Sign-reversible valley-dependent Berry phase effects in 2D valley-half-semiconductors

Xiaodong Zhou1,2,6, Run-Wu Zhang1,2,6, Zeying Zhang3,6, Wanxiang Feng1,2,6, Yuriy Mokrousov4,5 and Yugui Yao1,2

INTRODUCTION
Recent advances in valleytronics are mainly based on the paradigm of time-reversal-connected valleys, which generates valley polarization by an external field, dynamically or statically1,2. However, for achieving widespread applications of valleytronics, intrinsic properties are to be prioritized higher than external tunability. More importantly, intrinsic valleytronics materials hosting spontaneous valley polarization are most desirable, owing to their advantages of robustness, power efficiency, and simplicity in operation. In this regard, alternatives to the existing paradigms are intensively sought after. Recently, the proposal of two-dimensional (2D) ferrovalley materials has laid out a magneto-valleytronics composite paradigm based on spontaneous quantum phase transitions induced by the integrated effects of magnetic order and spin-orbit coupling (SOC), which can radically reduce additional costs of the applied external fields3. Specifically, when valley couples with intrinsic ferromagnetic order, the valley-dependent Berry phase effects can generate emergent valley anomalous transport phenomena – e.g., valley Hall effect5–11 and valley Nernst effect12–17 – making it possible to realize high-performance quantum devices and thus raising an intensive interest in materials systems which host magneto-valley traits.

From the perspective of potential applications in valleytronics, exploring various phase transitions in 2D magneto-valley materials has played a vital role in promoting our understanding, discovery, and characterization of emerging quantum states of matter. A general law of phase transitions is that they can drive not only rich quantum states but also intriguing physical properties. However, possibly due to the fewer number of 2D ferrovalley materials14–18 and the difficulty in characterizing the magnetic ordering and topological phase transitions, high-quality magneto-valley material candidates with a wide range of phase transitions have been scarcely discussed. On the other hand, an interplay between different phase transitions accompanied by distinct valley anomalous transport manifestations in such materials has not been seriously considered yet, which poses a great challenge for the research on potential high-performance valleytronics devices. A crucial but thought-provoking issue for magneto-valley coupling is finding a way to effectively generate spontaneous valley polarization utilizing the spin degree of freedom, and thereby lead to a revolution in magneto-valley-based information storage and operation principles. To tackle the current challenges of valleytronics, we break out the present ferrovalley paradigm to provide a general classification of magneto-valley coupling states through an effective model analysis, where the low energy electronic inter-valley nature enjoys a giant valley splitting with fully spin-polarized fermions. Consistent with the model, as a concrete example, V$_2$N$_4$ showcases emerging magneto-valley coupling phases – including valley-half-semiconductor, valley-half-semimetal as well as valley-unbalanced quantum anomalous Hall states (see Fig. 1) – transitions between which can be controlled by using external stimuli such as biaxial strain, electric field, and correlation effects. Remarkably, we find that topological phase transitions with magneto-valley coupling can exhibit valley-dependent Berry phase effects which manifest in prominent valley sign-reversible anomalous transport fingerprints.

RESULTS AND DISCUSSION
Valley-half-semiconductors and their topological phase transitions
According to the relationship between the valence band maximum and conduction band minimum of the same spin channel at the Fermi level, magneto-valley coupling (MVC) gives rise to a wide spectrum of quantum phase transitions ranging from gapped to gapless ones. To better capture the key physics underlying topological phase transitions, we construct a simple tight-binding (TB) model for describing the topological phase transitions of MVC states. Without loss of generality, we take a 2D triangular lattice with a magnetic space group $P6/m2'$ as an example, and assume that the direction of the spins of magnetic atoms is parallel to the $z$-axis.
(i.e., out of the plane). To conveniently describe the atomic basis of \(P6m'2'\) symmetry, the minimal set of \([d_{x^2-y^2}^\uparrow]\) and \([d_{x^2-y^2}^\downarrow]\) (or \([p_y^\uparrow]\) and \([p_y^\downarrow]\)) orbitals is taken as the basis in our TB model. The Hamiltonian containing only the nearest-neighbour hopping reads:

\[
H = \sum_{\langle ij \rangle} t_{ij}^{a\beta} c_{i\alpha}^\dagger c_{j\beta} + H.c.,
\]

