Electric-field-tuned anomalous valley Hall effect in A-type hexagonal antiferromagnetic monolayers

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The combination of antiferromagnetic (AFM) spintronics and anomalous valley Hall effect (AVHE) is of great significance for potential applications in valleytronics. Here, we propose a way for achieving AVHE in *A*-type hexagonal AFM monolayer. The proposed way involves the introduction of layer-dependent electrostatic potential caused by an out-of-plane external electric field, which can break the combined symmetry (*PT* symmetry) of spatial inversion (*P*) and time reversal (*T*), producing spin splitting. The spin order of spin splitting can be reversed by regulating the direction of electric field. Based on first-principles calculations, the way can be verified in AFM Cr_2CH_2 . The layer-locked hidden Berry curvature can give rise to layer-Hall effect, including a valley layer–spin Hall effect and layer-locked AVHE. Moreover, we propose Janus monolayer Cr_2CHF with internal electric polarization, which can also realize the AVHE. Our works provide an experimentally feasible way to realize AVHE in AFM monolayer.

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I. INTRODUCTION

Since the discovery and successful preparation of rich two-dimensional (2D) materials, valley has recently received extensive attention, which lays the foundation for processing information and performing logic operations with low power consumption and high speed [1-8]. Transition-metal dichalcogenide (TMD) monolayers are typical valleytronic materials with a pair of degenerate but inequivalent -K and K valleys in the reciprocal space [9–14]. The -K and K valleys exhibit opposite Berry curvature and selective absorption of chiral light. When including spin-orbit coupling (SOC), the valleys of -K and K points produce opposite spin splitting characterized by spin-valley locking. However, these nonmagnetic TMD monolayers lack spontaneous valley polarization, which hinders the wide application of valleytronic devices. To achieve valley splitting, many methods have been applied, such as external magnetic field [14,15], proximity effect [13,16,17], and light excitation [18,19].

To realize intrinsic valley polarization, the ferrovalley semiconductor (FVS) has been proposed [20], which appears in ferromagnetic (FM) materials with broken spatial inversion symmetry. Many 2D materials have been predicted to be FVS, such as 2H-VSe₂ [20], $CrSi_2X_4$ (X = N and P) [21], $VAgP_2Se_6$ [22], $LaBr_2$ [23,24], VSi_2P_4 [25], NbX_2 (X = S and Se) [26], Nb₃I₈ [27], TiVI₆ [28], and FeClBr [29]. Realizing valley polarization in antiferromagnetic (AFM) materials is more meaningful for valleytronic application, because AFM materials possess the high storage density, robustness against external magnetic field, as well as ultrafast writing speed [30]. Thus, it is of fundamental importance and high interest to achieve valley polarization in AFM materials, accompanied by the anomalous valley Hall effect (AVHE). For AVHE the presence of additional charge Hall current originates from the spontaneous valley polarization, which is similar to the anomalous Hall effect in ferromagnetic materials.

However, both spontaneous valley polarization and AVHE are rarely reported in AFM monolayer. By stacking AFM monolayer MnPSe₃ or Cr₂CH₂ on ferroelectric monolayer Sc₂CO₂, the AVHE has been achieved due to the introduction of nonuniform potentials to break the simultaneous time reversal and spatial inversion [31,32]. Spin splitting and spontaneous valley polarization, accompanied by AVHE, have also been predicted in AFM Janus $Mn_2P_2X_3Y_3$ (X, Y = S, Se Te; $X \neq Y$) monolayers by introducing an out-of-plane potential gradient [33]. Here, we propose a way for achieving AVHE in A-type hexagonal AFM monolayer by the introduction of layer-dependent electrostatic potential caused by out-of-plane external electric field. The spin order of spin splitting can be reversed when the direction of the electric field is flipped. By first-principles calculations, we translate the proposal into Cr₂CH₂ monolayer and clarify the electric-field-tuned valley physics. Moreover, Janus monolayer A-type AFM Cr₂CHF with intrinsic polar electric field has also been used to realize the AVHE.

