Supporting Information for

Layer-Polarized Anomalous Hall Effect in Valleytronic van der Waals Bilayers

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Table 1. Interlayer magnetic couplings, lattice constants (*a*), interlayer distances (*d*), ferroelectric polarizations, ferroelectric barriers and magnetocrystalline anisotropy energies (MAE) of bilayer VSi_2P_4, VSi_2N_4, $FeCl_2$, $RuBr_2$ and VClBr.

Bilayer	**Interlayer Coupling**	***a* (Å)**	***d* (Å)**	**Ferroelectric Polarization (pC/m)**	**Ferroelectric Barrier (meV/f.u.)**	**MAE (meV/f.u.)**
VSi_2P_4	AFM	3.46	3.17	0.42	8.6	8.84×10^{-2}
VSi_2N_4	AFM	2.87	3.01	1.93	5.2	-2.23×10^{-2}
$FeCl_2$	AFM	3.36	3.12	0.05	4.1	-1.90
$RuBr_2$	AFM	3.68	3.08	0.21	7.9	-0.75
VClBr	AFM	3.30	3.12	0.13	6.4	-0.25

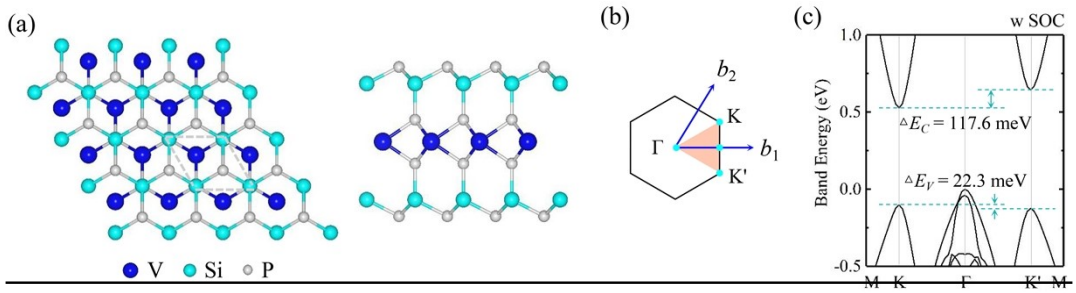

Fig. S1 (a) Crystal structures of single-layer VSi_2P_4 from and side views. (b) 2D Brillouin zone for single-layer VSi_2P_4. (c) Enlarged low-energy band dispersions of single-layer VSi_2P_4 with SOC. The Fermi level is set to 0 eV.

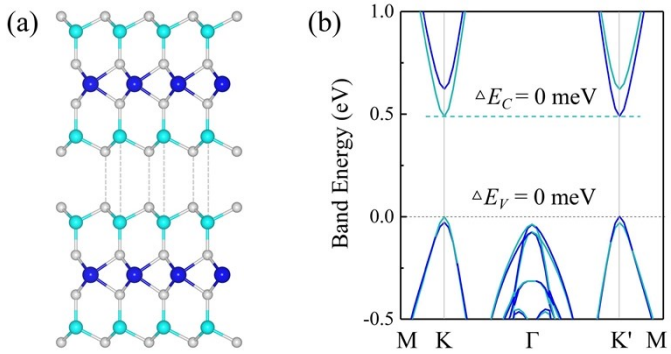

Fig. S2 (a) Crystal structure and (b) enlarged low-energy band dispersions of AA-stacked bilayer VSi_2P_4. The Fermi level is set to 0 eV.

