Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information

Nonvolatile electrical control of valley splitting by ferroelectric polarization switching in two-dimensional $AgBiP_2S_6/CrBr_3$ multiferroic heterostructure

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Table S1. Interlayer Distances d_i (Å) between $AgBiP_2S_6$ and $CrBr_3$, Binding Energy E_b (meV/atom), Band-Gap E_g (eV), Total Magnetic Moments M_{tot} (μ_B), and Energy Difference $\Delta E_{FM\text{-}AFM}$ (meV) for different configurations of the $AgBiP_2S_6/CrBr_3$ vdW heterostructures.

Model		d_i (Å)	E_b (meV/atom)	E_g (eV)	M_{tot} (μ_B)	$\Delta E_{FM\text{-}AFM}$（meV）
I	**P+**	3.15	-188.32	1.19	5.84	-42.86
	P-	3.73	-182.14	1.42	5.84	-41.09
II	**P+**	3.34	-185.62	1.15	5.84	-41.29
	P-	3.76	-179.47	1.41	5.84	-40.90
III	**P+**	3.26	-187.60	1.17	5.84	-42.39
	P-	3.80	-179.46	1.39	5.84	-40.70

Table S2. The lattice vectors and fractional coordinates of each atom in $AgBiP_2S_6/CrBr_3$ heterostructure with P+ state.

a[Å]	6.3516737391921954	0.0000000338711972	0.0000000000000000
b[Å]	-3.1758368989294152	5.5007107977067626	0.0000000000000000
c[Å]	0.0000000000000000	0.0000000000000000	29.9647006988999998
S	0.9909565350833234	0.6153824755154375	0.4476054072655399
S	0.3846175114845642	0.3755740305678862	0.4476054072655399
S	0.6244259944321126	0.0090435039166768	0.4476054072655399
S	0.3376691530403998	0.0519300521851825	0.3323641611108283
S	0.9480699058148216	0.2857391048552255	0.3323641611108283
S	0.7142609531447792	0.6623308659595973	0.3323641611108283
Br	0.0172715030579112	0.3322965585541545	0.5528608831528239
Br	0.6677034714458490	0.6849749345037568	0.5528608831528239
Br	0.3150250724962477	0.9827284889420868	0.5528608831528239
Br	0.6662523392471248	0.9823661886040973	0.6488545151122515
Br	0.0176338493958982	0.6838861076430283	0.6488545151122515
Br	0.3161138873569764	0.3337477077528752	0.6488545151122515
P	0.6666667060000009	0.3333333380000028	0.3501788018525476
P	0.6666667060000009	0.3333333380000028	0.4257640105547345
Ag	-0.0000000000000000	0.0000000000000000	0.4255378381306612
Bi	0.3333333380000028	0.6666666750000019	0.3798650698726054
Cr	-0.0000000000000000	0.0000000000000000	0.6009813130007060
Cr	0.3333333380000028	0.6666666750000019	0.6007581156644234

Table S3. The lattice vectors and fractional coordinates of each atom in $AgBiP_2S_6/CrBr_3$ heterostructure with P− state.

a[Å]	6.3479298765991805	0.0000001374165001	0.0000000000000000
b[Å]	-3.1739650573057689	5.4974684658205364	0.0000000000000000
c[Å]	0.0000000000000000	0.0000000000000000	29.9647006988999998
S	0.9904235723877730	0.6172551697405315	0.3247562426483977
S	0.6268316253527615	0.0095764396122271	0.3247562426483977
S	0.3827448272594640	0.3731684036472387	0.3247562426483977
S	0.3386597105855122	0.0513976193629148	0.4403088212043350
S	0.7127379277773972	0.6613402904144827	0.4403088212043350
S	0.9486023436370893	0.2872621002226043	0.4403088212043350
Br	0.6666200061735178	0.6849993356892514	0.5648679352483319
Br	0.0183794035157319	0.3333799998264828	0.5648679352483319
Br	0.3150006453107513	0.9816205814842700	0.5648679352483319
Br	0.3158648572492148	0.3332540523854736	0.6609852731176955
Br	0.0173891871362584	0.6841351397507880	0.6609852731176955
Br	0.6667459386145294	0.9826108368637442	0.6609852731176955
P	0.6666666750000019	0.3333333380000028	0.4220389154458148
P	0.6666666750000019	0.3333333380000028	0.3463370249089215
Ag	0.0000000000000000	0.0000000000000000	0.3519161150966360
Bi	0.3333333380000028	0.6666667060000009	0.3922070613437507
Cr	0.3333333380000028	0.6666667060000009	0.6130059525070672
Cr	0.0000000000000000	0.0000000000000000	0.6129801780415449

