# Supplemental Material for "Electronic and magnetic properties modulated by nonvolatile switching in the multiferroic $Cr_2Cl_3S_3/Ga_2O_3$ van der Waals heterostructure"

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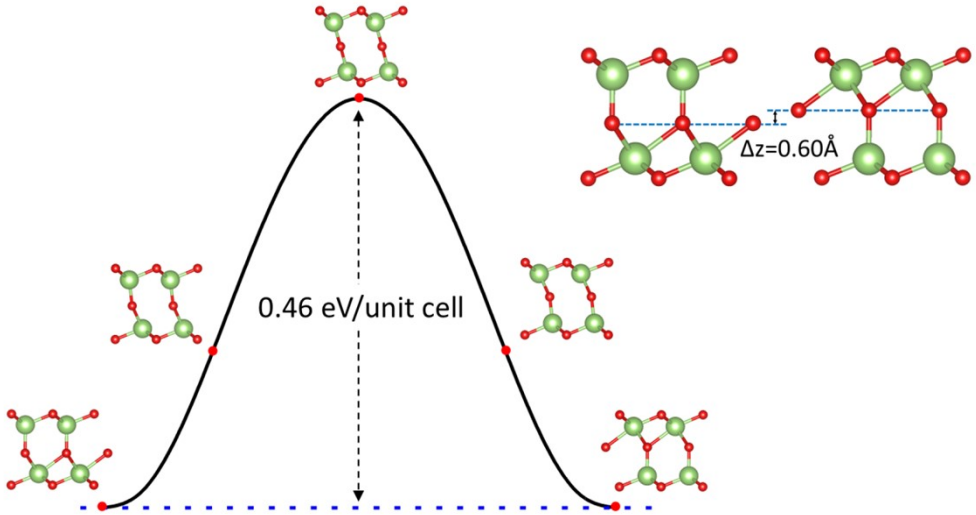


Fig. S1 The Kinetic pathway of polarization reversal process and ionic displacement (Δz) from P↑ to P↓ for $Ga_2O_3$ monolayer.

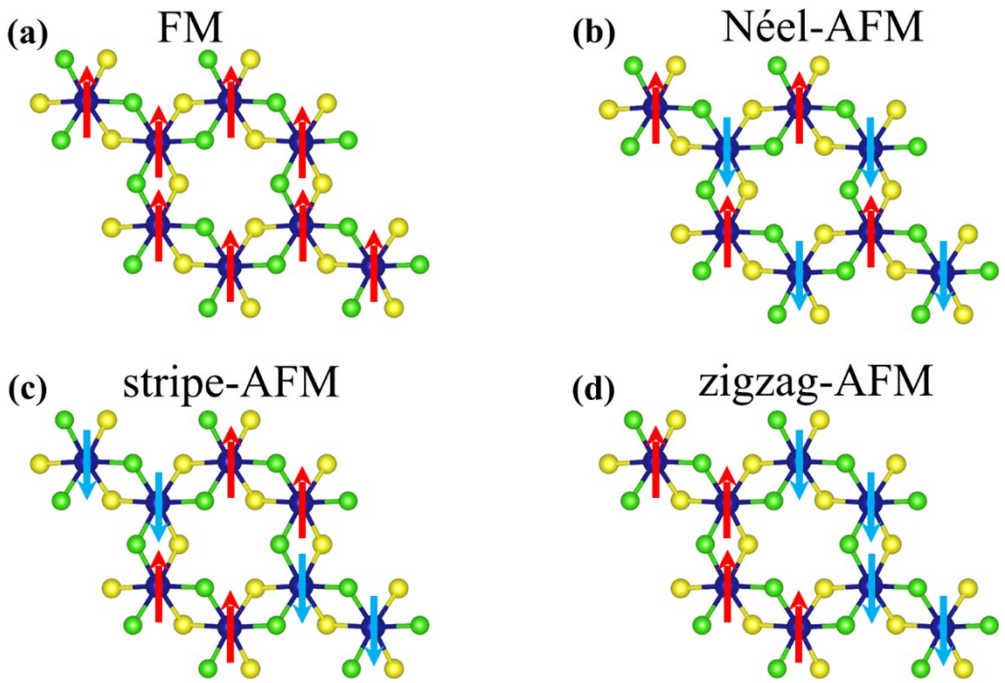


Fig. S2 (a) FM configuration, and the three AFM configurations: (b) Néel-AFM (nAFM), (c) stripe-AFM (sAFM), (d) zigzag-AFM (zAFM) states considered for $Cr_2Cl_3S_3$ monolayer.

