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## Electric-controlled Half-metallicity in Magnetic van der Waals Heterobilayer

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Figure S1. Energy difference of laterally-shifted Cr-H with respect to the (a) AB and (b) AB" stack under ferromagnetic (FM) and ferromagnetic (FiM) orders. Two high-symmetry directions of [100] (red lines) and [110] (blue lines) are adopted.



Figure S2. The spin configurations of ferromagnetism (FM), ferrimagnetism (FiM) and intralayer antiferromagnetism (I-AFM). The intralayer magnetic couplings of nearest-neighboring Cr in CrI<sub>3</sub> ( $J_{CrI}$ ) and CrGeTe<sub>3</sub> ( $J_{CrGeTe}$ ) and magnetic couplings of interlayer nearest-neighboring (J) and second-nearest-neighboring (J') Cr atoms are marked in each stacking Cr-H. Red and blue arrows represent the settings of spin up and down for Cr atoms, respectively.

Table S1. Total energies (the unit of eV) under different spin configurations for the stable and metastable Cr-H. The results including the spin orbital coupling (SOC) effect are displayed as a comparison. All of the energies are obtained without any restriction.

Stack orders	AB	AA	AB'	AC1	AC <sub>2</sub> '	AD <sub>1</sub> '	AD <sub>2</sub> '
GGA+U							
FM	-76.764278	-76.708827	-76.742676	-76.739958	-76.749612	-76.693814	-76.706868
FiM	-76.760788	-76.706077	-76.747028	-76.737715	-76.755930	-76.696397	-76.709770
FiM-1	-76.703346	-76.651654	-76.688867	-76.681077	-76.695603	-76.637432	-76.655102
FiM-2	-76.660946	-76.607818	-76.654053	-76.648559	-76.659365	-76.603040	-76.620217
I-AFM	-76.614452	-76.556177	-76.600088	-76.593127	-76.604981	-76.551667	-76.562124
GGA+U+SOC							
FM	-78.263343	-78.210958	-78.242785	-78.240082	-78.250671	-78.192333	-78.205899
FiM	-78.259603	-78.209699	-78.246296	-78.235519	-78.25509	-78.194654	-78.209115
FiM-1	-78.200794	-78.151133	-78.185761	-78.178599	-78.193244	-78.134853	-78.147612
FiM-2	-78.150289	-78.110263	-78.144495	-78.138834	-78.149099	-78.092690	-78.109468
I-AFM	-78.102579	-78.045667	-78.083754	-78.081036	-78.092767	-78.039032	-78.048037



Figure S3. Spin density of (a) AB, (b) AA, (c) AB', (d)  $AC_2'$  (e)  $AC_1'$ , (f)  $AD_1'$  and (g)  $AD_2'$  stack Cr-H with the isovalue of 0.0002  $e/Å^3$ . The yellow and blue areas represent opposite spin components.

### Details on the magnetic coupling parameters calculation

Based on the Heisenberg model, the energies (E) for different magnetic configurations in a unit cell can be expressed as

$$E_{FM} = E_0 + 3(J_{CrI} + J_{CrGeTe})|\vec{S}|^2 + mJ_{inter}|\vec{S}|^2$$
  

$$E_{FiM} = E_0 + 3(J_{CrI} + J_{CrGeTe})|\vec{S}|^2 - mJ_{inter}|\vec{S}|^2$$
  

$$E_{FiM-1} = E_0 + 3(J_{CrGeTe} - J_{CrI})|\vec{S}|^2 + pJ_{inter}|\vec{S}|^2$$
  

$$E_{FiM-2} = E_0 + 3(J_{CrI} - J_{CrGeTe})|\vec{S}|^2 + qJ_{inter}|\vec{S}|^2$$

where, E0 is the ground state energy for each stacking order, which is independent of the spin configuration.  $J_{Crl}$ ,  $J_{CrGeTe}$  and  $J_{inter}$  represent the hopping of Crl<sub>3</sub> and CrGeTe<sub>3</sub> intralayer and interlayer between nearest neighboring Cr atoms. Besides, *m*, *p*, *q* are the net magnetic coupling of interlayer Cr atoms under different spin configurations (As shown in Table S2).

 $|\vec{S}|$  is the value of magnetic monment of Cr atoms, it is simply fixed to be  $2 \mu_B$  in our calculations. Then, we can estimate the hopping parameters ( $J_{Crl}$ ,  $J_{\underline{CrGeTe}}$  and  $J_{inter}$ ) by solving the above equations.

