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

Spin Gapless van der Waals Heterostructure for Spin Gating Through Magnetic Injection Device

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For the transition group metals, the typical U-values are generally about 3.0 eV, mostly between 2.0 and 4.0 eV. We have tested the Cr_2NOF structure with U values between 0 eV and 5 eV (Fig. S1), and it can be seen that the change in U value has little effect on the properties, and the system keeps its magnetic semiconductor properties. When U is greater than 3eV, the morphology of the energy band structure is almost constant, and based on this 3eV was chosen as the U value selected for this study.

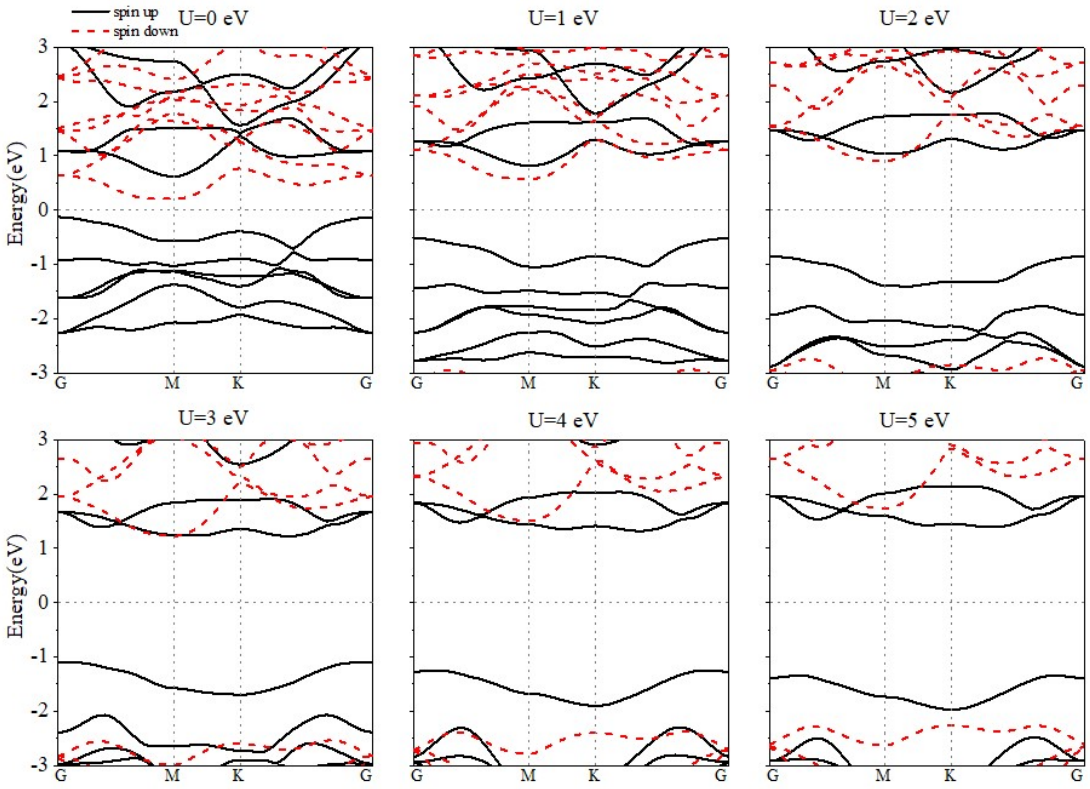

Fig. S1. Energy band structure of Cr_2NOF at different U values.

We compared the average binding energy per atom of Cr_2NOF with structures such as Nb_2C,[1] Ti_2C,[2] and V_2C.[3] For Cr_2NOF, the average binding energy per atom is -4.782 eV:

$$E_b = \frac{E_{Total} - 2E_{Cr} - E_N - E_O - E_F}{5} = -4.782\ eV$$

The binding energies of Nb_2C, Ti_2C, and V_2C were calculated to be -3.67 eV, -4.08 eV, and -5.09 eV, respectively, and the binding energy of Cr_2NOF lies between these synthesized 2D materials, indicating that it is stable.

Fig. S2 shows a schematic diagram of a lattice structure containing four different heterostructures, the mode of construction is the O-side and 2DH contact of Cr_2NOF.