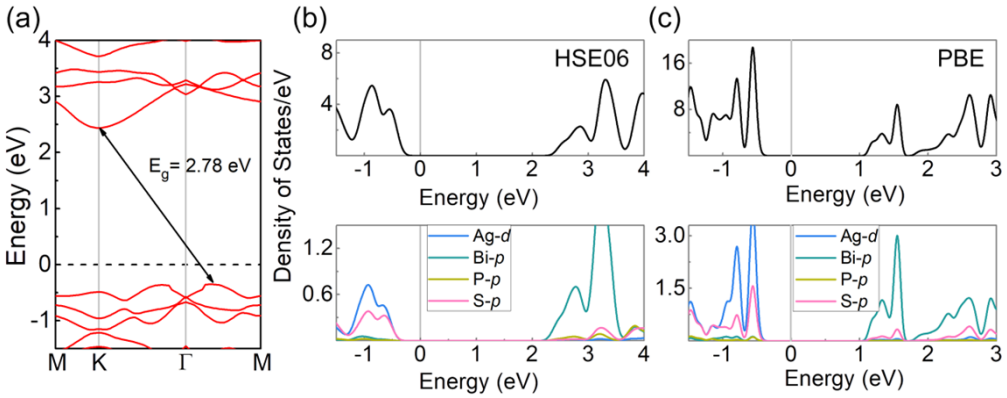

Fig. S1 (a) Band structure of the monolayer $AgBiP_2S_6$ calculated by HSE06 method. The total DOS and orbital-resolved PDOS for each atom calculated by (b) HSE06 and (c) PBE method, respectively.

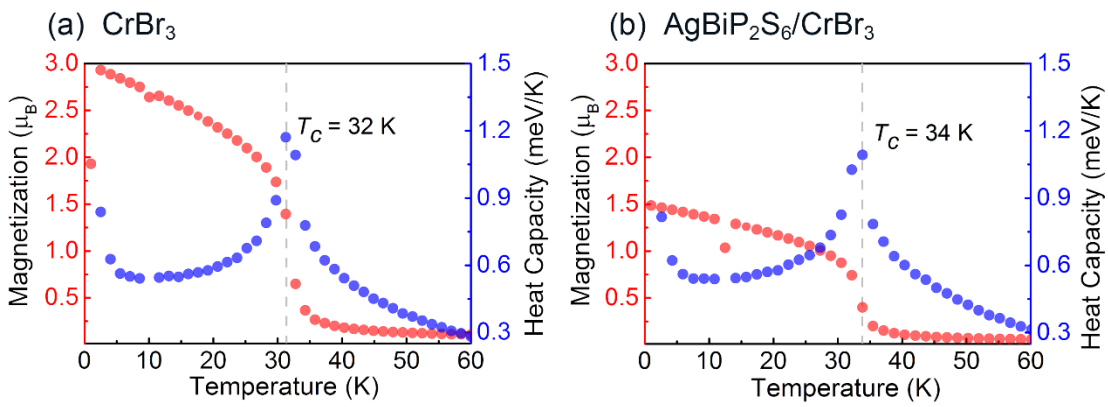

Fig. S2 The magnetization and heat capacity as functions of temperature for (a) $CrBr_3$ monolayer and (b) $AgBiP_2S_6/CrBr_3$ heterostructure.