where \(c_{i\alpha}^\dagger\) (\(c_{i\alpha}\)) is the electron creation (annihilation) operator for the orbital \(\alpha\) (\(\beta\)) on site \(i\) (\(j\)). By using the MagneticTB package\(^{19}\) that was recently developed to incorporate the group representation theory, all independent nearest-neighbour hopping integrals \(t_{ij}^{a\beta}\) can be screened under a given symmetry constraint. Under the magnetic space group of \(P6m'2'\), the topological phase transitions of MVC states can be achieved with only three parameters \(t_2, r_2\), and \(e_1\), see Eqs. (23) and (24)). The Chern number for characterizing topologically nontrivial phases can be analytically written as,

\[
|\mathcal{C}| = \frac{1}{2} \left( \text{Sign} \left( e_1 - \frac{3\sqrt{3}r_2}{2} \right) - \text{Sign} \left( e_1 + \frac{3\sqrt{3}r_2}{2} \right) \right)
\]

For our model, Fig. 1a schematically shows that K and K' valleys bare semiconductor characteristics with full spin-polarization in the same spin channel. Such a valley-half-semiconductor (VHSC) state is highly promising for generating, transporting, and manipulating spin currents in spin-valleytronics. Topological phase transitions starting from the VHSC state can be realized by changing the model parameters \(r_2\) and \(e_1\) simultaneously. When the bandgap at K valley decreases, the other one at K' valley is reduced as well. During the transition, a critical state, namely the valley-half-semimetal (VHSM) state, is inevitably encountered [Fig. 1b]; it is gapless at K' valley but it is gapped at K valley. The gapless crossing point is two-fold degenerate with linear dispersion, similar to that of Weyl semimetals. In Fig. 1c, the gap reopening at K' valley indicates a topological phase transition, and the valley-unbalanced quantum anomalous Hall state (VQAH) state is confirmed by a nonzero Chern number \(|\mathcal{C}| = 1\), which is contributed by both K and K' valleys. The VQAH state predicted here differs from the previously considered quantized valley Hall state or valley-polarized quantum anomalous Hall state. The quantized valley Hall state is found in spatial inversion broken non-magnetic materials, in which both K and K' valleys host nonzero Chern numbers but with opposite signs, while the valley-polarized quantum anomalous Hall state is predicted in magnetic materials with nonzero Chern number originating from only one valley\(^{5,20,21}\). Further increasing the parameter \(e_1\) will force K valley to first close the gap and then reopen it again, as shown in Fig. 1d, e. The summary of topological phase transitions is shown in Fig. 1f. Note that the SOC has been taken into account in our model, and therefore the emergence of MVC states having 100% spin polarization is remarkable and promising for spin-valleytronics\(^{12-15}\). An interplay between valley degree of freedom, magnetism, and topology provides an excellent platform for researching valley-related anomalous transport properties of MVC states, realizing a rich set of exotic quantum phenomena, such as valley anomalous Hall effect (VAHE), valley anomalous Nernst effect (VANE), as well as valley magneto-optical Kerr effect (VMOKE) and valley magneto-optical Faraday effect (VMOFE). Specifically, pursuing a single material that simultaneously exhibits topological phase transitions and valley sign-reversible Berry phase effects has been rarely done to date, although such a material would be highly valuable for multi-functional miniaturized devices. In addition to considering artificially constructed MVC heterostructures, an alternative is to seek intrinsic MVC materials that can harbor topological phase transitions driven by external means, such as biaxial strain, electric field, and correlation effects.

**High-quality candidate materials**

In this work not only do we provide a classification of MVC states, but also propose a series of MVC materials [Fig. 2a, b], including 2D ferromagnetic VSi\(_2\)N\(_4\) as well as other eleven M\(_A\)Z\(_N\) (\(M = V, Nb; A = Si, Ge; Z = N, P, As\)) candidates\(^{26,27}\). All these candidate materials form on a hexagonal lattice with the same magnetic space group that has been employed above, which can be regarded as the magnetic counterparts of the valley Hall semiconductors MoSi\(_2\)N\(_4\) family\(^{28,29}\). While aforementioned MVC states and their topological phase transitions exist in all materials,
undergoes topological phase transitions that can be identified by the closing and reopening of band gaps at K and K’ valleys, resulting in the VHSM (1.4%), VQAH (1.6%), VHSM (1.75%), and VHSC (2%) states. The gap evolution together with corresponding Chern numbers is summarized in Fig. 2e. Furthermore, the orbital-projected band structures of MVC states (Fig. 2g) are another indicator for topological phase transitions. In the balanced state of VSi$_2$N$_4$, the band minimum is dominated by the $d_{x^2-y^2}$ and $d_{z^2}$ orbitals of V atoms, and the conduction band minimum mainly comes from the $d_{z^2}$ orbital. At the strain of 1.4%, the orbital composition reverses at K’ valley, driving the system into the VQAH state; further increasing strain to 1.75%, the orbital inversion occurs also at K valley, restoring the system to the VHSC state. This valley-related topological phase transition can also be driven by the correlation effect, as realized in FeCl$_2$ and VSi$_2$P$_4$.53,54.