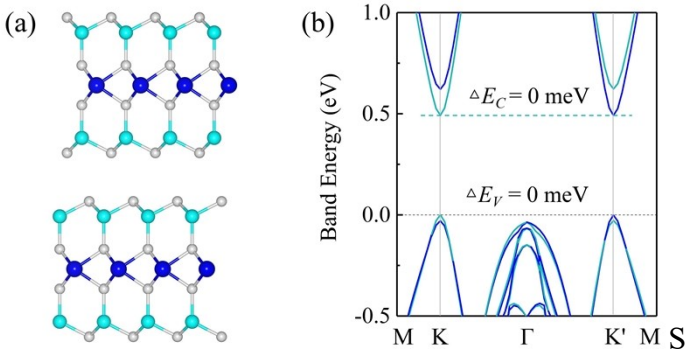

Fig. S3 (a) Crystal structure and (b) enlarged low-energy band dispersions of the paraelectric state in bilayer VSi_2P_4. The Fermi level is set to 0 eV.

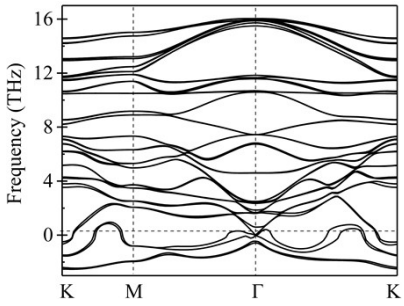

Fig. S4 Phonon spectra of the paraelectric state of bilayer VSi_2P_4.

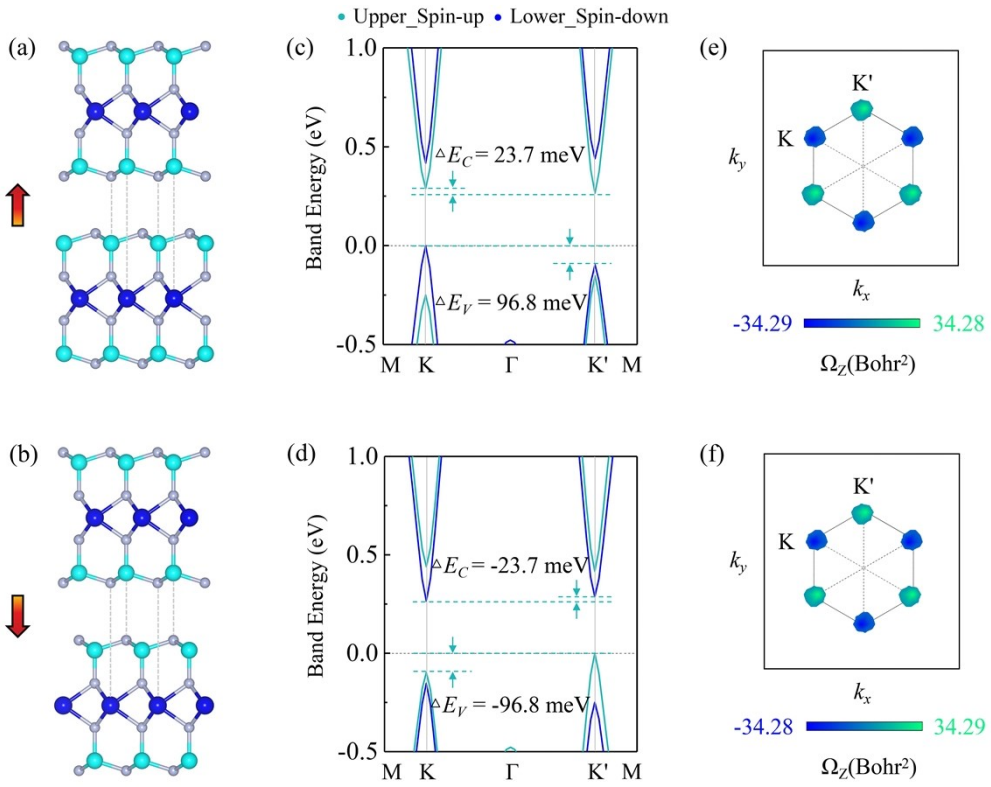

Fig. S5 Crystal structures, enlarged low-energy band dispersions and Berry curvatures of (a,c,e) AB- and (b,d,f) BA-stacked bilayer VSi_2N_4. The Fermi level is set to 0 eV. The arrows in (a) and (b) indicate the directions of ferroelectric polarization.

