Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information for

The Effect of Switchable Electronic Polarization States on the Electronic Properties of Two-dimensional Multiferroic $TMBr_2/Ga_2SSe_2$ (TM=V-Ni) Heterostructures

Jinlian Lu[a#], Nini Guo[b#], Yuanyuan Duan[c], Shu Wang[c], Yuxuan Mao[c], Sun Yi [c], Lijuan Meng[a], Xiaojing Yao [b], Xiuyun Zhang[c,*]

[a]Department of Physics, Yancheng Institute of Technology, Yancheng, Jiangsu 224051, China.

[b]College of Physics and Hebei Advanced Thin Films Laboratory, Hebei Normal University, Shijiazhuang 050024, China.

[c]College of Physics Science and Technology & Microelectronics Industry Research Institute, Yangzhou University, Yangzhou 225002, China.

[#]Jinlian Lu and Nini Guo contributed equally to this work.

*Corresponding author: xyzhang@yzu.edu.cn

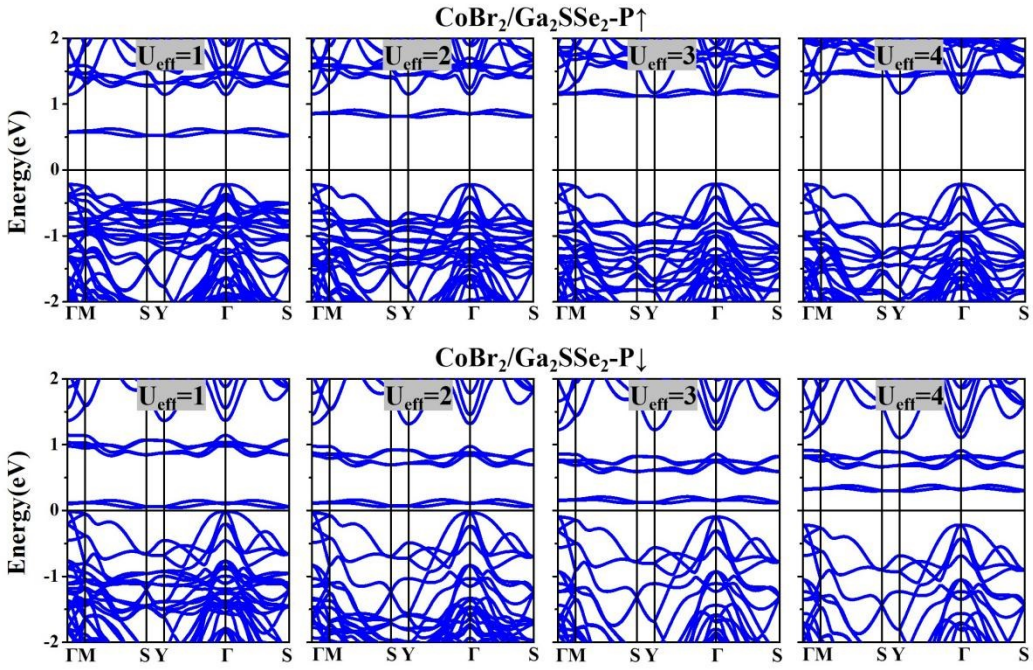

Fig. S1. The band structure of $CoBr_2/Ga_2SSe_2$-P↑ and $CoBr_2/Ga_2SSe_2$-P↓ HSs calculated using different U_{effs} (=1.0 ~4.0 eV).

