

Supplementary Information

Nonvolatile Switching of Intra- and Interlayer Magnetism in 2D CrSe₂/Sc₂CO₂ Multiferroic Heterostructures

Yueyue Lu,¹ Jiayi Wang,¹ Lili Kang,^{1, a)} Zhenzhen Feng,¹ Gaofeng Zhao,^{1, 2, b)} and Peng Jiang^{4, c)}

1)Institute for Computational Materials Science, School of Physics and Electronics, International Joint Research Laboratory of New Energy Materials and Devices of Henan Province, Henan University, Kaifeng 475004, China

2)School of Flexible Electronics (SoFE) and Henan Institute of Flexible Electronics (HIFE), Henan University, 379 Mingli Road, Zhengzhou 450046, China

3)School of Physics and Electronic Engineering, Jiangsu Normal University, Xuzhou 221116, China

a) Electronic mail: llkang@henu.edu.cn

b) Electronic mail: gfzhao@henu.edu.cn

c) Electronic mail: pjiang@jsnu.edu.cn

Table S1: The total energy of the CrSe₂ rectangular supercell under four different magnetic configurations.

Magnetic order	ΔE (eV)
FM	0
sAFM-aabb	-0.05
sAFM-abab	-0.12
zAFM	-0.13

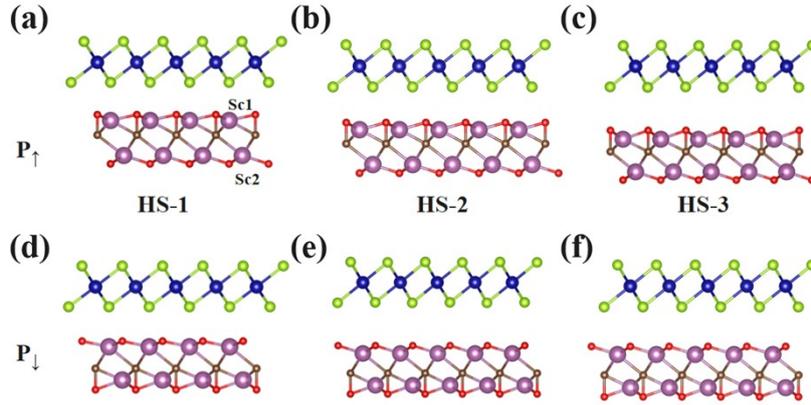


Fig. S1: Three different stacking configurations of the 1L-CrSe₂/Sc₂CO₂ vdW heterostructures. (a) and (d) are the HS-1 configuration, in which the Cr atoms of the CrSe₂ are vertically positioned above the O atoms of Sc₂CO₂; (b) and (e) are the HS-2 configuration, in which the Cr atoms of the CrSe₂ are vertically positioned above the Sc1 atoms of Sc₂CO₂; (c) and (f) are the HS-3 configuration, in which the Cr atoms of the CrSe₂ are vertically positioned above the Sc2 atoms of Sc₂CO₂.

Table S2: The calculated energies and the formation energies of the 1L-CrSe₂/Sc₂CO₂ heterostructures with HS-1, HS-2, and HS-3 configurations and different magnetic orders. The unit is eV.

	Magnetic order	$\Delta E (P_{\uparrow})$	$\Delta E (P_{\downarrow})$	$E_b(P_{\uparrow})$	$E_b(P_{\downarrow})$
HS-1	sAFM-abab	0.05	0.20	-0.157	-0.130
	sAFM -aabb	-0.01	0.13	-0.158	-0.131
	zAFM	-0.07	0.10	-0.158	-0.131
	FM	0.00	0.00	-0.158	-0.132
HS-2	sAFM-abab	1.49	1.15	-0.141	-0.122
	sAFM -aabb	1.46	1.14	-0.142	-0.123
	zAFM	1.38	1.02	-0.142	-0.123
	FM	1.49	1.19	-0.139	-0.120
HS-3	sAFM-abab	2.30	0.75	-0.132	-0.126
	sAFM -aabb	2.26	0.73	-0.133	-0.127
	zAFM	2.17	0.62	-0.134	-0.128
	FM	2.34	0.76	-0.130	-0.123

