Electronic Supplementary Information for

Theoretical prediction of two-dimensional ferromagnetic Mn_2X_2 (X=As, Sb) with strain-controlled magnetocrystalline anisotropy

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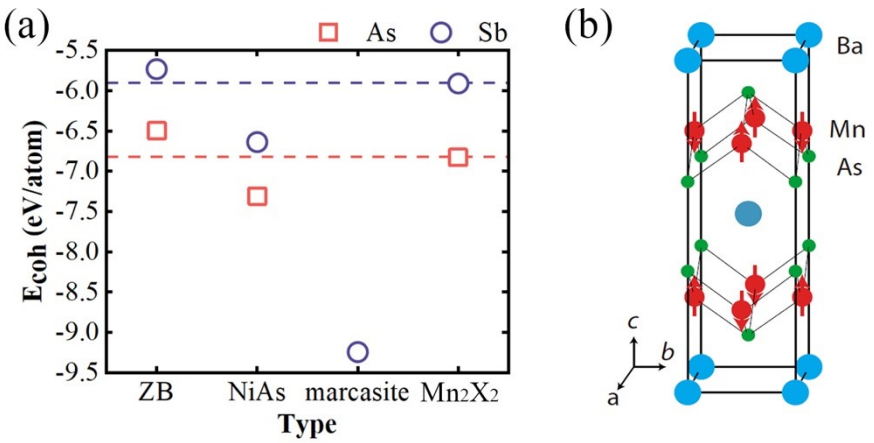

Figure S1. (a) Cohesive energy of different structures. The predicted Mn_2X_2 in the tetragonal structures are $BaMn_2As_2$ type. (b) The crystallographic structure of synthesized $BaMn_2As_2$.[40]

Table S1. The calculated magnetic moments of Mn (μ_B) for Mn_2X_2 (X = As, Sb) with HSE06 and GGA+U (U_{eff} = 3.5 eV) level, respectively.

Method	Mn_2As_2	Mn_2Sb_2
HSE06	3.711	4.103
GGA+U	3.681	4.095

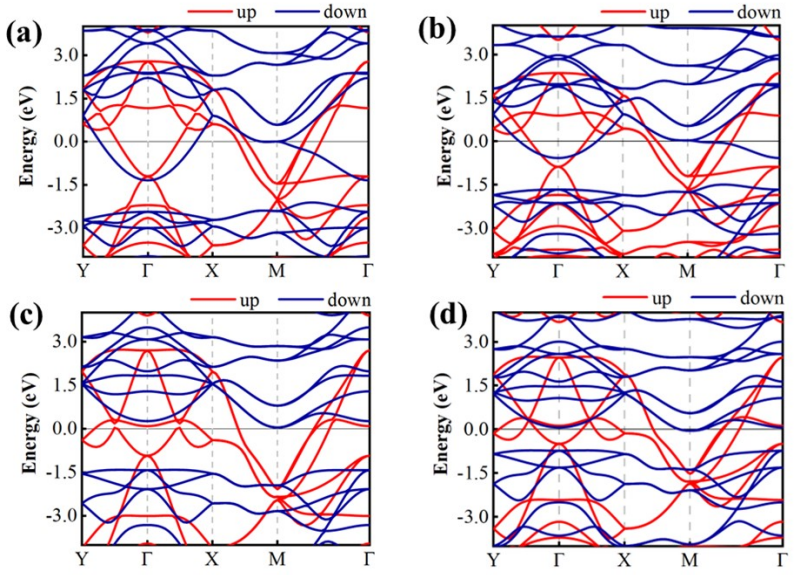

Figure S2. The band structures of the (a) Mn_2As_2 and (c) Mn_2Sb_2 using HSE06 functional and those of the (b) Mn_2As_2 and (d) Mn_2Sb_2 with GGA+U (U_{eff} = 3.5 eV) method.

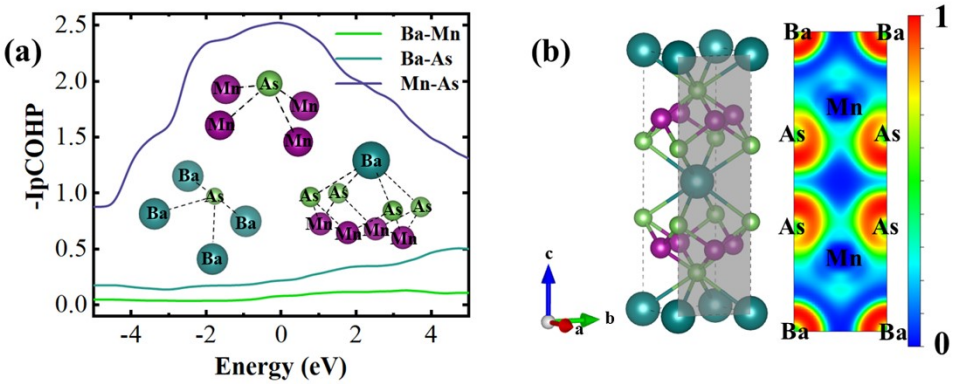

Figure S3. (a) Integrated projected crystal orbital Hamilton population for Ba-As, Ba-Mn and Mn-As bonds with the corresponding structure schematics. (b) Crystal structure and projection of the electron localization function along (100) plane of $BaMn_2As_2$.

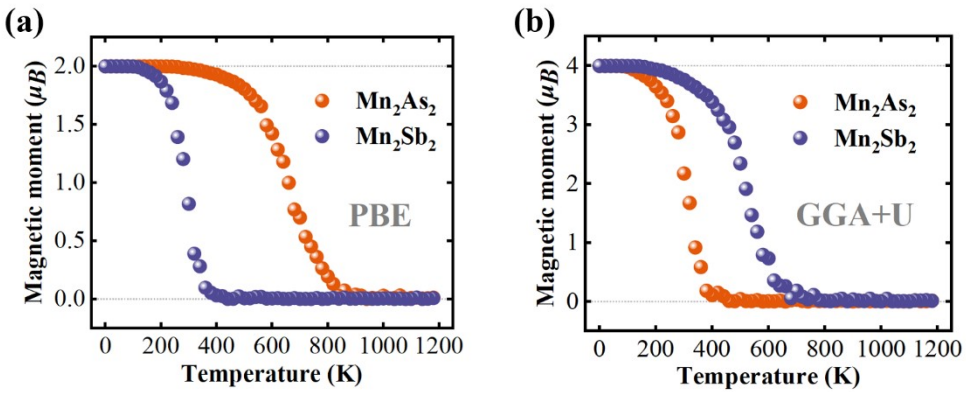

Figure S4. Variation of the Mn magnetic moment with respect to the temperature for the 2D Mn_2X_2 in (a) PBE and (b) GGA+*U* level.