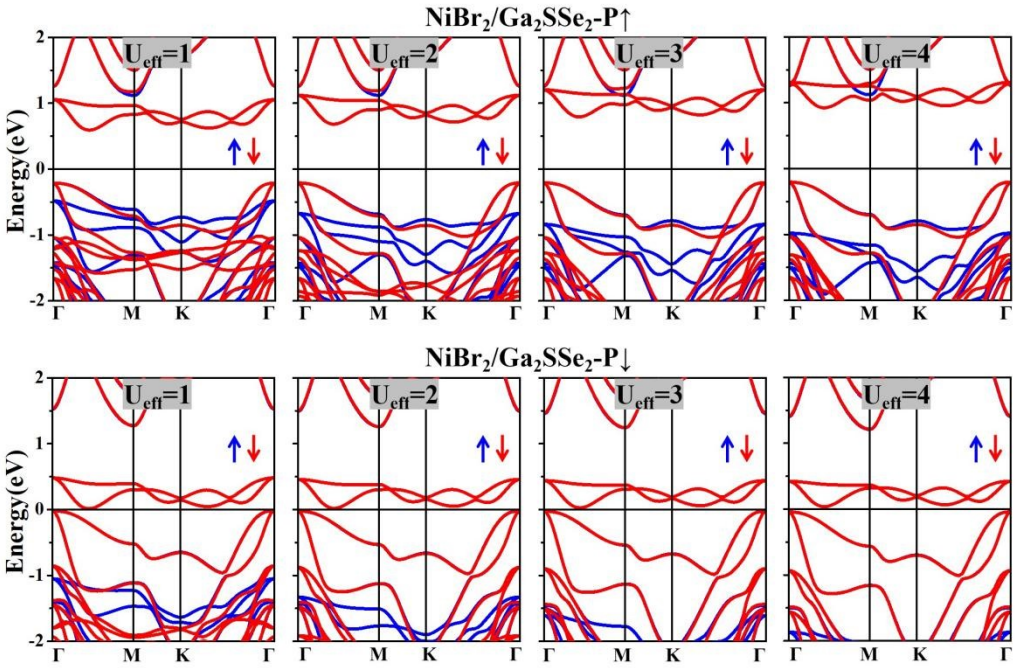

Fig. S2. The band structure of $NiBr_2/Ga_2SSe_2$-P↑ and $NiBr_2/Ga_2SSe_2$-P↓ HSs calculated using different U_{effs} (=1.0 ~4.0 eV).

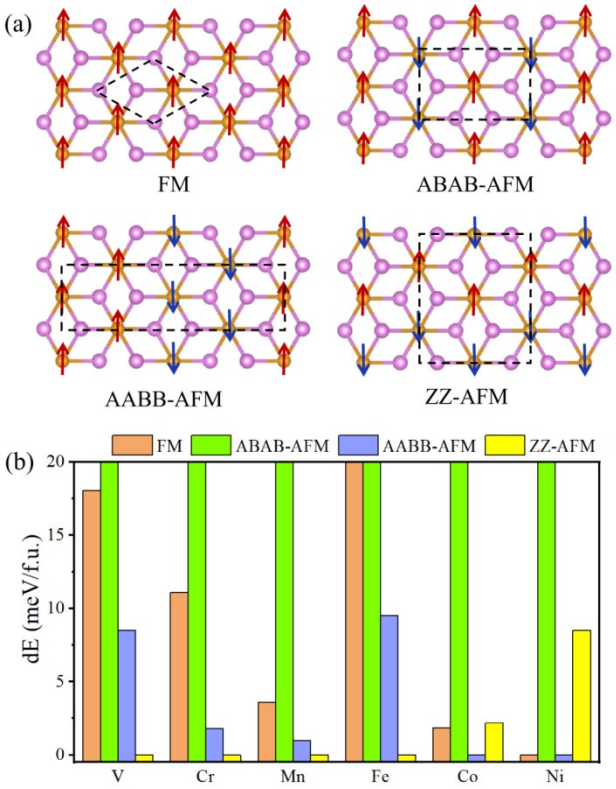

Figure S3. (a) Four possible magnetic states of $TMBr_2$ monolayers (TM=V-Ni). (b)Energy diagram of $TMBr_2$ monolayers with different magnetic states, Where the ground state is set to zero.