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Table S2. The net magnetic coupling (m, n and p) of interlayer nearest neighboring Cr atoms for different stacking orders under different magnetic configurations. Positive and negative values represent the same and opposite directions for spins, respectively.

	AB	AA	AB'	AC <sub>1</sub> '	AC <sub>2</sub> '	AD <sub>1</sub> '	AD <sub>2</sub> '
<i>m</i> (FM)	1	2	4	4	4	2	3
p (FiM-1)	-1	0	0	0	0	0	1
q (FiM-2)	1	0	0	0	0	0	1

#### **Interlayer Magnetic Analysis**

In order to study the relationship between the interlayer magnetic coupling and stacking orders in Cr-H, we calculate the difference in energy between different spin configurations. These total energies are obtained from the unrestricted relaxation and shown in Table S1, and confirmed by the relaxations with spin orbital coupling (SOC) effect. As shown in Table 1, the intralayer exchange coupling parameters of Cr-H are an order of magnitude larger than that of interlayer, and close to those of Crl<sub>3</sub> and CrGeTe<sub>3</sub> monolayers, also leading to the comparable Curie (Néel) temperature to 2D Crl<sub>3</sub> and CrGeTe<sub>3</sub>. For such FM coupling, it is determined by the Cr-I-Cr (Cr-Te-Cr) super-exchange with the bond angle of 90° in Crl<sub>3</sub> (CrGeTe<sub>3</sub>) monolayer, which is consistent with pioneering studies.<sup>1-2</sup>

We next reveal the mechanism of the strongly stacking-dependent interlayer magnetic coupling in Cr-H. Here, the AB and AC<sub>2</sub>' stacks are chosen as two typical structures to illustrate the interlayer exchange mechanism. As shown in Figure S4, the electrons mainly accumulate at the interlayer region, and deplete near neighboring I and Te atoms. Such distribution of interlayer differential charge density indicates the electronic hybridization between the interlayer I and Te atoms, also suggesting the strong Cr-I...Te-Cr super-super-exchange (SSE) interactions. In Figure S4c, we discuss two possible SSE interactions (FM and AFM) between interlayer Cr. (The AFM coupling replaces FiM to clearly show the opposite spin configurations for the interlayer Cr atoms.) Here, Cr<sup>3+</sup> simultaneously possesses the half-filled t<sub>2g</sub> and empty eg orbitals, and I<sup>-</sup> and Te<sup>2-</sup> both have full-filled p orbitals. The interlayer hopping of t<sub>2g</sub>-p...p-eg are allowed in both FM and AFM SSE interactions, whereas that of t<sub>2g</sub>-p...p-t<sub>2g</sub> is permitted in AFM exchange but prohibited in FM exchange. Thus, the hopping of t<sub>2g</sub>-p...p-t<sub>2g</sub> indicates AFM SSE interactions, while t<sub>2g</sub>-p...p-e<sub>g</sub> hybridization leads to FM interactions, due to the Hund's rules.<sup>3</sup> As a result, the interlayer magnetic coupling is dominated by the competition between AFM and FM Cr-I...Te-Cr SSE (or I-Te direct) interactions.<sup>1, 4</sup> Figure S4d and S4e schematically display two paths (as marked in Figure S4a and S4b) for interlayer magnetic coupling. Interlayer AFM and FM couplings are coexist in Cr-H, however, their coordination number is different in different stacking orders. For instance, the AB stack possesses one AFM and nine FM couplings per unit cell, indicating its strong interlayer ferromagnetism. For the AC<sub>2</sub>' stack, the coordination numbers of AFM coupling is increased to four, while the FM coupling is

decreased to eight. Therefore, the combined effect makes the AC<sub>2</sub>' stack the interlayer AFM coupling.



Figure S4. Top and side views of differential charge density (isovalue of 0.0002  $e/Å^3$ ) for the (a) AB and (b) AC<sub>2</sub>' stacks Cr-H. Yellow and cyan areas represent electron accumulation and depletion, respectively. Black and magenta arrows are two different paths between interlayer Cr<sup>3+</sup>. (c) Schematic of Cr-I...Te-Cr SSE interactions under AFM and FM configurations. Schematics for AFM (black) and FM (magenta) magnetic coupling between interlayer Cr<sup>3+</sup> of the (d) AB and (e) AC<sub>2</sub>' stacks, involving p orbitals of I<sup>-</sup> and Te<sup>2-</sup> and t<sub>2g</sub> and e<sub>g</sub> orbitals of Cr<sup>3+</sup>.