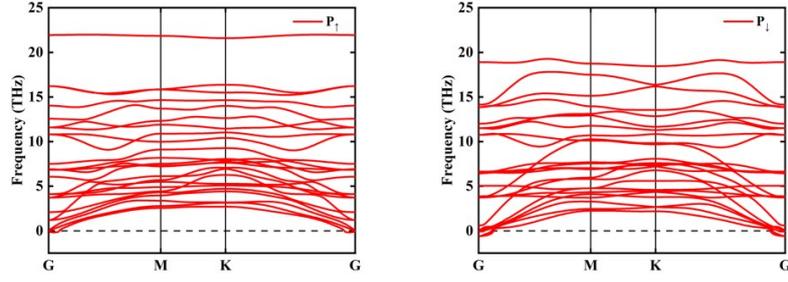


Fig. S2: The phonon spectrum of the heterostructure with HS-1 configuration for P_{\uparrow} and P_{\downarrow} states. At the Γ point, there is a small imaginary frequency in the phonon spectrum. After examination, it is found that the vibration mode is the rigid translation/rotation of the entire system. This is due to the precision limit of the numerical calculation and does not affect the thermodynamic stability of the structure.

Table S3: The MAE of the monolayer CrSe_2 (1L- CrSe_2) and the 1L- $\text{CrSe}_2/\text{Sc}_2\text{CO}_2$ vdW heterostructure for P_{\uparrow} and P_{\downarrow} states under different U values.

U	Configurations	MAE (meV/Cr)	EMA
2.5eV	1L- CrSe_2	0.15	x
	HS- P_{\uparrow}	0.23	y
	HS- P_{\downarrow}	0.26	z
3.0eV	1L- CrSe_2	0.17	x
	HS- P_{\uparrow}	0.24	y
	HS- P_{\downarrow}	0.11	z
3.5eV	1L- CrSe_2	0.14	x
	HS- P_{\uparrow}	0.25	y
	HS- P_{\downarrow}	0.15	z
4.0eV	1L- CrSe_2	0.05	x
	HS- P_{\downarrow}	0.13	x

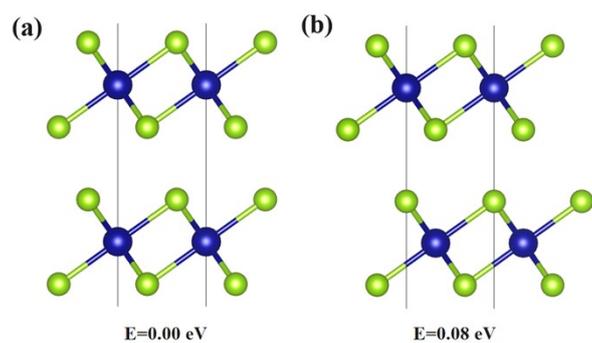


Fig. S3: The atomic structures of the 2L-CrSe₂: (a) and (b) are the AA and AB stacking configurations, respectively.