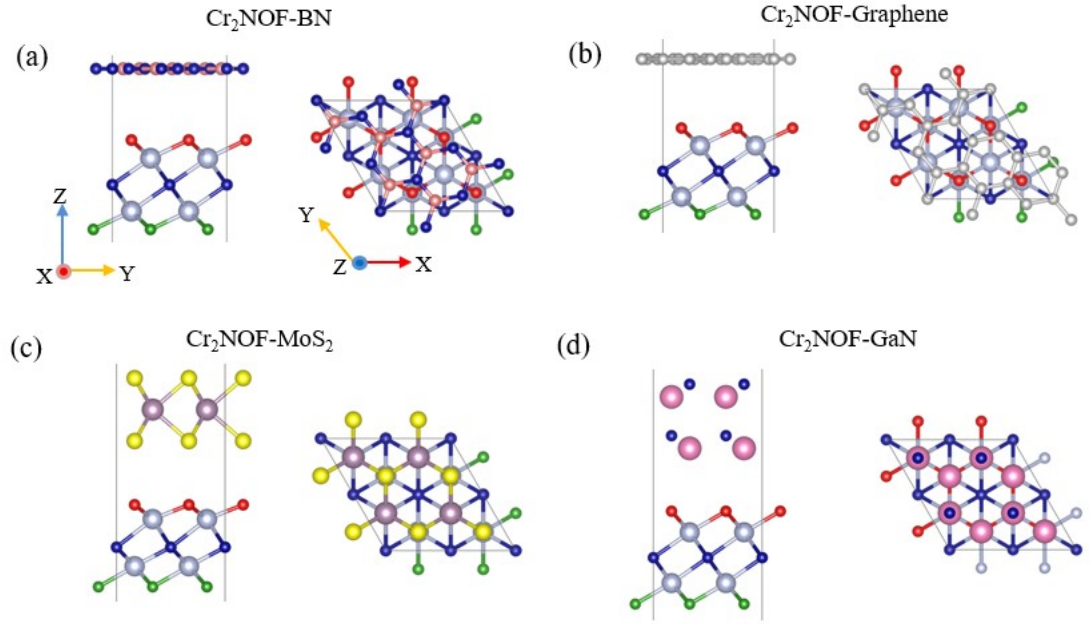

Fig. S2. Schematic illustration of the crystal structures of (a) Cr_2NFO/BN, (b) Cr_2NFO/Graphene, (c) Cr_2NFO/MoS_2, (d) Cr_2NFO/GaN vdW heterostructures.

Fig. S3 shows the work function of Cr_2NOF (F-side) contact with 2DH in heterojunction Cr_2NFO/2DH. Due to the high original work function of 6.64 eV at the O-side, the O-side is exposed on the outside in this construction, resulting in a high work function on the right side of the four heterojunctions.

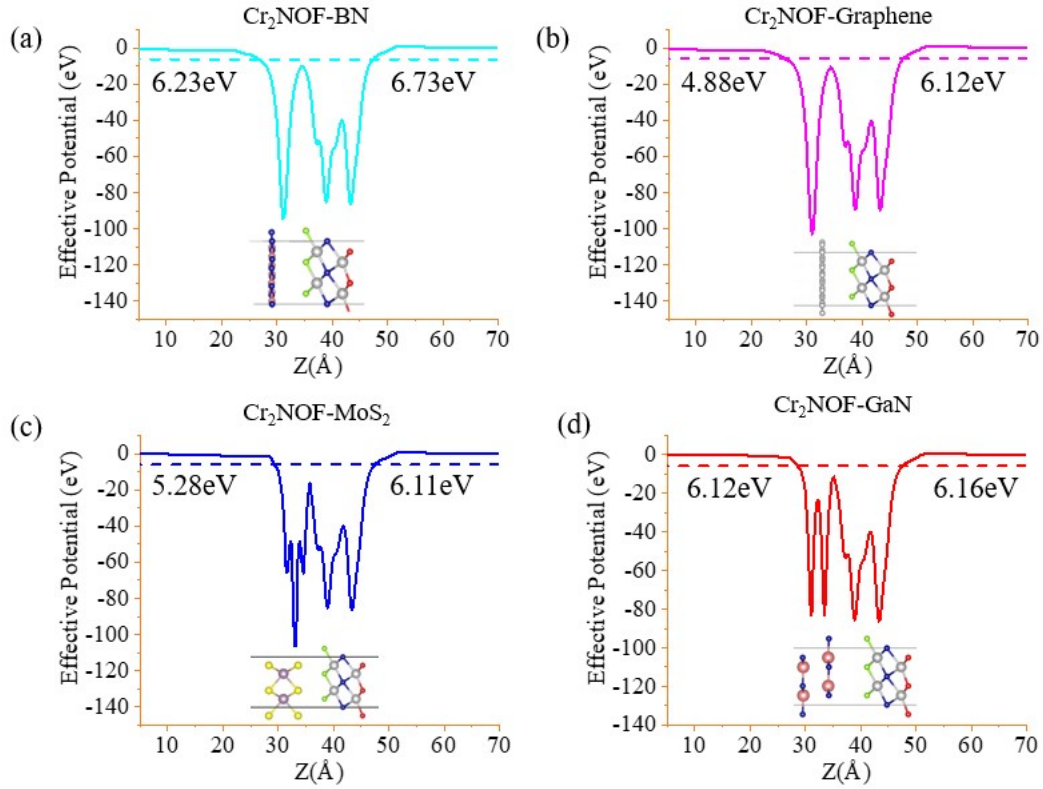

Fig. S3. The plane-averaged electrostatic potential difference of Cr_2NOF after the formation of (a) Cr_2NFO/BN, (b) Cr_2NFO/Graphene, (c) Cr_2NFO/MoS_2, (d) Cr_2NFO/GaN heterostructures. The inserts show the crystal structure of four heterojunctions in contact with the F-side.