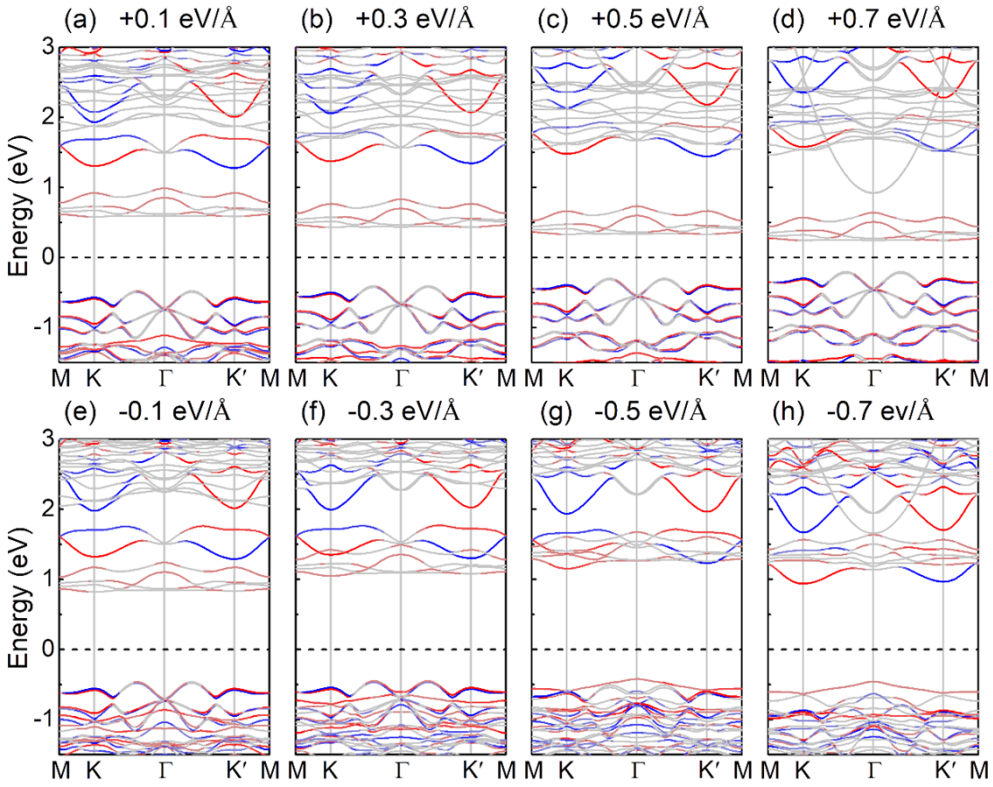

Fig. S3 Calculated band structures of the $AgBiP_2S_6/CrBr_3$ heterostructure in the polarized state P+ under different electric field from –0.7 V/Å to +0.7 V/Å. The Fermi level is set to zero.

