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

Supplementary Information

Bilayer hexagonal structure MnN_2 nanosheet with room-temperature ferromagnetic half-metal behavior and tunable electronic structure

Yuan Gao, Honggang Pan,* Baozeng Zhou*

Tianjin Key Laboratory of Film Electronic & Communicate Devices, School of Integrated Circuit Science and Engineering, Tianjin University of Technology, Tianjin 300384, China

*Corresponding Authors

phg022@163.com (H. Pan)

baozeng@tju.edu.cn (B. Zhou)

Table S1. Elastic constants of BHS-MnN_2, BHS-MnP_2 and BHS-$MnAs_2$.

	Elastic Constants (N/m)					Young's Modulus (N/m)	Shear Modulus (N/m)	Poisson's Ratio
	C_{11}	C_{22}	C_{12}	C_{66}	C_{11}-$\|C_{12}\|$	*x* or *y*		*x* or *y*
MnN_2	190.28	190.28	64.37	62.96	＞0	168.51	62.96	0.34
MnP_2	102.62	102.62	31.16	35.73	＞0	93.15	35.73	0.30
$MnAs_2$	87.21	87.21	17.82	34.69	＞0	83.56	34.69	0.20

Table S2. Energy difference between antiferromagnetic and ferromagnetic states of MnN_2, MnP_2 and $MnAs_2$.

System	ΔE_{FM}(eV)	ΔE_{AFM1}(eV)	ΔE_{AFM2}(eV)	ΔE_{AFM3}(eV)	ΔE_{AFM4}(eV)	ΔE_{AFM5}(eV)
MnN_2	0	8.12	6.71	4.68	6.24	6.34
MnP_2	0	3.65	3.30	2.07	2.91	2.58
$MnAs_2$	0	6.13	4.48	2.62	4.47	4.07

Table S3. Localized magnetic moment (M, μ_B), exchange parameters J_1 and J_2, magnetic anisotropic energy and calculated Curie temperature of BHS-MnN_2, BHS-MnP_2 and BHS-$MnAs_2$.

Substance	$M_{Mn}(\mu_B)$	$M_{N、P、As}(\mu_B)$	$M_{Tot}(\mu_B)$	J_1(eV)	J_2(eV)	$E_{MAE}(\mu$eV)	T_C(K)
BHS-MnN_2	2.43	-0.55	5.23	0.169	0.419	3.67	563
BHS-MnP_2	2.84	-0.38	8.28	0.076	0.206	8.27	705
BHS-$MnAs_2$	3.37	-0.42	10.19	0.088	0.194	41.58	1122