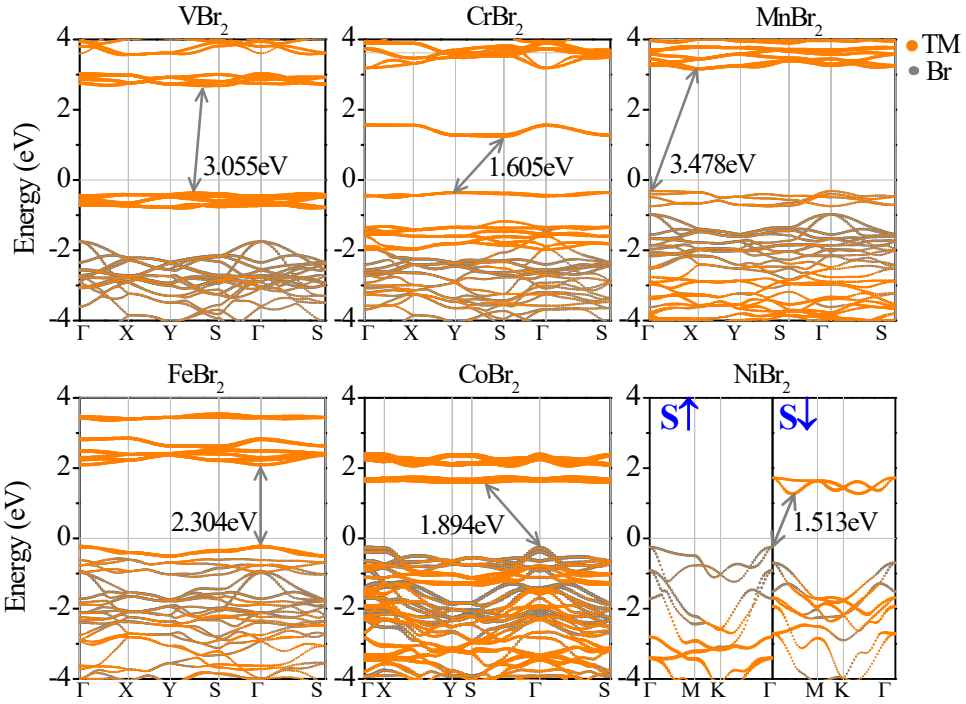

Figure S4. Band structures of $TMBr_2$ monolayers.

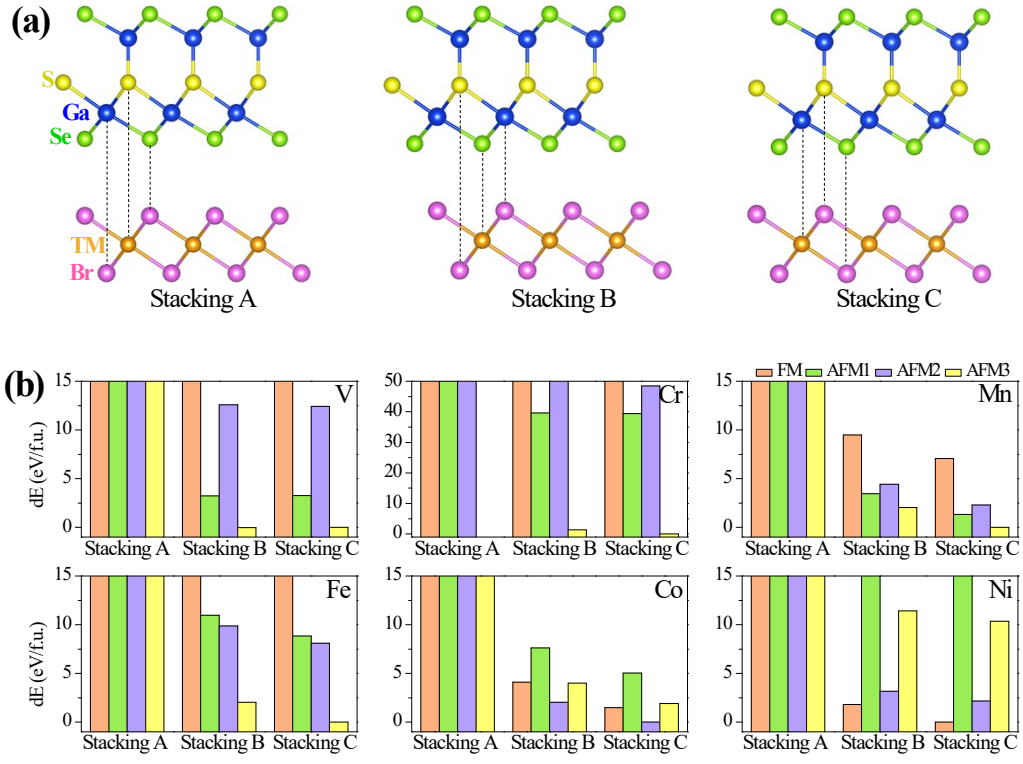

Figure S5. (a) Side view of three different interfacial contact types for the $TMBr_2/Ga_2SSe_2$ heterostructures. (b) Energy diagram of $TMBr_2/Ga_2SSe_2$ HSs with different magnetic states, Where the ground state is set to zero.