Sign reversal of valley-dependent Berry phase effects

An in-depth investigation of MVC topological phase transitions provides a platform for exploring valley-related anomalous transport phenomena. In this context, it is remarkable that VSi$_2$N$_4$, while hosting topological phase transitions, exhibits also sign change of valley-dependent Berry phase effects. Regarding the VHSC state (0%), for which the calculated k-resolved Berry curvature $\Omega(k)$ is shown in Fig. 3a, one can clearly identify the hot spots in the Berry curvature around two valleys with opposite signs and different magnitudes. By introducing a tiny biaxial strain, the VHSC state experiences a topological phase transition into the VQAH state, bridged by the VHSM state. Within the strain of 1.4—1.6%, the sign of $\Omega(k)$ at K’ valley flips [Fig. 3b]. Further increasing strain from 1.6—2%, K valley also experiences a topological phase transition, akin to the case of K’ valley, resulting in the sign change of $\Omega(k)$ at K valley [Fig. 3c]. Such dynamics of $\Omega(k)$ is bound to influence valley-related anomalous transport phenomena such as VAHE, VANE, VMOKE, and VMOFE.

Our predictions concerning the valley-related anomalous transport phenomena are presented in Fig. 3. Since VAHE is calculated by the integration of $\Omega(k)$ in a small region centered at each valley, the sign changes of VAHE are in full accordance with $\Omega(k)$ during the topological phase transitions. This is also the case for VANE as well as VMOKE and VMOFE. The former is calculated by integrating the Berry curvature together with a weighting...
factor around each valley (see Eq. (4)). The latter phenomenon can be actually regarded as the counterpart of VAHE. Physically, these anomalous transport phenomena are intimately related to each other. Therefore, the change in their signs strongly depends on the nature of topological phase transitions, exhibiting exotic sign reversal of valley-dependent Berry phase effects.

Interestingly, due to the different magnitudes of $\Omega(k)$ at two valleys, a net fully spin-polarized valley current is produced by the anomalous Hall, anomalous Nernst, and magneto-optical effects. Notably, the VQAH phase emerges during the phase transition, accompanied by a quantized anomalous Hall conductivity $\sigma_{xy} = e^2/h$ [second row of Fig. 3b]. Besides, one can utilize the non-contact magneto-optical technique to detect this topological phase transition. In the bottom of Fig. 3b, one can clearly observe the quantization behavior of $\theta_K$ and $\theta_F$ in the low-frequency limit.

Generally, the tunable sign of $\Omega(k)$ has been witnessed by reversing the magnetization and ferroelectric polarization. However, these two means seem to be sub-optimal and often suffer from various drawbacks, making it difficult to utilize this effect. As an alternative avenue, the manipulation by a small biaxial strain is more suitable for practical purposes. It is worth noting that topological phase transitions through biaxial strain may not only change the sign of anomalous transport characteristics but also modify their magnitude, and more importantly, the mediated quantization of transport characteristics is indispensable for experimental observation. We further calculate the effects of temperature and disorder on the valley-dependent Berry phase effects, and the results (see Supplementary Fig. 10) show that the sign changes of VAHE and VANE, being the key feature of...
topological phase transitions, are robust against disorder and temperature. In this work, we introduce a general framework to realize topological magneto-valley phase transitions in 2D VHSC, and we propose a series of feasible candidate materials harboring valley-dependent Berry phase effects, which are triggered by external means such as biaxial strain, electric field, and correlation effects. Taking VSi2N4 as a representative, we demonstrate that such intrinsic VHSC states display long-sought fully spin-polarized valley index. The proposal of sign reversal of valley-dependent Berry phase effects and high-quality materials realization greatly expand the ferrovalley family and provide an exciting playground for spintronics and valleytronics applications.