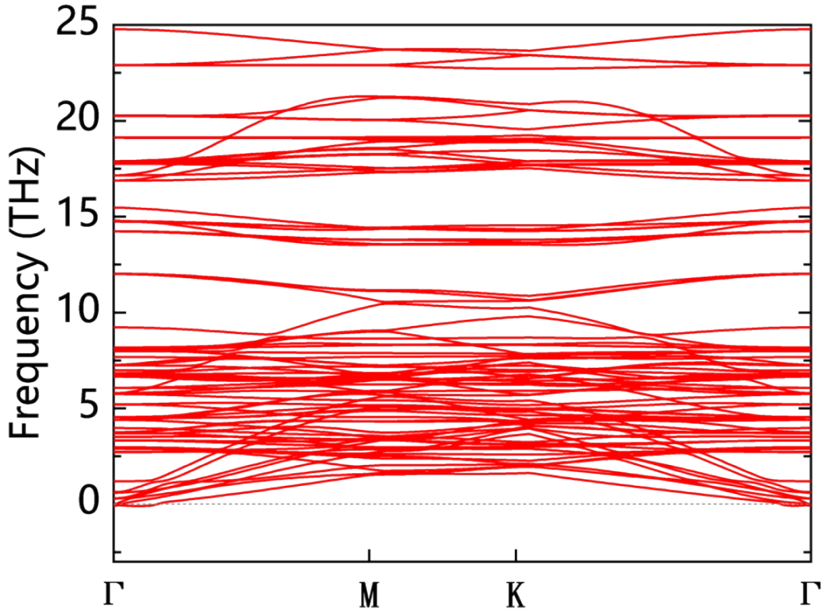


Fig. S3 Phonon dispersion for the $Cr_2Cl_3S_3/Ga_2O_3$ heterostructure.

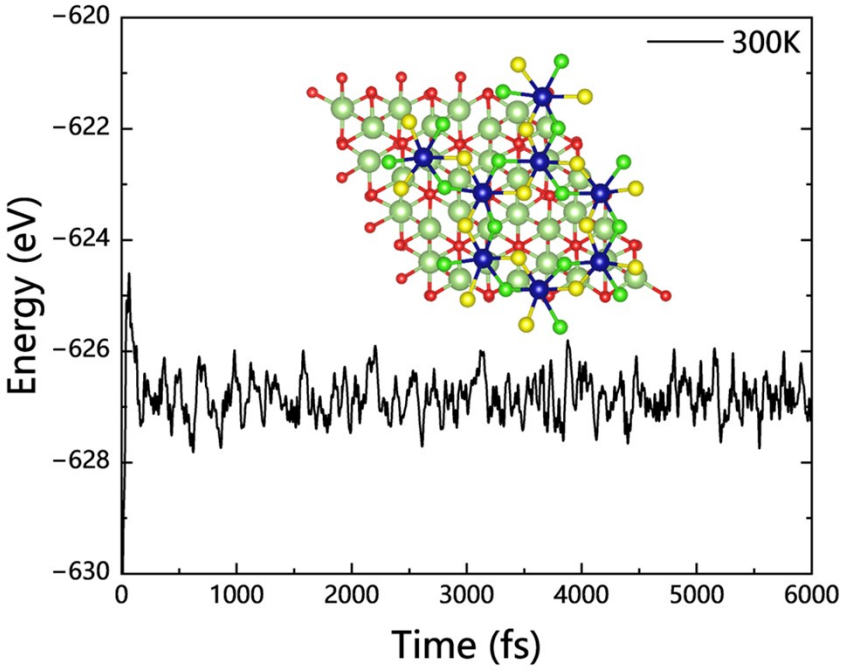


Fig. S4 Fluctuation of total energy and the final structure of $Cr_2Cl_3S_3/Ga_2O_3$ heterostructure after 6000 fs at 300 K from AIMD simulations.