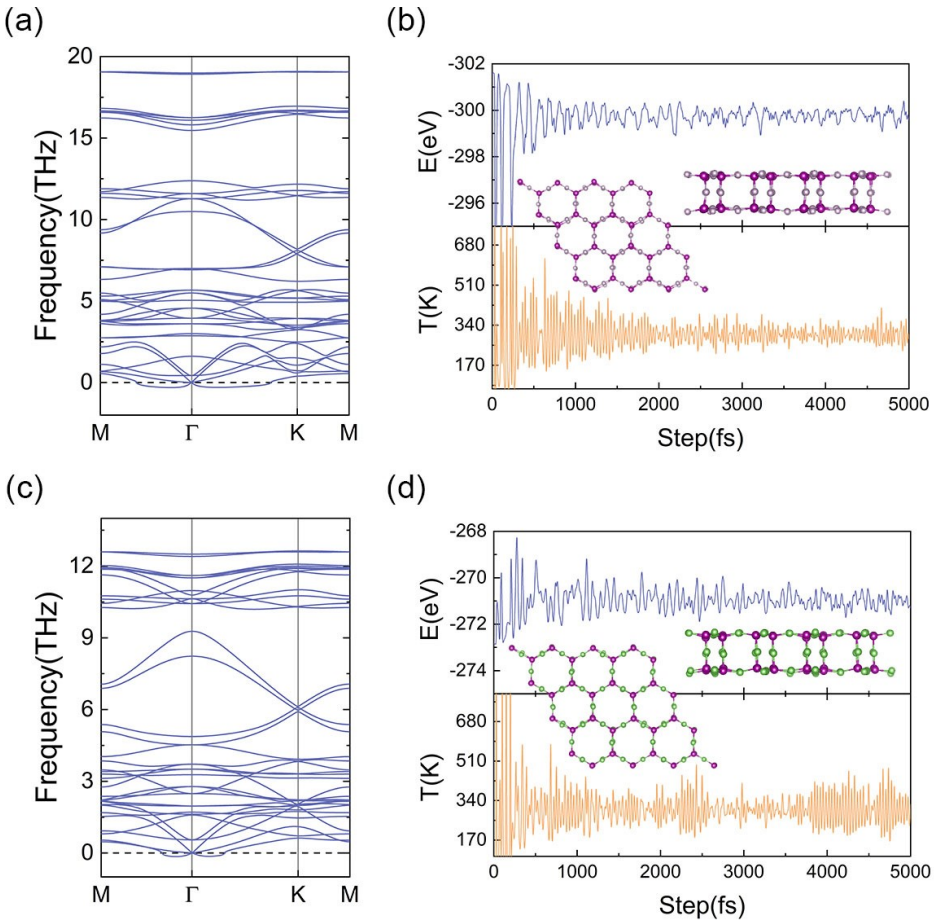

Fig. S1 (a) Phonon dispersions of MnP_2 nanosheet, (b) The fluctuations of energy and temperature as well as the final configuration of MnP_2 are obtained from AIMD simulation, (c) Phonon dispersions of $MnAs_2$ nanosheet, (d) The fluctuations of energy and temperature as well as the final configuration of $MnAs_2$ are obtained from AIMD simulation.

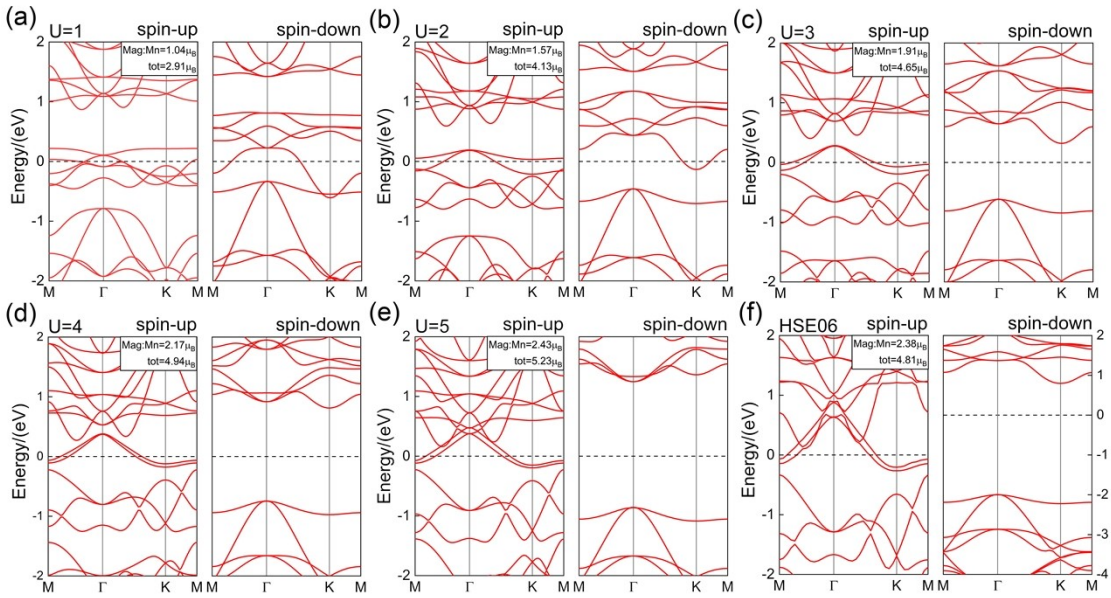

Fig. S2 (a)-(e) Band structures of BHS-MnN_2 nanosheet calculated by PBE+*U* (U_{eff}=1, 2, 3, 4, 5), and (f) Band structure of BHS-MnN_2 calculated by HSE method.