**METHODS**

**First-principles calculations**

The first-principles calculations were carried out employing the projected augmented wave method, as implemented in the Vienna ab initio simulation package (VASP). The exchange-correlation effect was treated by the Perdew-Burke-Ernzerhof parameterized generalized-gradient approximation (PBE-GGA). The energy cut-off of 500 eV and the k-mesh of 25 × 25 × 1 were used in the static calculations. The force and energy convergence criteria were set to be 10⁻³ eV/Å and 10⁻⁷ eV, respectively. The on-site Coulomb correlation of V and Nb atoms is considered within the GGA + U scheme. Different effective Hubbard energy U were tested in Supplementary Fig. S and the U = 3 eV was used in the main text, which has been also used in ref. The more accurate Heyd-Scuseria-Ernzerhof hybrid functional method (HSE06) was used to check the electronic structure. A vacuum thickness of 13 Å was used to avoid the interactions between the neighboring slabs. The phonon spectrum was performed based on the density functional perturbation theory (DFPT). The mostly localized Wannier functions including the d-orbitals of V atom, the s- and p-orbitals of Si atom, and the p-orbitals of N atom were constructed on a k-mesh of 8 × 8 × 1, using the WANNIER90 package.

**Anomalous Hall and anomalous Nernst effects**

The intrinsic anomalous Hall conductivity (AHC) and anomalous Nernst conductivity (ANC) were calculated on a dense k-mesh of 501 × 501 × 1, using the Berry phase theory,

\[
\sigma_{xy} = -\frac{e^2}{h} \int \sum_{\text{bands}} \Omega_{\sigma}(k) w_{\sigma}(k),
\]

where \(\Omega_{\sigma}(k)\) is the band- and momentum-resolved Berry curvature,

\[
\Omega_{\sigma}(k) = \sum_{\text{bands}} \frac{2\text{Im}\left\langle \phi_{\sigma} | \hat{v}_x | \phi_{\sigma} \right\rangle \left\langle \phi_{\sigma} | \hat{v}_y | \phi_{\sigma} \right\rangle}{(\omega_{\sigma} - \omega_{\sigma})^2}.
\]

Here, \((x, y)\) denote the Cartesian coordinates, \(\hat{v}_x, \hat{v}_y\) are the velocity operators, and \(\phi_{\sigma}(\textbf{k})\) is the eigenvector (eigenvalue) at band index \(n\) and momentum \(\textbf{k}\). The two unitless weighting factors \(w_{\uparrow}(\textbf{k})\) and \(w_{\downarrow}(\textbf{k})\) in Eqs. (3) and (4) are written as

\[
w_{\sigma}(k) = f_{\sigma}(k),
\]

\[
W_{\sigma}(k) = \frac{k_B T}{E_{\sigma} - \mu} f_{\sigma}(k) + k_B T \ln[1 + e^{-(E_{\sigma} - \mu)/k_B T}].
\]

**Magneto-optical Kerr and Faraday effects**

Extending the AHC to the ac case, the optical Hall conductivity \(\sigma_{xy}(\omega)\) is given by,

\[
\sigma_{xy}(\omega) = \frac{e\omega}{c} \sum_{\sigma} \sum_{\text{bands}} \frac{2\text{Im}\left\langle \phi_{\sigma} | \hat{v}_x | \phi_{\sigma} \right\rangle \left\langle \phi_{\sigma} | \hat{v}_y | \phi_{\sigma} \right\rangle}{(\omega_{\sigma} - \omega_{\sigma})^2} \frac{\omega_{\sigma}^2}{\omega_{\sigma}^2 - \omega^2 + i\Gamma}. \quad \text{(10)}
\]

where \(\omega_{\sigma}\) is the electronic frequency, \(\omega\) is the photon energy, \(\Gamma\) is the optical damping, and \(\text{Re}\) and \(\text{Im}\) denote the real and imaginary parts of the optical conductivity.