II. WAYS TO ACHIEVE AVHE

The proposal for electric-field-tuned AVHE in AFM monolayer is schematically illustrated in Fig. 1. The proposed way can be considered as an application of the design principle in Ref. [32]. We consider a hexagonal centrosymmetric monolayer with two-layer magnetic atoms with intralayer FM and interlayer AFM orderings (A-type AFM ordering), which possesses energy extrema of conduction or valance bands located at high-symmetry -K and K points. The lattice of our proposed system has inversion symmetry, but the opposite spin vectors of the two sublattices break spatial inversion (*P*) symmetry and time-reversal (*T*) symmetry, which produces spontaneous valley polarization [Fig. 1(a)]. However, the spin degeneracy of -K and K valleys is maintained



FIG. 1. (a) A hexagonal *A*-type AFM monolayer with spin degeneracy but nonequivalent –K and K valleys; (b) by applying an out-of-plane electric field, the spin degeneracy is removed, and the –K and K valleys still are unequal; and (c) when the direction of electric field is reversed, the order of spin splitting is also reversed. The spin-up and spin-down channels are depicted in blue and red. The green arrow represents the electric field.

due to *PT* symmetry, which prohibits the AVHE in AFM monolayer.

There is zero Berry curvature $[\Omega(k)]$ everywhere in the momentum space due to *PT* symmetry. However, layer-locked hidden Berry curvature can appear, because each layer breaks the *PT* symmetry. Due to layer-spin locking, the Berry curvatures for the spin-up and spin-down channels are equal in magnitude and opposite in sign. The layer-locked hidden Berry curvature can give rise to layer-Hall effect.

Here, an out-of-plane electric field is applied to break the *PT* symmetry, which can remove spin degeneracy of -K and K valleys. The out-of-plane electric field can produce layer-dependent electrostatic potential $\propto eEd$ (*e* and *d* denote the electron charge and the layer distance), which induces the spin-splitting effect. With breaking *PT* symmetry, the spin splitting at the -K and K valleys can be realized [Fig. 1(b)], resulting in the AVHE. Moreover, it is expected that the spin order at both the -K and K valleys can be reversed through manipulating the direction of the out-of-plane electric field [Fig. 1(c)].

To achieve AVHE in the AFM monolayers, spin splitting is a necessary factor. Our recent work proposes a way to produce spin splitting in AFM materials by making the magnetic atoms with opposite spin polarization locate in the different environment (surrounding atomic arrangement) [34]. In Ref. [31], the spin splitting is mainly achieved by a layerdependent electrostatic potential caused by the introduction of substrate. In Ref. [32], the spin splitting is achieved by making the magnetic atoms with opposite spin polarization locate in a different environment caused by the substrate. In fact, the approach to achieve spin splitting in Ref. [31] also fundamentally aligns with the proposition of our initial research [34]. The spin splitting in our present work is realized by a layer-dependent electrostatic potential caused by an outof-plane electric field, so *A*-type AFM order and an external out-of-plane electric field are necessary. A hexagonal lattice is needed to produce spontaneous valley polarization and layer-locked hidden Berry curvature combined with *A*-type AFM order.

The H-functionalized MXene Cr_2C (Cr_2CH_2) [35] can be used as a real material to verify our proposal. The Cr_2C is a half-metallic ferromagnet which has been successfully synthesized in experiments [36]. By surface functionalization with H in Cr_2C , a ferromagnetic-antiferromagnetic transition and a metal-insulator transition can be induced simultaneously [35]. In addition to this, Cr_2CH_2 possesses A-type AFM ordering with energy extrema of valance bands located at -Kand K points, which meets the requirements of our proposal.