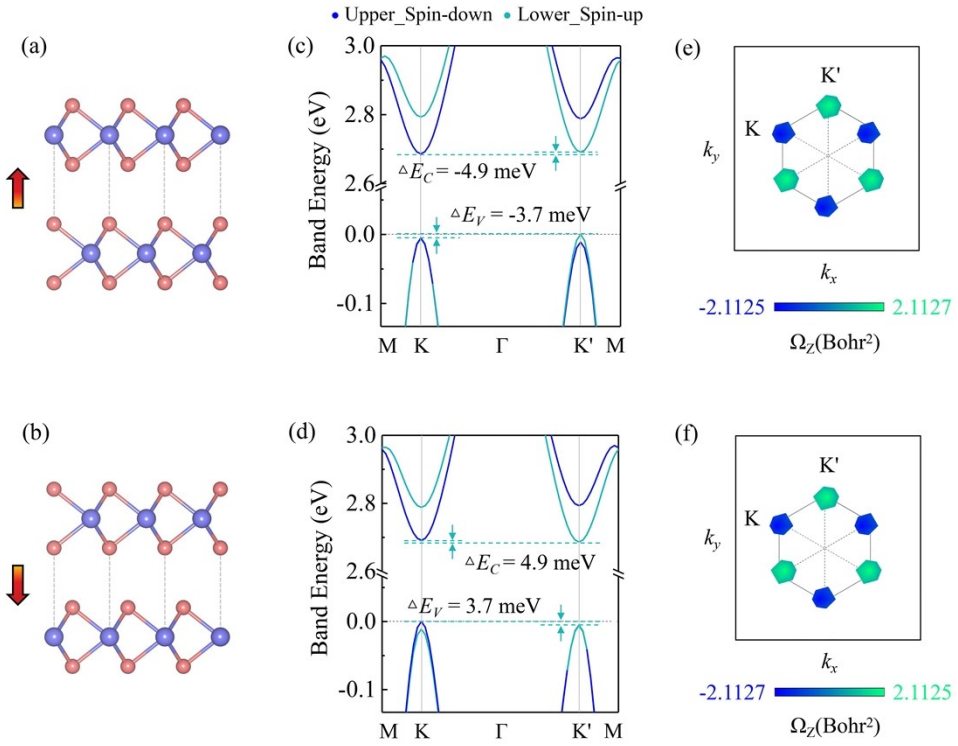

Fig. S6 Crystal structures, enlarged low-energy band dispersions and Berry curvatures of (a,c,e) AB- and (b,d,f) BA-stacked bilayer $FeCl_2$. The Fermi level is set to 0 eV. The arrows in (a) and (b) indicate the directions of ferroelectric polarization.