The MO Kerr and Faraday rotation angles in the Chern insulator are written as

\[
\theta_K = \text{Re} \left\{ \arg(E_x) - \arg(E_y) \right\}. \quad \text{(13)}
\]

\[
\theta_F = \text{Re} \left\{ \arg(E_x^\prime) - \arg(E_y^\prime) \right\}. \quad \text{(14)}
\]

The optical conductivities for a non-magnetic substrate (the SiO2 is used, and the optical conductivity is given by

\[
\sigma_{xy} = \frac{e^2}{c} \int \frac{d\omega}{2\pi} \sum_{\sigma} \text{Im}\left\langle \phi_{\sigma} | \hat{v}_x | \phi_{\sigma} \right\rangle \left\langle \phi_{\sigma} | \hat{v}_y | \phi_{\sigma} \right\rangle \frac{\omega^2}{\omega^2 - \omega^2 + i\Gamma}.
\]

where \(\omega_{\sigma}\) is the electronic frequency, \(\omega\) is the photon energy, \(\Gamma\) is the optical damping, and \(\text{Re}\) and \(\text{Im}\) denote the real and imaginary parts of the optical conductivity.
indicating the potential applications of 2D spintronics. The strained structure (with different bandgaps at \( K \) and \( K' \)) can be realized via the 'magnetic TB' model using a minimal basis set of magnetic orbitals and magnetic interaction. By using the MagneticTB package that is recently developed on the top of group representation theory, all independent nearest-neighbour hopping integrals \( t_{ij} \) can be screened under a given symmetry constrain. Under magnetic space group of \( PMn2' \), we found there are only three parameters are necessary to realize the magneto-valley coupling states and their topological phase transitions. The Hamiltonian is recast to

\[
H(k) = \left( \varepsilon_i + t_1 (\sin k_x - \sin k_y) \right) \sigma_z + t_2 (\cos k_x - \cos k_y) \sigma_x - t_3 (\cos k_x \cos k_y + \sin k_x \sin k_y) \sigma_y + H.c.,
\]

where \( \sigma_i \) (\( \sigma_z \)) is the electron creation (annihilation) operator for the orbital \( \alpha \) (\( \beta \)) on-site (\( i \)). Note that this Hamiltonian is in general including spin-orbit coupling and magnetic interaction. The Chern number for characterizing topological phases can be written by

\[
\mathcal{C} = \frac{1}{2} \left( \text{sign} \left( \varepsilon_i - \frac{3\sqrt{3}r_3}{2} \right) - \text{sign} \left( \varepsilon_i + \frac{3\sqrt{3}r_3}{2} \right) \right).
\]

## Curie temperature

Regarding the ferromagnetic VSi\(_2\)N\(_4\), a key physical quantity is Curie temperature (\( T_C \)). Here, \( T_C \) is the nearest-neighboring exchange interaction, \( S \) is the spin magnetic moment on the V or Nb atom. The calculated \( T_C \) for strain-free (\( \varepsilon = 0\% \)) VSi\(_2\)N\(_4\) is about 100 K (see Supplementary Fig. 3b), which is more than two times larger than those of \( \text{Cr}_2\) or \( \text{CrGeTe}_{2} \) of less than 30 K\(^{36}\), indicating the potential applications of 2D spintronics. The \( T_C \) (75 K) of strained-structure (\( \varepsilon = 1.6\% \)) is slightly smaller than that of the balanced system but is much larger than that of the famous antiferromagnetic topological insulator \( \text{MnBi}_2\text{Te}_4 \) (24 K\(^{36}\)), suggesting the great application prospects to realize the high-temperature VQAHE state.

## Tight-binding model

The magnetic space group of monolayer VSi\(_2\)N\(_4\) with out-of-plane magnetization is \( \text{PMn2'2'2} \). Considering the magnetic V atoms, it is a 2D triangular lattice with out-of-plane ferromagnetic order. We now introduce a two-band tight-binding model using a minimal basis set of \( \{ \sigma_{-}, \sigma_{+} \} \) and \( \{ \gamma_{-}, \gamma_{+} \} \) orbitals, where \( \sigma \) means spin up. Then, the generators of the magnetic space group \( \text{PMn2'2'2} \) are represented by \( \gamma_{0} = e^{i\theta_{0}} \), \( \gamma_{3} = \gamma_{0}^{3} \), \( \gamma_{6} = \gamma_{0}^{6} \), and \( \gamma_{7} = \gamma_{0}^{7} \). The magnetic space group of monolayer VSi\(_2\)N\(_4\) with out-of-plane magnetization is \( \text{PMn2'2'2} \). Here, \( J_{ij} \) is the nearest-neighboring exchange interaction, \( S \) is the spin magnetic moment on the V or Nb atom. The calculated \( T_C \) for strain-free (\( \varepsilon = 0\% \)) VSi\(_2\)N\(_4\) is about 100 K (see Supplementary Fig. 3b), which is more than two times larger than those of \( \text{Cr}_2\) or \( \text{CrGeTe}_{2} \) of less than 30 K\(^{36}\), indicating the potential applications of 2D spintronics. The \( T_C \) (75 K) of strained-structure (\( \varepsilon = 1.6\% \)) is slightly smaller than that of the balanced system but is much larger than that of the famous antiferromagnetic topological insulator \( \text{MnBi}_2\text{Te}_4 \) (24 K\(^{36}\)), suggesting the great application prospects to realize the high-temperature VQAHE state.