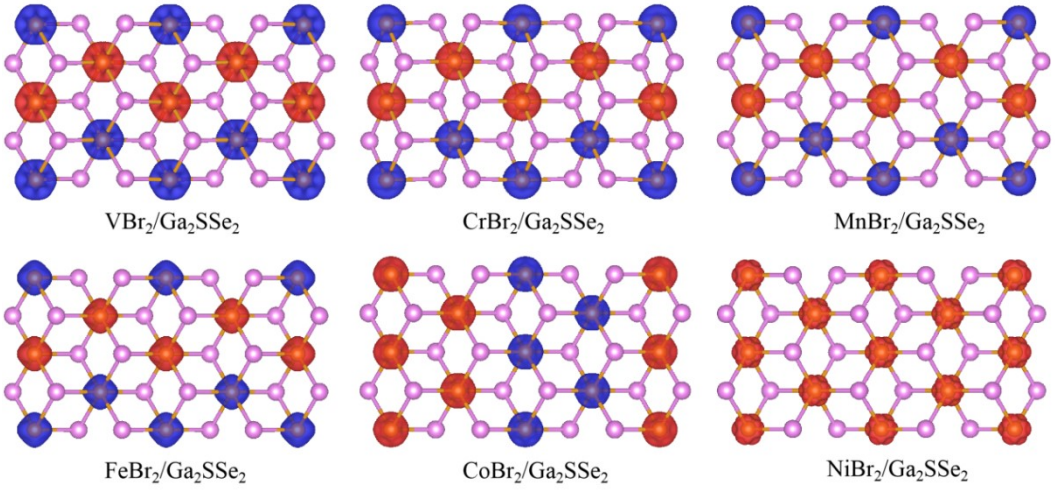

Figure S6. Top views of atomic structures with spin-polarized charge density distribution for $TMBr_2/Ga_2SSe_2$ HSs.

