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

Supplementary Information

Magnetic phase transition regulated by interface coupling effect in $CrBr_3$/electride Ca_2N van der Waals heterostructures

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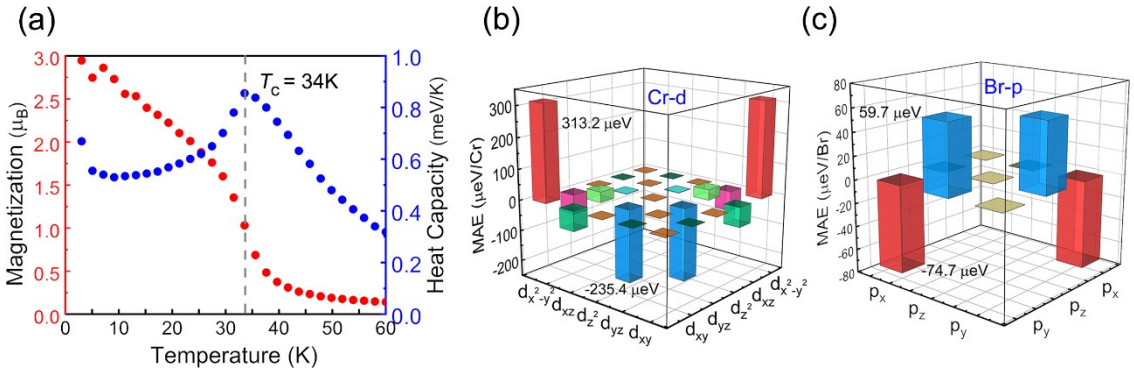

Fig. S1 (a) The magnetization and heat capacity as functions of temperature for $CrBr_3$ monolayer. Orbital-resolved MAE for the (b) Cr and (c) Br ion in the $CrBr_3$ monolayer.

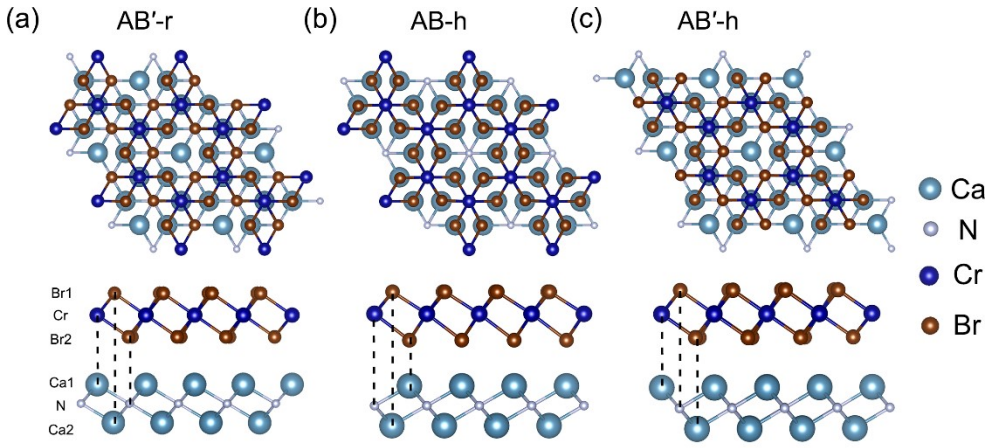

Fig. S2 The top and side views of $CrBr_3/Ca_2N$ heterostructure with different stacking orders: (a) AB'-r, (b) AB-h, and (c) AB'-h.

Table S1. The energy of FM and various AFM states with different stacking orders for the $CrBr_3/Ca_2N$ heterostructure. The light gray part indicates the most stable energy ground state under each structure.

Model	**AB-r**	**AB'-r**	**AB-h**	**AB'-h**
E_{FM} **(eV)**	–320.6	–318.4	–312.2	–320.6
$E_{Neel\text{-}AFM}$ **(eV)**	–321.0	–319.2	–311.6	–319.5
$E_{zigzag\text{-}AFM}$ **(eV)**	–316.2	–319.3	–312.7	–311.8
$E_{stripy\text{-}AFM}$ **(eV)**	–311.1	–309.9	–300.4	–308.0

Table S2. The binding energies (E_b) of the $CrBr_3/Ca_2N$ heterostructures in the most stable ground state under different stacking orders.

	AB-r (Neel-AFM)	**AB'-r** (zigzag-AFM)	**AB-h** (zigzag-AFM)	**AB'-h** (FM)
E_b **(eV/atom)**	–0.474	–0.425	–0.246	–0.457