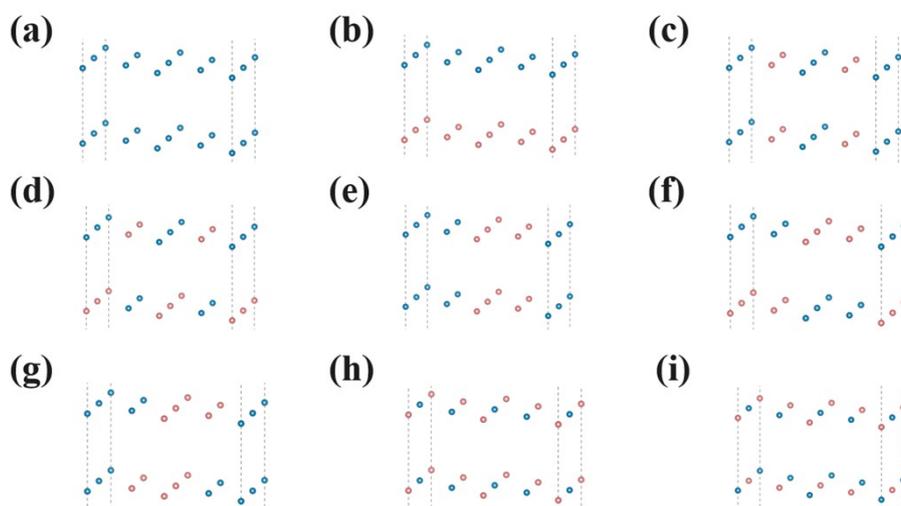


Fig. S4: The different magnetic configurations of 2L-CrSe₂. The spheres in the figure represent Cr atoms. The blue and red colors indicate spin-up and spin-down, respectively.

Table S4: The calculated energies of the nine magnetic configurations of the 2L-CrSe₂ as plotted in Fig. S4.

Magnetic order	ΔE (eV)
a	0.00
b	-0.12
c	0.33
d	0.39
e	0.30
f	0.30
g	0.46
h	0.40
i	1.07

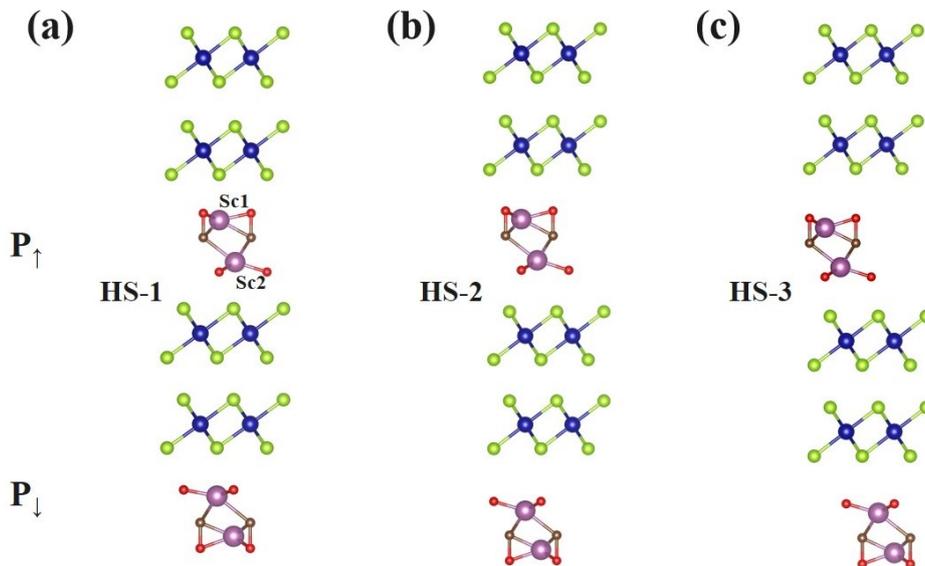


Fig. S5: Three configurations of the 2L-CrSe₂/Sc₂CO₂ heterostructures.

Table S5: The calculated energies and the formation energies of the 2L-CrSe₂/Sc₂CO₂ heterostructures with HS-1, HS-2, and HS-3 configurations and different magnetic orders. The unit is meV.

	Magnetic order	$\Delta E (P_{\uparrow})$	$\Delta E (P_{\downarrow})$	$E_b (P_{\uparrow})$	$E_b (P_{\downarrow})$
HS-1	AFM	11.79	-0.63	-0.199	-0.176
	FM	0.00	0.00	-0.201	-0.177
HS-2	AFM	196.87	179.02	-0.183	-0.164
	FM	190.61	174.76	-0.184	-0.165
HS-3	AFM	309.49	119.28	-0.173	-0.169
	FM	302.08	114.36	-0.175	-0.170