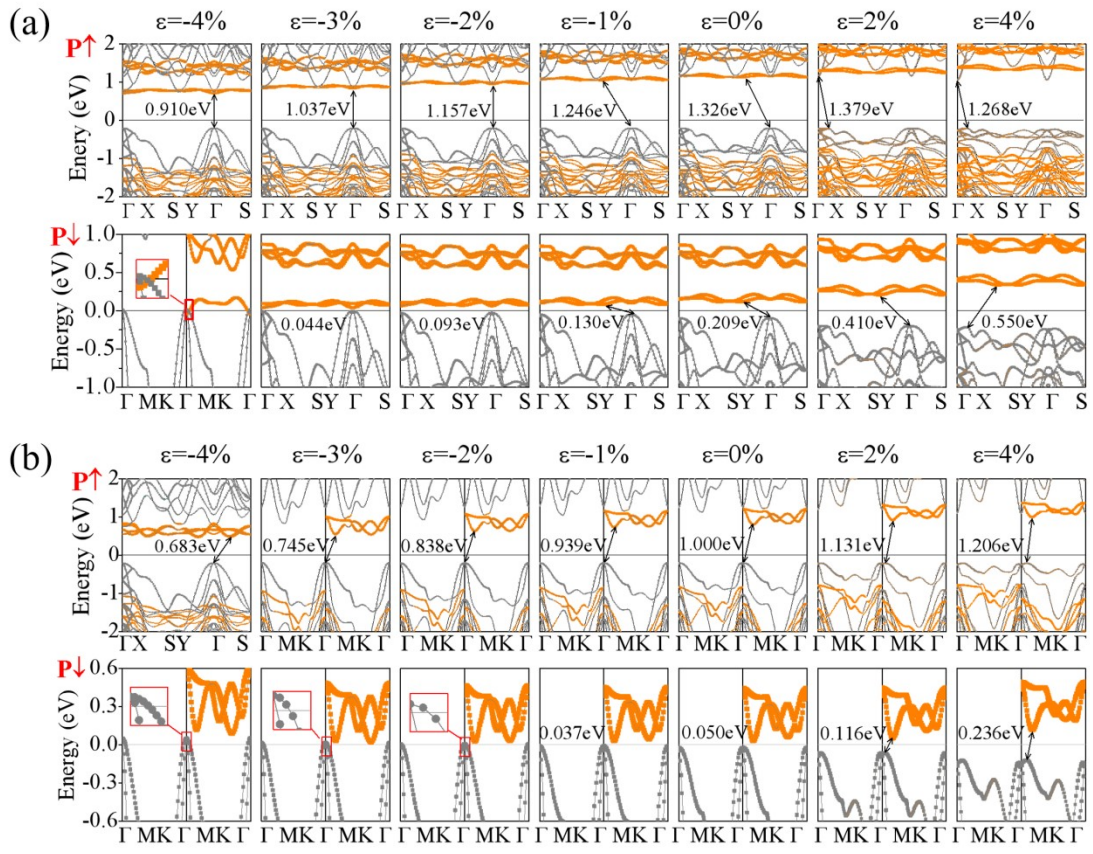

Figure S7. Band structures of (a) $CoBr_2/Ga_2SSe_2$ and (b) $NiBr_2/Ga_2SSe_2$ HSs under biaxial strains with upward and downward ferroelectric polarization.

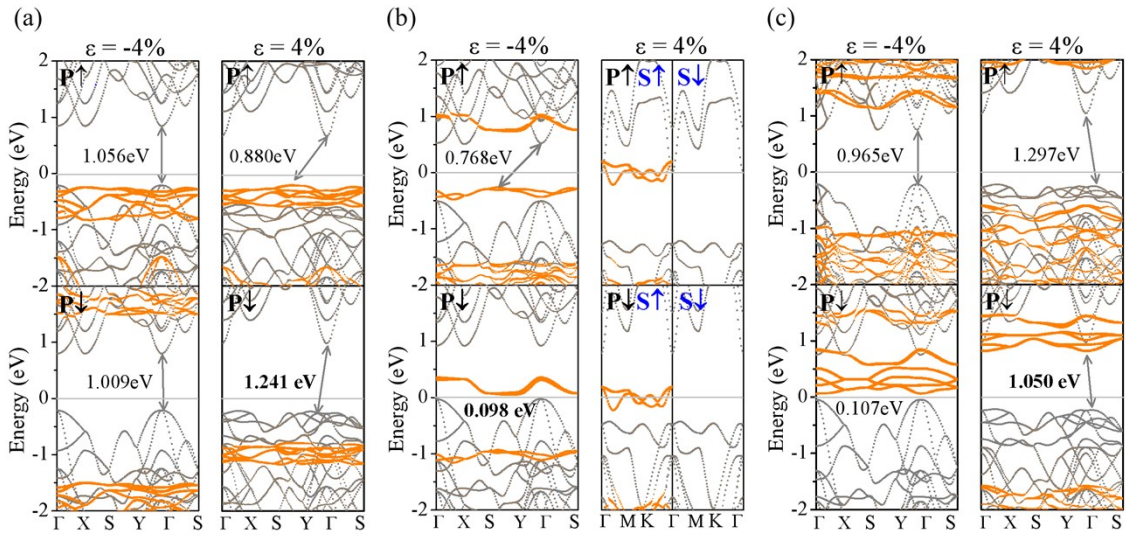

Figure S8. Band structures of (a) VBr_2/Ga_2SSe_2, (b) $CrBr_2/Ga_2SSe_2$, and $FeBr_2/Ga_2SSe_2$ HSs under biaxial strains 4% with upward and downward ferroelectric polarization.

Table S1. The energy differences of ferromagnetic (FM) and antiferromagnetic (AFM) states in Co(Ni)Br_2/Ga_2SSe_2-P↑ and Co(Ni)Br_2/Ga_2SSe_2-P↓ HSs.

