Supplementary Information

Nonvolatile Magnetoelectric Coupling in Two-dimensional Ferromagnetic-bilayer /Ferroelectric van der Waals Heterostructure

Wenxuan Wang,[a] Wei Sun,[a] Hang Li,[a]* Ying Bai,[a] Fengzhu Ren,[a] Caiyin You,[b] Zhenxiang Cheng[c]*

[a] *School of Physics and Electronics, Henan University, Kaifeng 475004, People's Republic of China*

[b] *School of Materials Science & Engineering, Xi'an University of Technology, Xi'an 710048, People's Republic of China*

[c] *Institute for Superconducting & Electronic Materials, Australian Institute of Innovative Materials, University of Wollongong, Innovation Campus, Squires Way, North Wollongong, NSW 2500, Australia*

* *Corresponding author, Zhenxiang Cheng Tel:13638644511, fax: 61-2-42215731 and E-mail address: cheng@uow.edu.au*

* *Corresponding author, Hang Li, E-mail address: hang.li@vip.henu.edu.cn*

Calculation for Hubbard U

Our calculations were performed by the DFT method implemented in Vienna *ab* initio Simulation package (VASP)[1]. Considering corrections on the strong correlation of the 3d electrons, spin-polarized GGA + U calculations were applied throughout the study and the Hubbard U of

Dudarev implementation[2] applied on the 3d orbitals of the Cr ions. The reasonable U value is determined by comparing with the experimental lattice constant, magnetic moment and band gap. Here, we tested the values of the Hubbard U for CrOBr (COB), and the results are summarized in the following table. It can be seen from the Table S1. that the calculated magnetic moments and lattice constants of monolayer COB and the bulk COB are close to each other for the same U values. Owing to the fact that we can nowhere to get the experimental data of monolayer COB, thus, we can determine the U value of Cr by comparing with the experimental lattice data of bulk COB. On the other hand, there is no experimental data about the band gap of bulk COB, so we consider its homologous compound CrOCl as a comparative, as shown in Table S1. We found that the lattice constant, magnetic moment and band gap increase as a function of the U value (from 0 to 7). Comparing the calculated results with the experimental data, we can draw the following conclusions:

1) When U = 0, the lattice constants are in good agreement with the experimental values.

2) When U = 7, the calculated band gap is closer to the experimental values.

3) In terms of magnetic moment, even if we increase U to 7, a great disparity still exists between the experiment values and the calculated ones. From a theoretical point of view, it is difficult for Cr3+ to reach its magnetic moment value above 3.6 μB measured in experiment, for example, the magnetic moment of CrOBr is 2.9 μB in theory [3], which is in good agreement with our results. While, there might be other reasons for it to reach 3.6 μB in the experiment.

4) For Cr ions in most 3d transition metal compounds, the U value is usually set as 2–5 eV, such as CrOX (X = F, Cl, Br)[3], CrSX (X = Cl, Br, I) [4-5], CrX3 (X=Cl, Br, I) [6-7], CrWI6[8], CrWGe2Te6 [8], CrXTe (X = S, Se)[9].

Combined, in order to consider both the lattice constant and the band gap, we choose the intermediate value U_{eff} = 4 eV to do calculation for our system.

Table S1 Calculated lattice constant *a*, *b*, *c* (Å), magnetization *m* (μB) and band gaps (E_g) for CrOBr monolayer, CrOBr bulk, and CrOCl bulk with different *U* values ($U = 0 \sim 7$). Their experimental values (exp.) are also listed for comparison. [10-11]

		0	1	2	3	4	5	6	7	exp.
CrOBr monolayer	*a*	3.3	3.31	3.32	3.33	3.34	3.36	3.36	3.38	—
	b	3.85	3.87	3.88	3.9	3.91	3.93	3.94	3.95	—
	m	2.88	2.93	2.98	3.02	3.06	3.1	3.13	3.16	—
CrOBr bulk	*a*	3.31	3.32	3.32	3.34	3.35	3.36	3.37	3.38	3.23
	b	3.85	3.87	3.88	3.9	3.91	3.93	3.94	3.95	3.86
	c	16.48	16.5	16.5	16.53	16.55	16.58	16.6	16.64	8.36
	m	2.88	2.93	2.98	3.02	3.06	3.1	3.13	3.16	3.74
CrOCl bulk	*a*	3.21	3.22	3.23	3.24	3.25	3.26	3.27	3.28	3.18
	b	3.85	3.86	3.88	3.9	3.91	3.92	3.93	3.94	3.86
	c	15.55	15.55	15.57	15.59	15.63	15.63	15.68	15.72	7.69
	m	2.87	2.92	2.96	2.99	3.03	3.06	3.09	3.12	3.68
	E_g	0.53	0.88	1.26	1.61	1.93	2.19	2.41	2.6	2.8

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