The magnetic space group of monolayer VSi\(_2\)N\(_4\) with out-of-plane magnetization is \( \text{PMn2'2'2} \). Considering the magnetic V atoms, it is a 2D triangular lattice with out-of-plane ferromagnetic order. We now introduce a two-band tight-binding model using a minimal basis set of \( \{ \sigma_{-}, \sigma_{+} \} \) and \( \{ \gamma_{-}, \gamma_{+} \} \) orbitals, where \( \sigma \) means spin up. Then, the generators of the magnetic space group \( \text{PMn2'2'2} \) are represented by \( \gamma_{0} = e^{i\theta_{0}} \), \( \gamma_{3} = \gamma_{0}^{3} \), \( \gamma_{6} = \gamma_{0}^{6} \), and \( \gamma_{7} = \gamma_{0}^{7} \). The magnetic space group of monolayer VSi\(_2\)N\(_4\) with out-of-plane magnetization is \( \text{PMn2'2'2} \). Here, \( J_{ij} \) is the nearest-neighboring exchange interaction, \( S \) is the spin magnetic moment on the V or Nb atom. The calculated \( T_C \) for strain-free (\( \varepsilon = 0\% \)) VSi\(_2\)N\(_4\) is about 100 K (see Supplementary Fig. 3b), which is more than two times larger than those of \( \text{Cr}_2\) or \( \text{CrGeTe}_{2} \) of less than 30 K\(^{36}\), indicating the potential applications of 2D spintronics. The \( T_C \) (75 K) of strained-structure (\( \varepsilon = 1.6\% \)) is slightly smaller than that of the balanced system but is much larger than that of the famous antiferromagnetic topological insulator \( \text{MnBi}_2\text{Te}_4 \) (24 K\(^{36}\)), suggesting the great application prospects to realize the high-temperature VQAHE state.
28. Li, S. et al. Valley-dependent properties of monolayer MoS2, WS2, and MoS2 and MoS2 and MoS2As monolayers. Phys. Rev. B 102, 235435 (2020).
29. Yang, C., Song, Z., Sun, X. & Lu, J. Valley pseudospin in monolayer MoS2 and MoS2 and MoS2As. Phys. Rev. B 103, 035308 (2021).
30. Bhalla, P., MacDonald, A. H. & Culcer, D. Resonant photovoltic effect in doped magnetic semiconductors. Phys. Rev. Lett. 124, 087402 (2020).
31. Kim, S. et al. Direct measurement of the Fermi energy in graphene using a double-layer heterostructure. Phys. Rev. Lett. 108, 116404 (2012).
32. Cui, Q., Zhu, Y., Li, P. & Yang, H. Spin-valley coupling in a two-dimensional 3D topological insulator. Phys. Rev. B 103, 085421 (2021).
33. Liu, S., Wang, Q., Zhang, C., Guo, P. & Yang, S. A. Correlation-driven topological and valley states in monolayer VSi2N4. Phys. Rev. B 104, 085149 (2021).
34. Feng, X. et al. Valley-related multiple Hall effect in monolayer VSi2N4. Phys. Rev. B 104, 075421 (2021).
35. Antonov, V., Harmon, B. & Yaresko, A. Electronic structure and magneto-optical properties of solids (Chap. 1.4). (Kluwer Academic Publishers, Dordrecht, 2004).
36. Huang, B. et al. Layer-dependent ferromagnetism in a van der waals crystal down to the monolayer Limit. Nature 546, 270 (2017).
37. Gong, C. et al. Discovery of intrinsic ferromagnetism in two-dimensional van der waals crystals. Nature 546, 265 (2017).
38. Wu, L. et al. Quantized Faraday and Kerr rotation and axion electrodynamics of a 3D topological insulator. Science 354, 1124 (2016).
39. Okada, K. N. et al. Terahertz spectroscopy on Faraday and Kerr rotations in a quantum anomalous Hall state. Nat. Commun. 7, 12245 (2016).