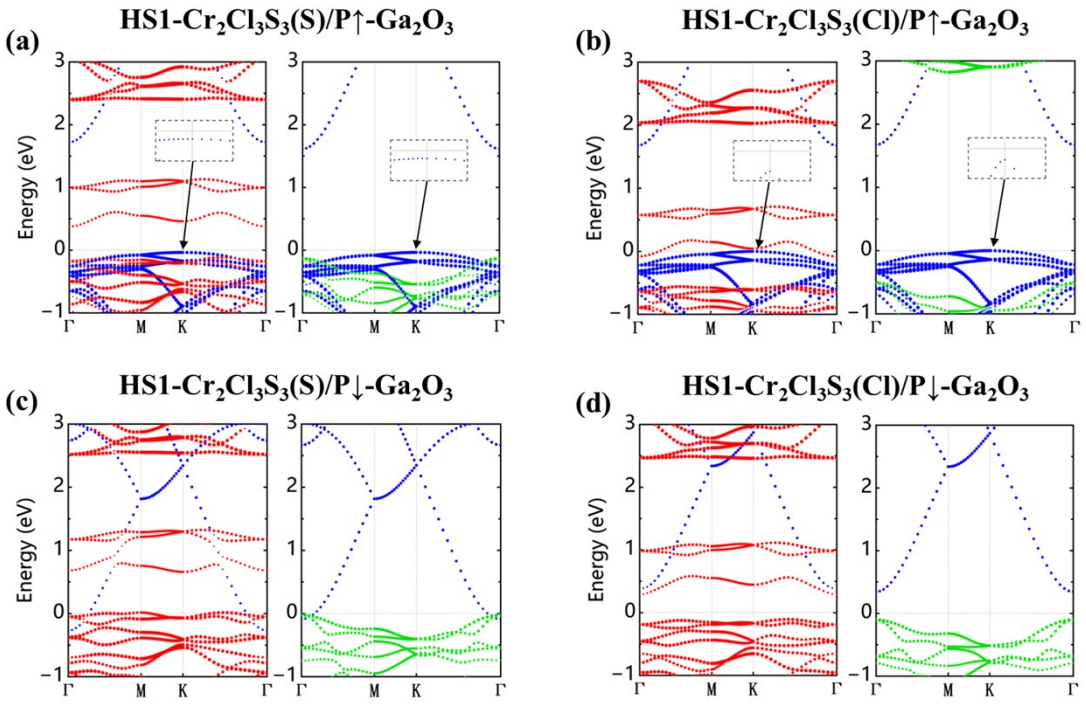


Fig. S5 Band structures of the four heterostructures (a) HS1-$Cr_2Cl_3S_3$(S)/P↑-$Ga_2O_3$, (b) HS1-$Cr_2Cl_3S_3$(Cl)/P↑-$Ga_2O_3$, (c) HS1-$Cr_2Cl_3S_3$(S)/P↓-$Ga_2O_3$ and (d) HS1-$Cr_2Cl_3S_3$(Cl)/P↓-$Ga_2O_3$, respectively. Red and green dots denote the spin-up and spin-down contributions of $Cr_2Cl_3S_3$, respectively, while blue dots represent the spin-degenerate projected bands of $Ga_2O_3$.

Fig. S6 Band structures of the four heterostructures (a) HS2-$Cr_2Cl_3S_3$(S)/P↑-$Ga_2O_3$, (b) HS2-$Cr_2Cl_3S_3$(Cl)/P↑-$Ga_2O_3$, (c) HS2-$Cr_2Cl_3S_3$(S)/P↓-$Ga_2O_3$ and (d) HS2-$Cr_2Cl_3S_3$(Cl)/P↓-$Ga_2O_3$, respectively. Red and green dots denote the spin-up and spin-down contributions of $Cr_2Cl_3S_3$, respectively, while blue dots represent the spin-degenerate projected bands of $Ga_2O_3$.