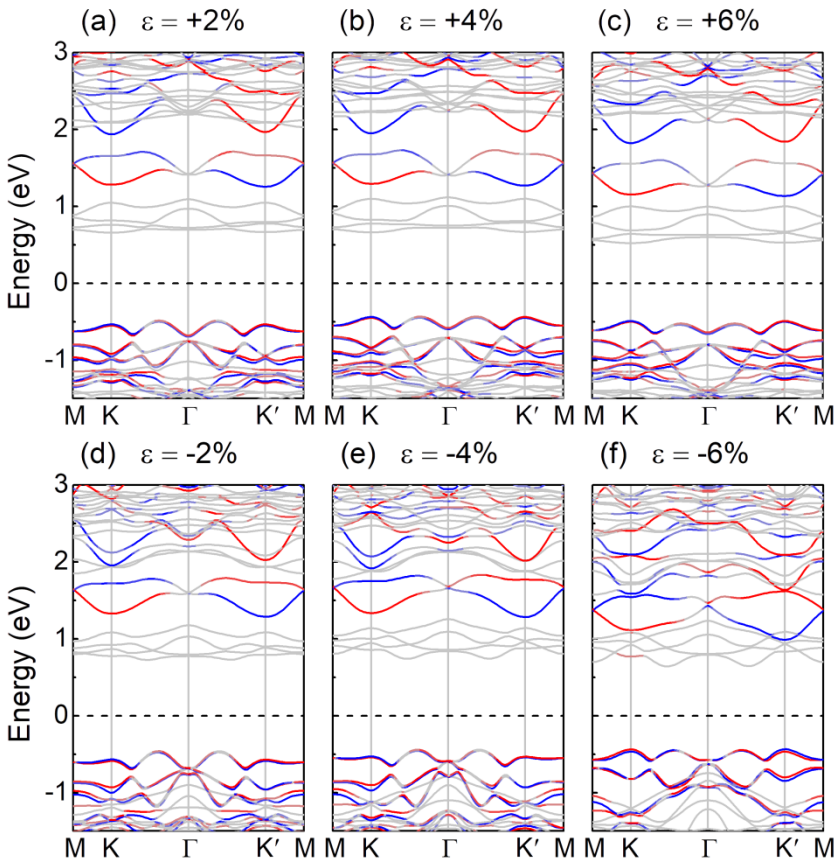

Fig. S4 Band structures of $AgBiP_2S_6/CrBr_3$ heterostructure in the polarized state P+ under different strain from −6% to +6%. The Fermi level is set to zero.

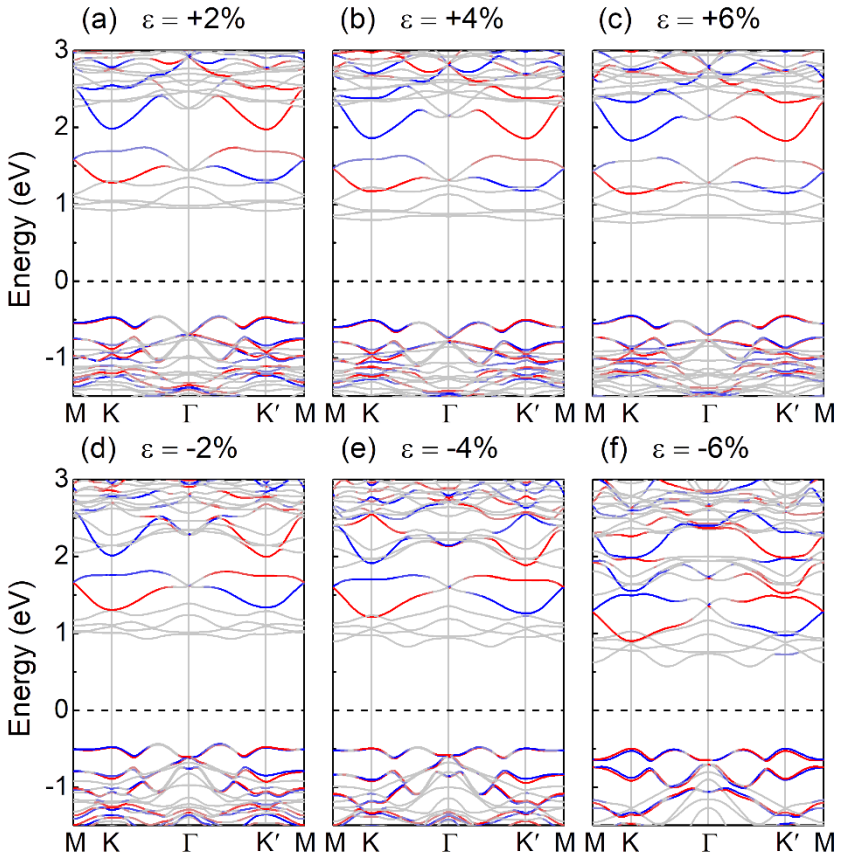

Fig. S5 Band structures of $AgBiP_2S_6/CrBr_3$ heterostructure in the polarized state P– under different strain from –6% to +6%. The Fermi level is set to zero.