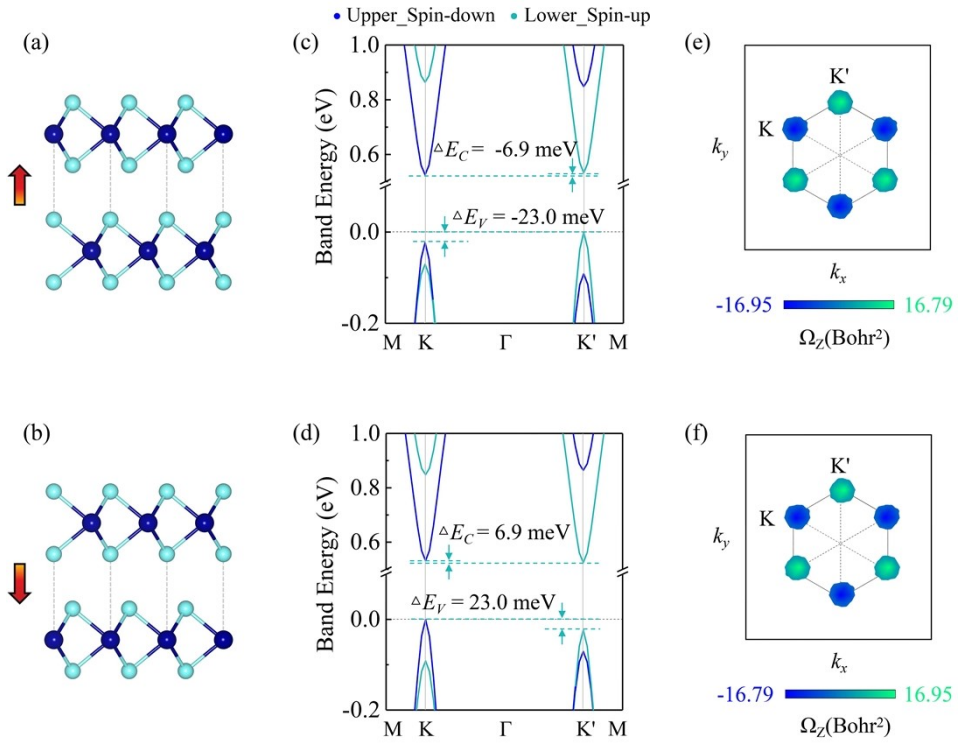

Fig. S7 Crystal structures, enlarged low-energy band dispersions and Berry curvatures of (a,c,e) AB- and (b,d,f) BA-stacked bilayer $RuBr_2$. The Fermi level is set to 0 eV. The arrows in (a) and (b) indicate the directions of ferroelectric polarization.

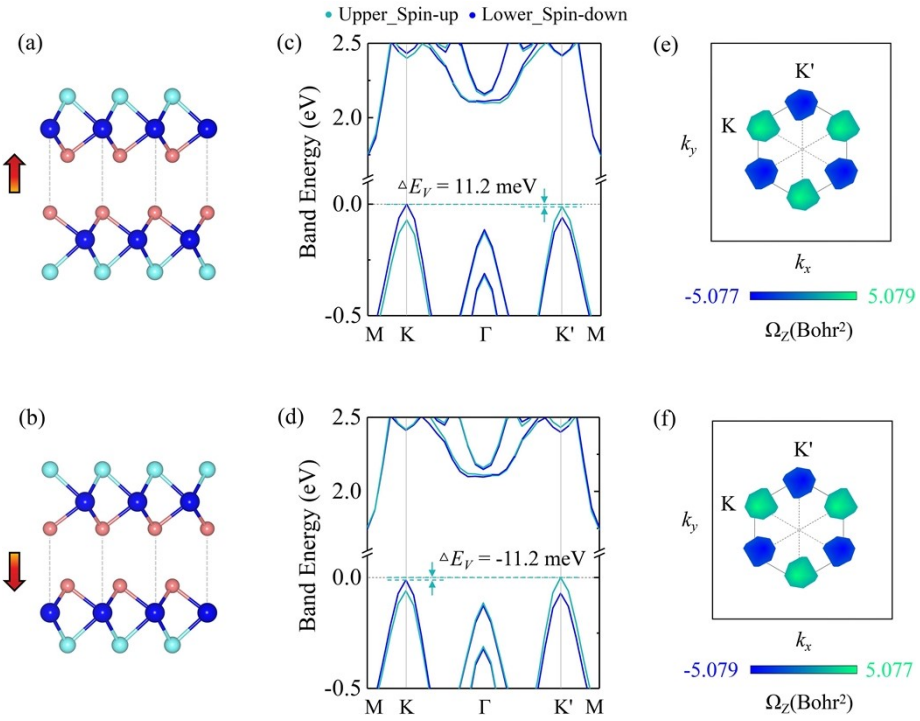

Fig. S8 Crystal structures, enlarged low-energy band dispersions and Berry curvatures of (a,c,e) AB- and (b,d,f) BA-stacked bilayer VClBr. The Fermi level is set to 0 eV. The arrows in (a) and (b) indicate the directions of ferroelectric polarization.