III. COMPUTATIONAL DETAIL

Within density-functional theory (DFT) [37], the spinpolarized first-principles calculations are carried out within the projector augmented-wave (PAW) method, as implemented in the Vienna Ab initio Simulation Package (VASP) [38–40]. The generalized gradient approximation of Perdew-Burke-Ernzerhof (PBE-GGA) [41] as the exchangecorrelation functional is adopted. The kinetic energy cutoff of 500 eV, total energy convergence criterion of 10^{-8} eV, and force convergence criterion of 0.0001 eV $Å^{-1}$ are set to obtain accurate results. To account for the localized nature of Cr-3*d* orbitals, a Hubbard correction $U_{\text{eff}} = 3.0 \text{ eV} [31,42]$ is used by the rotationally invariant approach proposed by Dudarev et al. [43]. The SOC is incorporated for investigation of valley splitting and magnetic anisotropy energy (MAE). The vacuum space of more than 20 Å along z direction is introduced to avoid interactions between neighboring slabs. $21 \times 21 \times 1$ Monkhorst-Pack k-point meshes are used to sample the Brillouin zone (BZ) for calculating electronic structures. Based on the finite displacement method, the interatomic force constants (IFCs) are calculated by employing a $5 \times 5 \times 1$ supercell with AFM ordering, and the phonon dispersion spectrum is obtained by the PHONOPY code [44]. The Berry curvatures are calculated directly from the calculated wave functions based on the method of Fukui et al. [45], as implemented in the VASPBERRY code [46-48]. The elastic stiffness tensor C_{ij} and piezoelectric stress tensor e_{ij} are calculated by using a strain-stress relationship (SSR) method and density functional perturbation theory (DFPT) method [49], respectively. The C_{ij}^{2D}/e_{ij}^{2D} has been renormalized by $C_{ij}^{2D} = L_z C_{ij}^{3D} / e_{ij}^{2D} = L_z e_{ij}^{3D}$, where L_z is the length of the unit cell along z direction.

IV. CRYSTAL AND ELECTRONIC STRUCTURES

It has been proved that Cr_2CH_2 monolayer possesses Atype AFM ordering with dynamical, mechanical, and thermal stabilities [35]. The crystal structures of Cr_2CH_2 along with the first BZ are shown in Figs. 2(a)-2(c), which crystallizes in the $P\bar{3}m1$ space group (No. 164), hosting inversion symmetry. It consists of five atomic layers in the sequence of H-Cr-C-Cr-H, and the magnetic Cr atoms distribute in two layers. The optimized equilibrium lattice constants are a = b = 2.99 Å by the GGA+U method, which agrees well with previous results (2.95 Å [35]). Based on its energy-band



FIG. 2. For monolayer Cr_2CH_2 with $a/a_0 = 0.96$, (a), (b) the top and side views of crystal structures, (c) the first BZ with high-symmetry points, and (d) the phonon dispersion spectrum.

structures in Fig. S1 [50] or in Ref. [35], the valence-band maximum (VBM) of Cr_2CH_2 is at the Γ point, not the –K or K point. To clearly clarify our proposal, the biaxial strain ($a/a_0 = 0.96$) is applied to change the VBM of Cr_2CH_2 from the Γ to the –K or K point. To determine the magnetic ground state of Cr_2CH_2 , the FM and three AFM configurations (AFM1, AFM2, and AFM3) are constructed, as shown in Fig. 3, and the AFM1 is called an *A*-type AFM state. Calculated results show that the energy of the AFM1 per unit cell is 281 meV, 50 meV, and 399 meV lower than those of FM, AFM2, and AFM3 cases by GGA+U, confirming that the strained Cr_2CH_2 still possesses the AFM1 ground state. The calculated phonon spectrum of strained Cr_2CH_2 with no obvious imaginary frequencies is shown in Fig. 2(d), indicating its dynamic stability.