Figure S5. Band structures and density of states (DOS) of monolayer (a, b)  $CrI_3$  and (c, d)  $CrGeTe_3$ , respectively. The Fermi level has been set to be zero.



Figure S6. Band structures of (a) AB, (b) AA, (c)  $AC_1'$ , (d) AB', (e)  $AC_2'$ , (f)  $AD_1'$  and (g)  $AD_2'$  stack Cr-H through HSE functional. The indirect bandgaps are marked in the figures.



Figure S7. Partial density of states of (a) AB and (b) AC<sub>2</sub>' stack Cr-H.



Figure S8. The exchange coupling parameters of (a) AB and (b)  $AC_2$ ' stacks as the function of the external electric fields.



Figure S9. The change of magnetic moments of Cr in CrI3 (cyan) and CrGeTe3 (blue) layers under external electric fields for (a) AB and (b)  $AC_2$ ' stacks.

### Electrically induced band structures of CrI<sub>3</sub> and CrGeTe<sub>3</sub> bilayers

As shown in Figure S10a, the gaps of spin up and down channels are both significantly decreased by applying external electric fields in Crl<sub>3</sub> and CrGeTe<sub>3</sub> bilayers. Notably, the bandgaps of two spin channels simultaneously drop to zero after reaching the critical applied field (1.2 V/Å for Crl<sub>3</sub> and 1.0 V/Å for CrGeTe<sub>3</sub>). Figure S10b and S10c clearly show the changes of bandgaps under the increased fields. When the applied field reaches critical field, the valence bands of spin up channel and conduction bands of spin down channel cross the fermi level, indicating the metallic transport in both Crl<sub>3</sub> and CrGeTe<sub>3</sub> bilayers. Such band crossing is due to the positively and negatively doped layers under external electric fields.



Figure S10. (a) Spin-resolved bandgaps of bilayer  $Crl_3$  and  $CrGeTe_3$ . Band structures of bilayer (b)  $Crl_3$  and (c)  $CrGeTe_3$  under external electric field of 0, 0.8 and 1.2 V/Å. Fermi levels (blue dashed lines) are set to be 0 eV.



Figure S11. Orbital-resolved band structures of (a, c, e, g) AB and (b, d, f, h) AC<sub>2</sub>' stacks with and without external electric field. The values of bandgaps for spin up and down are marked in the figures.



Figure S12. Decomposed charge density corresponding to the marked positions (Figure 4) for (a) AB and (b)  $AC_2$ ' stacks Cr-H without external electric field.



Figure S13. Electrostatic vacuum potential of (a) CrI<sub>3</sub>, (b) CrGeTe<sub>3</sub> and (c) Cr-H layers.



Figure S14. Electrically induced charge densities defined as  $\Delta \rho = \rho(\vec{E}) - \rho(0)$  of (a and b) AB and (c and d) AC<sub>2</sub>' stacks under external electric field  $\vec{E} = \pm 0.8 \text{ V/Å}$ . Yellow and blue areas represent electrons accumulation and depletion, respectively.



Figure S15. (a) Schematic structure of Cr-H based field effect transistor. (b) The gate voltage and (c) corresponding output spin-polarized current as a function of time.

#### Electric-field-dependent interlayer distance

In order to understand the effect of the external field on the Cr-H geometry, we make further discussion on the electrically-induced changes of the thickness of two monolayers, as well as their interlayer distance. Herein, the thickness of CrI<sub>3</sub> monolayer is changed in a small range (3.20-3.22 Å), while that of CrGeTe<sub>3</sub> is fluctuated between 3.56-3.58 Å. We believe such small distortion of monolayers would show tiny impacts on the magnetic and electronic properties of heterostructures. However, the interlayer distances of these two layers exhibit electric-field-dependent characters as shown in Figure s16. The interlayer distances are 3.48 and 3.50 Å for AB and AC<sub>2</sub>' stacks under the field of 0 eV/Å, respectively. Compared to AB stack, the increased interlayer distance of AC<sub>2</sub>' phase induces the positive total energy. Besides, for both two stacks, the interlayer distance is significantly increased under positive fields, while slightly increased under negative fields. It is worthy to note that the interlayer distance of AC<sub>2</sub>' stack is larger than AB stack with and without external fields, possibly leading to the more positive binding energy.



Figure S16. The optimized geometry of (a) AB and (b)  $AC_2$ ' stacks. The vdW interlayer distance are defined in the figure. (c) The changes of interlayer distance of AB and  $AC_2$ ' stacks with respect to the electric fields.

# Reference

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