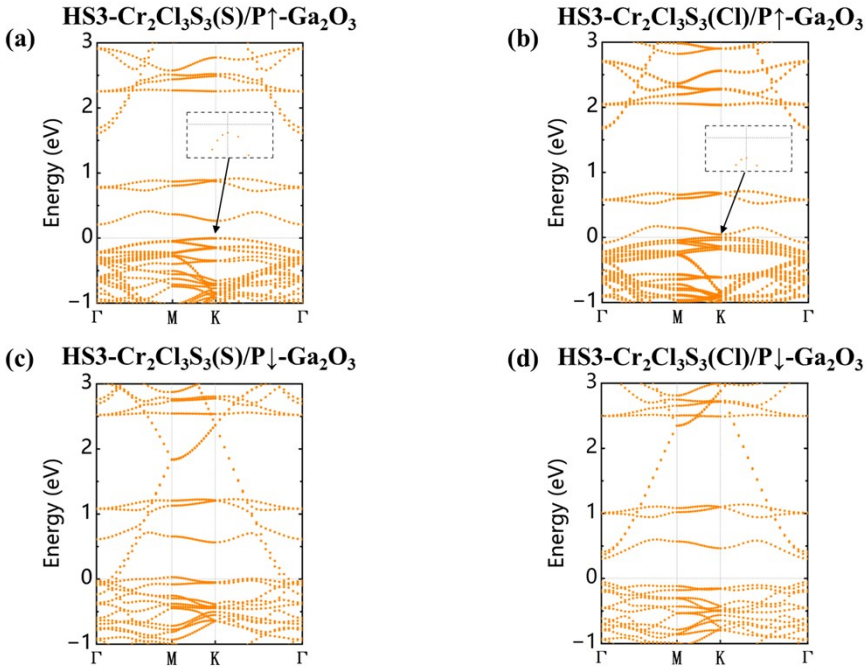


Fig. S7 Band structures of the four heterostructures with SOC effect: (a) HS3-$Cr_2Cl_3S_3$(S)/P↑-$Ga_2O_3$, (b) HS3-$Cr_2Cl_3S_3$(Cl)/P↑-$Ga_2O_3$, (c) HS3-$Cr_2Cl_3S_3$(S)/P↓-$Ga_2O_3$, and (d) HS3-$Cr_2Cl_3S_3$(Cl)/P↓-$Ga_2O_3$, respectively.

Table SI Work functions ($\Phi_1$ and $\Phi_2$), spin-up and spin-down conduction band offset ($\Delta E_{CBO}^{\uparrow/\downarrow}$), and valence band offsets ($\Delta E_{VBO}^{\uparrow/\downarrow}$) of the four HS3-$Cr_2Cl_3S_3$/$Ga_2O_3$ heterostructures

| Configuration | $\Phi_1$ (eV) | $\Phi_2$ (eV) | $\Delta E_{CBO}^{\uparrow}$ (eV) | $\Delta E_{VBO}^{\uparrow}$ (eV) | $\Delta E_{CBO}^{\downarrow}$ (eV) | $\Delta E_{VBO}^{\downarrow}$ (eV) |
|---|---|---|---|---|---|---|
| HS3-$Cr_2Cl_3S_3$(S)/P↑-$Ga_2O_3$ | 8.43 | 7.33 | -1.49 | -0.29 | 1.4 | -0.28 |
| HS3-$Cr_2Cl_3S_3$(S)/P↓-$Ga_2O_3$ | 4.21 | 7.41 | 0.80 | 1.83 | 3.40 | 1.78 |
| HS3-$Cr_2Cl_3S_3$(Cl)/P↑-$Ga_2O_3$ | 8.69 | 6.20 | -1.77 | -0.55 | 1.14 | -0.47 |
| HS3-$Cr_2Cl_3S_3$(Cl)/P↓-$Ga_2O_3$ | 4.89 | 6.49 | -0.10 | 1.08 | 2.88 | 1.10 |

The work functions $\Phi_1$and $\Phi_2$, corresponding to the $Ga_2O_3$ side and the $Cr_2Cl_3S_3$ side, respectively.