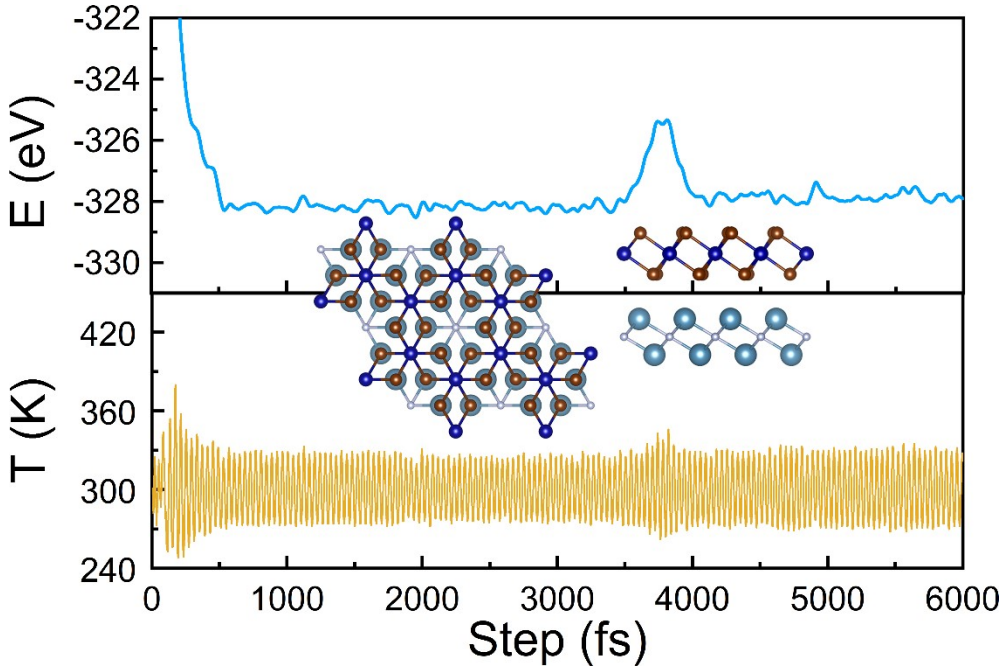

Fig. S3 The fluctuations of energy and temperature as well as the final configuration of the AB-r heterostructure are obtained from AIMD simulation.

Table S3. The exchange coupling parameters (J_1, J_2), Atomic Magnetic Moments (M_{Cr}), Curie temperature (T_C), Magnetic Anisotropy Energy (MAE), and Energy Difference between FM and AFM Spin Ordering ($\Delta E_{FM\text{-}AFM}$) of the 2×2×1 supercell for the $CrBr_3$ with the electron/hole injection (Q_e).

Q_e (e/u.c)	J_1 (meV)	J_2 (meV)	M_{Cr} (μ_B)	T_C (K)	MAE (μeV/Cr)	$\Delta E_{FM\text{-}AFM}$ (meV)
1	**28.66**	**−1.02**	**3.355**	**32**	**375**	**−68**
0.75	**26.75**	**0.37**	**3.327**	**34**	**282**	**−78**
0.5	**24.83**	**1.38**	**3.299**	**35**	**166**	**−84**
0.25	**23.14**	**2.00**	**3.271**	**36**	**26**	**−86**
0	**21.73**	**2.26**	**3.243**	**34**	**−156**	**−85**
−0.25	**24.97**	**3.41**	**3.239**	**46**	**−1172**	**−107**
−0.5	**28.14**	**4.74**	**3.233**	**55**	**−1788**	**−130**
−0.75	**31.47**	**6.14**	**3.23**	**66**	**−2177**	**−155**
−1	**34.63**	**7.63**	**3.225**	**75**	**−2464**	**−180**

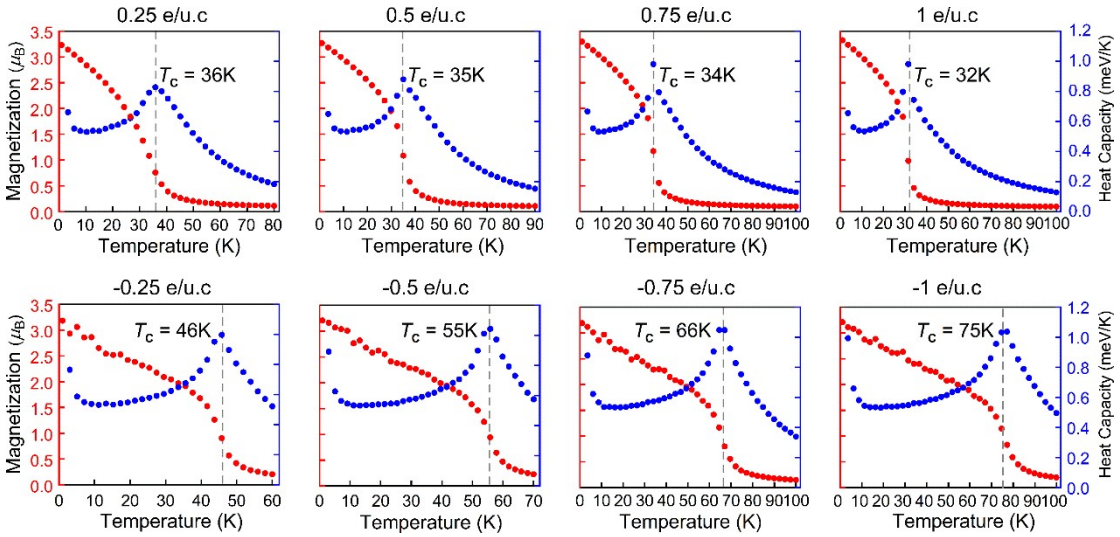

Fig. S4 The magnetization and heat capacity as functions of temperature for $CrBr_3$ monolayer with charge doping.