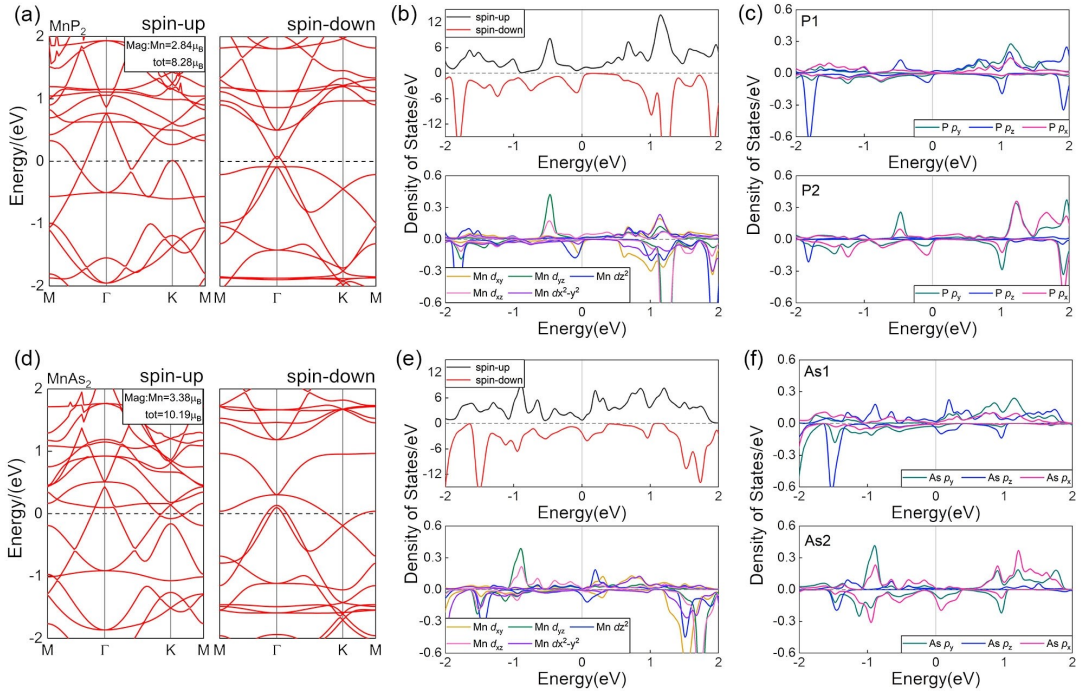

Fig. S3 (a) BHS-MnP_2 spin polarization band structure. (b) Calculate the total DOS of BHS-MnP_2 and the *d*-orbital projection DOS of Mn with U_{eff}=5 eV. (c) *p*-orbital projection DOS for different coordination P. (d) BHS-$MnAs_2$ polarization band structure. (e) Calculate total DOS for BHS-$MnAs_2$ and *d*-orbital projection DOS for Mn with U_{eff}=5 eV. (f) *p*-orbital projection DOS for different coordination As.

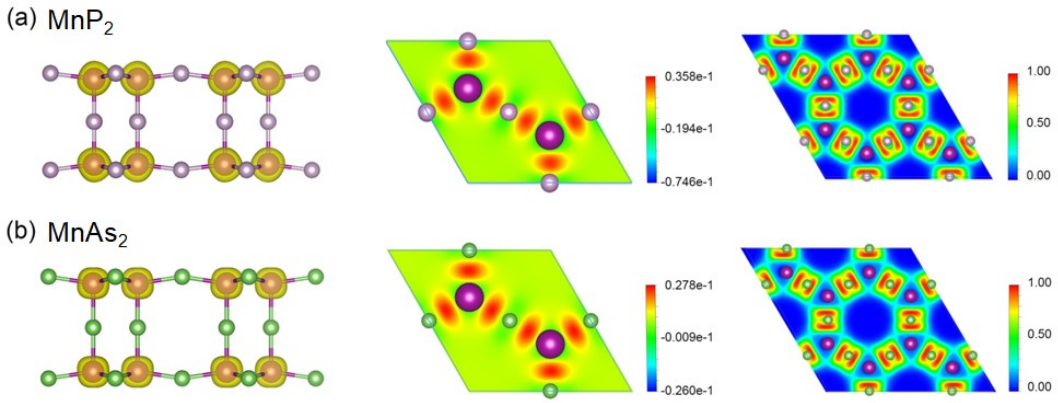

Fig. S4 (a) The spin density distribution, deformed charge density and ELF of BHS-MnP_2. (b) The spin density distribution, deformed charge density and ELF of BHS-$MnAs_2$.