The spin-resolved conduction-band and valence-band offsets are defined as:

$\Delta E_{CBO}^{\uparrow/\downarrow} = E_{CBM}^{\uparrow/\downarrow}(Cr_2Cl_3S_3) - E_{CBM}^{\uparrow/\downarrow}(Ga_2O_3)\#(1)$

$\Delta E_{VBO}^{\uparrow/\downarrow} = E_{VBM}^{\uparrow/\downarrow}(Cr_2Cl_3S_3) - E_{VBM}^{\uparrow/\downarrow}(Ga_2O_3)\#(2)$

Where ↑ and ↓ indicate the spin-up and spin-down channels, respectively, and are distinguished from the symbols P↑ and P↓, which represent the ferroelectric polarization direction of the $Ga_2O_3$ layer. $E_{CBM}^{\uparrow/\downarrow}(Cr_2Cl_3S_3)$ and $E_{VBM}^{\uparrow/\downarrow}(Cr_2Cl_3S_3)$ represent the spin-up/spin-down conduction band minimum (CBM) and valence band maximum (VBM) of the $Cr_2Cl_3S_3$ layer in the heterostructure, respectively, while $E_{CBM}^{\uparrow/\downarrow}(Ga_2O_3)$ and $E_{VBM}^{\uparrow/\downarrow}(Ga_2O_3)$ correspond to those of the $Ga_2O_3$ layer, using the Fermi level as a common reference.

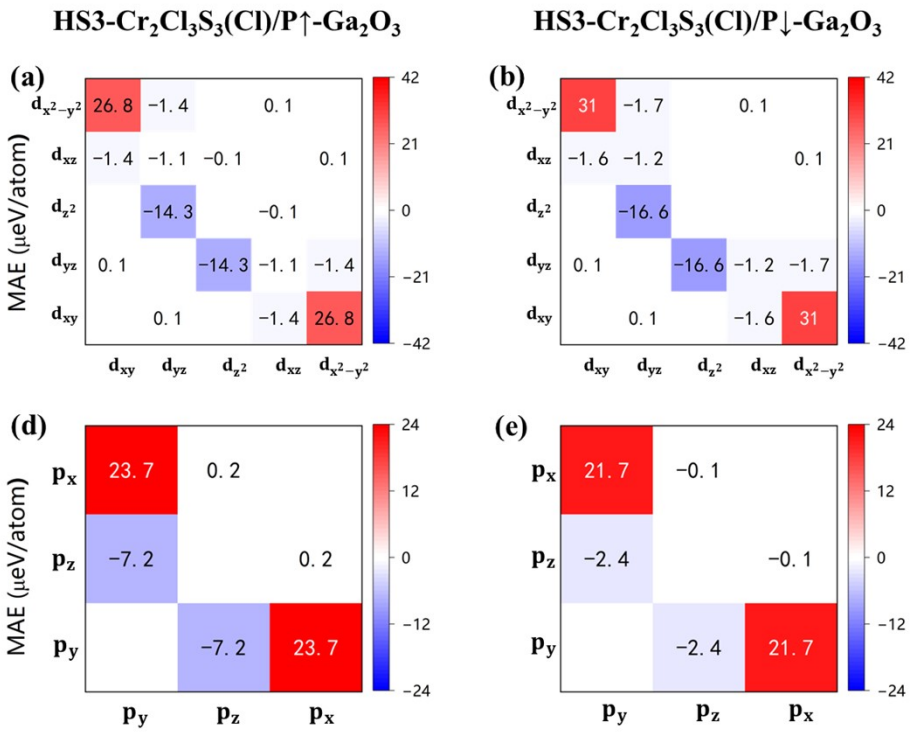


Fig. S8 The orbital-resolved MAE of Cr-*d* orbital for (a) HS3-$Cr_2Cl_3S_3$(Cl)/P↑-$Ga_2O_3$, (b) HS3-$Cr_2Cl_3S_3$(Cl)/P↓-$Ga_2O_3$, the orbital-resolved MAE of S-*p* orbital for (c) HS3-$Cr_2Cl_3S_3$(Cl)/P↑-$Ga_2O_3$, (d) HS3-$Cr_2Cl_3S_3$(Cl)/P↓-$Ga_2O_3$.