We also calculate the fat band structures for the 2DH and F-side contact heterojunctions (Fig. S4). The BN contribution in Cr_2NFO/BN is biased towards deeper energy levels, with a larger band gap compared to the O-side contact case. The Dirac cone in Cr_2NFO/Graphene does not undergo a significant upward shift and remains near the Fermi level. There is no significant difference between the band structure of the Cr_2NOF/GaN heterojunction for the F-side contact and that of the O-side contact.

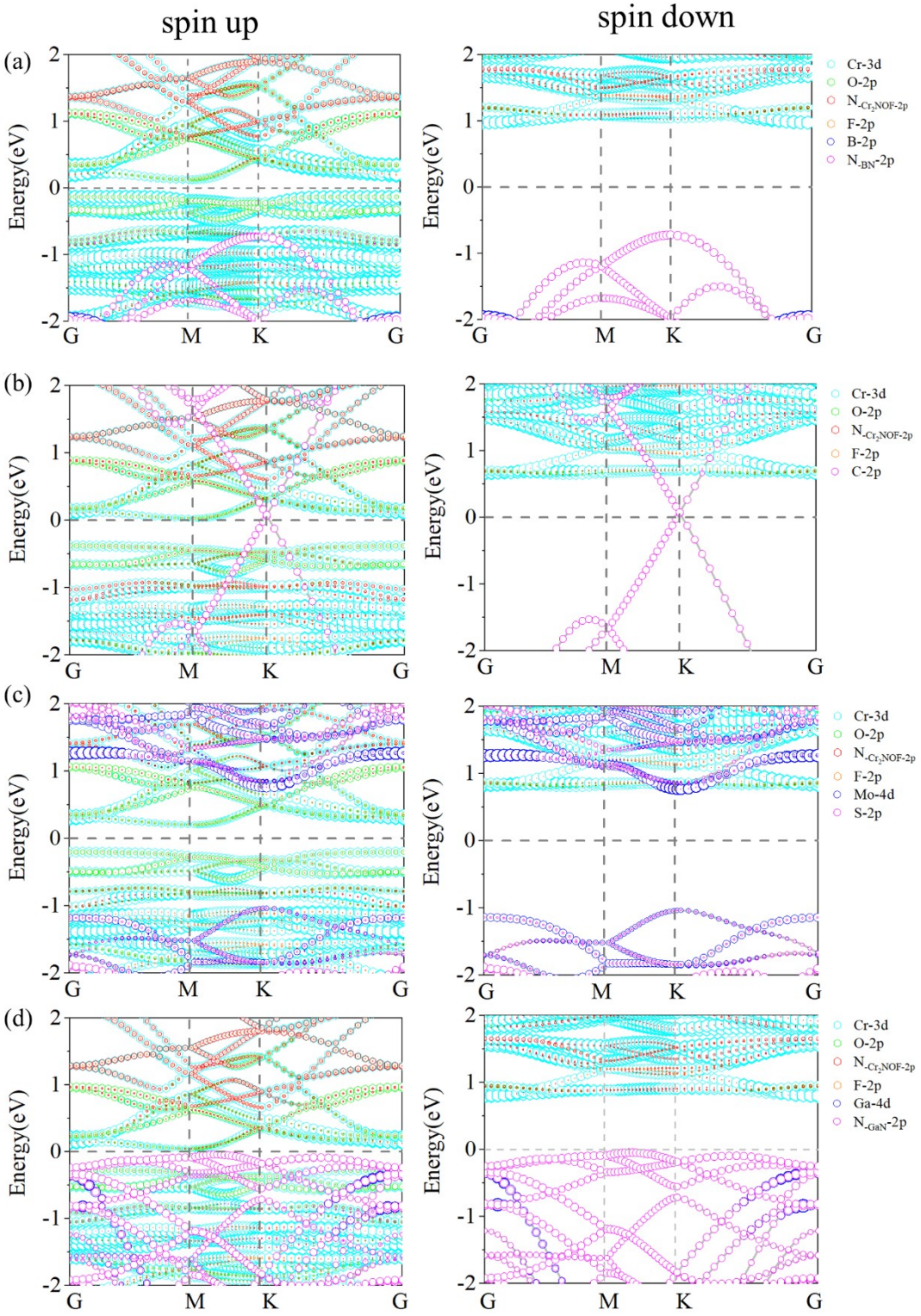

Fig. S4. The spin-polarized band structures of (a) Cr_2NOF/BN, (b) Cr_2NOF/Graphene, (c) Cr_2NOF/MoS_2, and (d) Cr_2NOF/GaN heterostructures. This is the case that 2DH and Cr_2NOF (F-side) contacts.

References

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