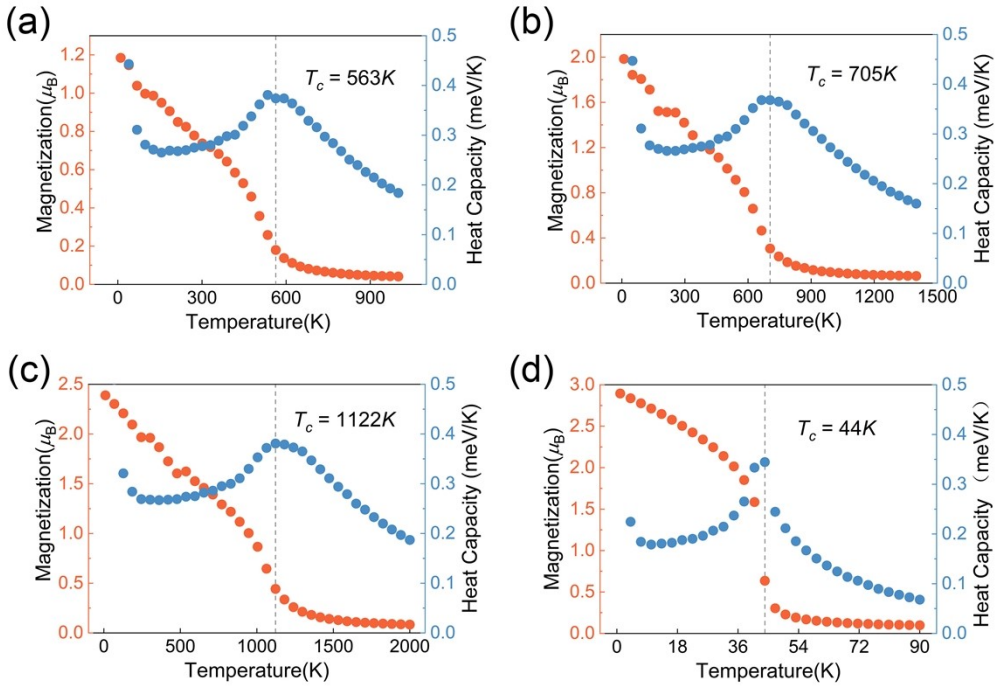

Fig. S5 Magnetic moment per unit cell (red) and specific heat (Cv) (blue) of (a) BHS-MnN_2, (b) BHS-MnP_2, (c) BHS-$MnAs_2$, (d) CrI_3.

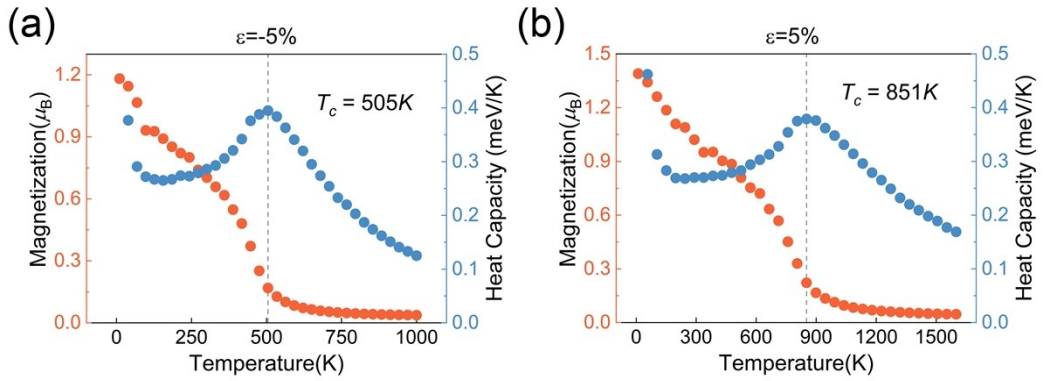

Fig. S6 Variations of magnetic moment and heat capacity of MnN_2 with temperature under (a) 5% and (b) -5% strain.