The energy-band structures of Cr_2CH_2 by using both GGA and GGA+SOC are plotted in Fig. 4. According to Fig. 4(a),



FIG. 3. Four magnetic configurations with side view of crystal structure: FM (a), AFM1 (b), AFM2 (c), and AFM3 (d) configurations. The blue (red) balls represent the spin-up (spin-down) Cr atoms.

134402-3



FIG. 4. For Cr₂CH₂ with $a/a_0 = 0.96$, the energy-band structures without SOC (a), and with SOC (b), (c), (d) for magnetization direction along the positive *z*, negative *z*, and positive *x* direction, respectively. In (a), the blue (red) lines represent the band structure in the spin-up (spin-down) direction.

no spin splitting can be observed due to the PT symmetry, and Cr₂CH₂ is an indirect-band-gap semiconductor. The energies of -K and K valleys in the valence band are degenerate. Figure 4(b) shows that the valley polarization can be induced by SOC, and the valley splitting is 49 meV. The energy of the K valley is higher than that of the –K valley, and the valley polarization can be switched when the magnetization direction is reversed [Fig. 4(c)]. When the magnetization direction of Cr_2CH_2 is in-plane along the x direction [Fig. 4(d)], no valley polarization can be observed. Therefore, the magnetic orientation is determined by MAE, which can be obtained by $E_{\text{MAE}} = E_{\text{SOC}}^{||} - E_{\text{SOC}}^{\perp}$, where || and \perp mean that spins lie in- and out-of-plane. The calculated MAE is 27 μ eV/unit cell, indicating the out-of-plane easy magnetization axis of Cr₂CH₂. The out-of-plane easy magnetization axis means that Cr₂CH₂ has spontaneous valley polarization, which confirms the realization of our proposal. The total magnetic moment of Cr_2CH_2 per unit cell is strictly 0.00 μ_B , with the magnetic moment of the bottom/top Cr atom being 3.09 μ_B /-3.09 μ_B . When the SOC is included, the spin degeneracy is still maintained for both the out-of-plane and in-plane magnetization directions.

For Cr₂CH₂, the distribution of Berry curvatures of total, spin-up, and spin-down are plotted in Fig. 5. It is observed that total Berry curvature everywhere in the momentum space is zero due to *PT* symmetry. The extremes of spin-resolved Berry curvatures locate at the –K and K valleys, which have opposite signs for the same spin channel. For the same valley at different spin channels, the Berry curvatures are also opposite. When a longitudinal in-plane electric field is applied, the Bloch carriers will acquire an anomalous transverse velocity $v_{\perp} \sim E_{\parallel} \times \Omega(k)$ [4]. By shifting the Fermi level between the



FIG. 5. For Cr_2CH_2 with $a/a_0 = 0.96$, the distribution of Berry curvatures of total (a), spin-up (b), and spin-down (c). In the presence of a longitudinal in-plane electric field, an appropriate hole doping for three cases in Fig. 1 produces valley layer–spin Hall effect (d) and layer-locked anomalous valley Hall effect (e and f). The upper and lower planes represent the top and bottom Cr layers.

-K and K valleys in the valence band, the spin-up and spindown holes from the K valley will accumulate along opposite sides of different layers under a longitudinal in-plane electric field, resulting in the valley layer–spin Hall effect [Fig. 5(d)], but the AVHE is absent.

V. ELECTRIC FIELD INDUCES SPIN SPLITTING

To induce spin splitting in Cr_2CH_2 , *PT* symmetry should be broken. Here, an out-of-plane electric field +*E* (0.00– 0.10 V/Å) is applied that can break *PT* symmetry and lift spin degeneracy of valleys. The -*E* generates exactly the same results except spin orders, since two Cr layers are related by a glide mirror G_z symmetry but have opposite magnetic moments. Firstly, the magnetic ground state under the out-of-plane electric field is determined by the energy difference between FM/AFM2/AFM3 and AFM1 configurations. Within the considered *E* range, based on Fig. S2 [50], the AFM1 ordering is always ground state. The MAE vs *E* is plotted in Fig. S3 [50], and the positive MAE confirms that the easy axis of Cr₂CH₂ is in the out-of-plane direction within the considered *E* range. These ensure that our proposed way can be realized in Cr₂CH₂.