	$CoBr_2/Ga_2SSe_2$-P↑				**$CoBr_2/Ga_2SSe_2$-P↓**			
U_{eff}	**FM**	**ABAB-AFM**	**AABB-AFM**	**ZZ-AFM**	**FM**	**ABAB-AFM**	**AABB-AFM**	**ZZ-AFM**
1	1.4	8.91	0	3.83	0.57	9.62	0	4.03
2	1.63	6.41	0	2.4	1.41	6.85	0	2.46
3	1.48	5.05	0	1.90	2.23	5.57	0	2.37
4	1.05	3.79	0	1.38	1.1	7.49	0	5.65
	$NiBr_2/Ga_2SSe_2$-P↑				**$NiBr_2/Ga_2SSe_2$-P↓**			
U_{eff}	**FM**	**ABAB-AFM**	**AABB-AFM**	**ZZ-AFM**	**FM**	**ABAB-AFM**	**AABB-AFM**	**ZZ-AFM**
1	0	13.09	1.69	7.88	0	25.42	3.83	15.86
2	0	21.01	2.75	12.71	0	20.53	3.18	12.93
3	0	17.01	2.16	10.35	0	16.59	2.36	10.43
4	0	13.75	1.88	8.42	0	13.65	2.03	8.56

Table S2. The lattice parameters and magnetic ground states of $TMBr_2$ (TM=V-Ni) monolayer.

	a	mag static	a	mag static	thickness
VBr2	3.76[1]	ABAB-AFM[1]	3.82	ZZ-AFM	3.029
CrBr2	3.74[1]	ABAB-AFM[1]	3.79	ZZ-AFM	3.069
MnBr2	3.80[1]	ABAB-AFM[1]	3.85	ZZ-AFM	3.093
FeBr2	3.63[1]	FM[1]	3.72	ZZ-AFM	3.024
CoBr2	3.62[1]	FM[1]	3.72	AABB-AFM	2.923
NiBr2	3.61[1]	FM[1]	3.64	FM	2.8925
Ga2SSe2	3.73[2]	/	3.73	/	6.162

[1]Journal of Materials Chemistry C, 2017, 5, 8734--8741.

[2]Physical Review B, 2022, 105, 075402.

Table S3. The magnetic anisotropy energy per TM atom (meV) along *x*, *y*, *z* axis of $TMBr_2/Ga_2SSe_2$-P↑ and $TMBr_2/Ga_2SSe_2$-P↓ (TM=V-Ni) HSs.

	x(meV)	*y*(meV)	*z*(meV)
VBr_2/Ga_2SSe_2 -P↑	0	0.02	0
VBr_2/Ga_2SSe_2 -P↓	0	0	0
$CrBr_2/Ga_2SSe_2$-P↑	-0.42	-0.13	0
$CrBr_2/Ga_2SSe_2$ -P↓	-0.20	0.08	0
$MnBr_2/Ga_2SSe_2$ -P↑	0.03	0.02	0
$MnBr_2/Ga_2SSe_2$ -P↓	0	0	0
$FeBr_2/Ga_2SSe_2$ -P↑	-1.98	7.36	0
$FeBr_2/Ga_2SSe_2$ -P↓	-0.01	-0.08	0
$CoBr_2/Ga_2SSe_2$ -P↑	-1.11	-1.26	0
$CoBr_2/Ga_2SSe_2$ -P↓	-1.11	-1.27	0
$NiBr_2/Ga_2SSe_2$ -P↑	0.05	0.06	0
$NiBr_2/Ga_2SSe_2$ -P↓	-0.04	-0.03	0

Table S4. Magnetic ground states $TMBr_2/Ga_2SSe_2$ (TM=V, Cr, Fe) heterostructures under biaxial strains 4% with upward and downward ferroelectric polarization.

	ε = -4%		ε = +4%	
	P↑	P↓	P↑	P↓
V	AFM3	AFM3	AFM3	AFM3
Cr	AFM3	AFM3	FM	FM
Fe	AFM2	AFM3	AFM3	AFM3