40. Shuavaev, A., Dziorn, V., Kvon, Z. D., Mikhailov, N. N. & Pimenov, A. Universal Faraday rotation in HgTe wells with critical thickness. Phys. Rev. Lett. 117, 117401 (2016).
41. Dziorn, V. et al. Observation of the universal magneto-electric effect in a 3D topological insulator. Nat. Commun. 8, 15197 (2017).
42. Qi, X.-L., Hughes, T. L. & Zhang, S.-C. Topological field theory of time-reversal invariant insulators. Phys. Rev. B 78, 195424 (2008).
43. Maciejko, J., Qi, X.-L., Drew, H. D. & Zhang, S.-C. Topological quantization in units of the fine structure constant. Phys. Rev. Lett. 105, 166803 (2010).
44. Tse, W.-K. & MacDonald, A. H. Giant magneto-optical Kerr effect and universal Faraday effect in thin-film topological insulators. Phys. Rev. Lett. 105, 057401 (2010).
45. Liu, X., Pyatakov, A. P. & Ren, W. Magneto-electric coupling in multiferroic bilayer V20. Phys. Rev. Lett. 125, 247601 (2020).
46. Bloch, P. E. Projector augmented-wave method. Phys. Rev. B 50, 17953 (1994).
47. Kresse, G. & Furthmüller, J. Efficiency of ab-initio total energy calculations for metals and semiconductors using a plane-wave basis set. Comput. Mater. Sci. 6, 15 (1996).
48. Perdew, J. P., Burke, K. & Ernzerhof, M. Generalized gradient approximation made simple. Phys. Rev. Lett. 77, 3865 (1996).
49. Dudarev, S. L., Botton, G. A., Savrasov, S. Y., Humphreys, C. J. & Sutton, A. P. Electron-energy-loss spectra and the structural stability of nickel oxide: an LSDA+U study. Phys. Rev. B 57, 1505 (1998).
50. Heyd, J., Scuseria, G. E. & Ernzerhof, M. Hybrid functionals based on a screened coulomb potential. J. Chem. Phys. 118, 8207 (2003).
51. Baroni, S., Gironcoli, S. D., Corso, A. D. & Giannozzi, P. Phonons and related crystal properties from density-functional perturbation theory. Rev. Mod. Phys. 73, 515 (2001).
52. Mostofi, A. A. et al. Wannier90: a tool for obtaining maximally-localised Wannier functions. Comput. Phys. Commun. 178, 685 (2008).
53. Yao, Y. et al. First principles calculation of anomalous Hall conductivity in ferromagnetic bcc Fe. Phys. Rev. Lett. 92, 037204 (2004).
54. Xiao, D., Yao, Y., Fang, Z. & Liu, B. Q. O. Q17 phase effect in anomalous thermoelectric transport. Phys. Rev. Lett. 97, 026603 (2006).
55. Czaja, P., Freimuth, F., Weischenberg, J., Blügel, S. & Mokrousov, Y. Anomalous Hall effect in ferromagnets with Gaussian disorder. Phys. Rev. B 89, 014411 (2014).
56. Spencer, C. S. et al. Helical magnetic structure and the anomalous and topological Hall effects in epitaxial 2B2Fe3C6Ge films. Phys. Rev. B 97, 214406 (2018).
57. Keiser, A. C., Raimondi, R. & Culcer, D. Sign change in the anomalous Hall effect and strong transport effects in a 2D massive Dirac metal due to spin-charge correlated disorder. Phys. Rev. Lett. 123, 126603 (2019).
58. Suzuki, Y., Katayama, T., Yoshida, S., Tanaka, K. & Sato, K. New magneto-optical transition in ultrathin Fe(100) films. Phys. Rev. Lett. 68, 3355 (1992).
59. Guo, G. Y. & Ebert, H. Band-theoretical investigation of the magneto-optical Kerr effect in Fe and Co multilayers. Phys. Rev. B 51, 12633 (1995).
60. Ravindran, P. et al. Magnetic, optical, and magneto-optical properties of MnX (X=As, Sb, or Bi) from full-potential calculations. Phys. Rev. B 59, 15680 (1999).