The energy-band structures of Cr_2CH_2 at representative *E* by using GGA+SOC are plotted in Fig. S4 [50], and the enlarged figures of spin-resolved energy-band structures near the Fermi level for the valence band are shown in Fig. 6. For the valence band, the valley splitting and spin splitting at –K and K valleys as a function of *E* are plotted in Fig. 7. The spontaneous valley polarization of about 49 meV is maintained within the considered *E* range. When an electric field is applied, it is clearly seen that there is spin splitting, which is due to the layer-dependent electrostatic potential caused by an out-of-plane electric field. Spin splitting and electric field strength show a linear relationship. In fact, the spin splitting can be approximately calculated by *eEd*. Taking *E* = 0.02 V/Å as a example, the *d* of Cr₂CH₂ is 2.37 A, and the



FIG. 6. For Cr₂CH₂ with $a/a_0 = 0.96$, the spin-resolved energyband structures near the Fermi level for the valence band with SOC at representative *E* (0.00, 0.02, 0.04, and 0.06 V/Å). The blue (red) lines represent the band structure in the spin-up (spin-down) direction.

estimated spin splitting is approximately 47 meV, which is close to the first-principles result, 41 meV. The coexistence of spin splitting and valley polarization is essential for the realization of AVHE. When the electric field changes from +E to -E, the layer-dependent electrostatic potential is also reversed. The sizes of spin splitting and valley splitting remain unchanged, but the spin order of spin splitting reversed (Fig. S5 [50]), which is consistent with our proposal.

For Cr_2CH_2 at E = 0.02 V/Å, the distribution of Berry curvatures of spin-up and spin-down are shown in Fig. S6 [50]. It is clearly seen that the Berry curvatures are opposite for the same valley at different spin channels and different valleys at the same spin channel. By shifting the Fermi level between the -K and K valleys in the valence



FIG. 7. For Cr₂CH₂ with $a/a_0 = 0.96$, the valley splitting (V_v) and spin splitting $(S_{-K}$ and S_K at -K and K valleys) for the valence band as a function of *E*.



FIG. 8. For Janus Cr₂CHF with $a/a_0 = 0.96$: the crystal structures (a), the phonon dispersion spectrum (b), the spin-resolved energy-band structures with SOC for magnetization direction along the positive *z* direction (c), and the distribution of Berry curvatures of spin-up (d). In (c), the spin-up and spin-down are depicted in blue and red.

band, only the spin-up holes from the K valley move to the bottom boundary of the sample under an in-plane electric field [Fig. 5(e)], producing layer-locked AVHE. Conversely, by reversing the electric field direction, the spin-down holes from the K valley will move to the top opposite side of the sample by proper hole doping [Fig. 5(f)]. This accumulation of spin-polarized holes produces a net charge/spin current, and Figs. 5(e) and 5(f) generate opposite voltage. Given the proximity and interconnectedness of these layers within such a thin structure, the manifestation of a layer-Hall effect poses significant challenges. However, the layer-locked hidden spin polarization (HSP) leads to nonreciprocal nonlinear conductivity, which has been exemplified by tetragonal A-type AFM CuMnAs via first-principles calculations [51]. By combining spin- and angle-resolved photoemission spectroscopy, the spin-momentum-layer locking phenomena has been reported in a centrosymmetric layered material BiOI [52].

VI. JANUS MONOLAYER CR2CHF WITH INTRINSIC POLAR ELECTRIC FIELD

An intrinsic polar electric field is equivalent to an external electric field, which can be induced by constructing a Janus structure. As illustrated in Fig. 8(a), the Janus monolayer is obtained by replacing the top H layer by F element, which leads to monolayer Cr₂CHF with the symmetry of the crystal to P3m1 (no. 156). The biaxial strain ($a/a_0 = 0.96$) is applied to change the VBM of Janus Cr₂CHF from Γ to -K or K point, and the corresponding *a* is 2.88 Å. Calculated results show that the energy of AFM1 per unit cell is 871 meV, 183 meV, and 887 meV lower than those of the FM, AFM2, and AFM3 cases by GGA+U, confirming that the strained Cr₂CHF has AFM1 ground state. The phonon spectrum of

strained Cr₂CHF is shown in Fig. 2(b), which shows no obvious imaginary frequencies, indicating its dynamic stability. The built-in electric field of strained Cr₂CHF is predicted to be 1.01 V/Å by the plane-averaged electrostatic potential along the *z* direction.

The energy-band structures of Cr₂CHF by using both GGA and GGA+SOC are plotted in Fig. S7 [50] and Fig. 8(c). According to Fig. S7 [50], the obvious spin splitting can be observed due to the broken PT symmetry, and Cr₂CHF is an indirect-band-gap semiconductor without valley polarization. Figure 8(c) shows a valley splitting of 50 meV when the SOC is included. The total magnetic moment of Cr₂CHF per unit cell is strictly 0.00 μ_B , with the magnetic moment of the bottom/top Cr atom being 3.09 μ_B /-3.06 μ_B . For Cr₂CHF, the distribution of Berry curvatures of spin-up is plotted in Fig. 8(d). It is observed that the extremes of Berry curvatures are located at the -K and K valleys, which have opposite signs. By shifting the Fermi level between the -K and K valleys in the valence band, the spin-up holes from the K valley will accumulate along the bottom boundary of the sample under a longitudinal in-plane electric field, resulting in the layer AVHE [Fig. 5(e)].

Compared with Cr_2CH_2 , the Cr_2CHF monolayer possesses piezoelectricity due to broken inversion symmetry, which has both in-plane and out-of-plane piezoelectricity. By using Voigt notation, when only considering the in-plane strain and stress, two independent piezoelectric strain coefficients d_{11} and d_{31} can be derived [53,54]:

$$d_{11} = \frac{e_{11}}{C_{11} - C_{12}}$$
 and $d_{31} = \frac{e_{31}}{C_{11} + C_{12}}$. (1)

The calculated elastic constant C_{11}/C_{12} of Cr₂CHF is 198.6 Nm⁻¹/58.8 Nm⁻¹, and the piezoelectric stress coefficient e_{11}/e_{31} is $-0.961 \times 10^{-10}/0.882 \times 10^{-10}$ C/m. Based on Eq. (1), the calculated d_{11} and d_{31} of Cr₂CHF are -0.69and 0.34 pm/V, respectively. Electric-field-induced spin splitting in Cr₂CH₂ has been confirmed. When a biaxial in-plane strain is imposed, only out-of-plane d_{31} appears in Cr₂CHF, which produces an out-of-plane electric field. Therefore, the spin splitting in Cr₂CHF can be tuned by piezoelectric effect, which has been used to tune Rashba spin splitting in a ZnO/P3HT nanowire array structure in experimentation [55].

VII. CONCLUSION

In summary, we present a model to induce AVHE in Atype hexagonal AFM monolayer by applying an electric field. The validity of our proposal is confirmed by an extensive study of Cr_2CH_2 within the first-principles calculations. The spontaneous valley polarization can occur in Cr_2CH_2 , but the spin splitting of -K and K valleys is absent. The introduction of an out-of-plane electric field results in spin splitting due to layer-dependent electrostatic potential. The layer-locked hidden Berry curvature produces layer-locked AVHE. Our work enriches the AVHE in AFM monolayers and provides great potential for developing energy-efficient and ultrafast valleytronic devices.

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