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

Supplemental Information for "Spontaneous magnetic merons in half-metallic $Mn_2I_3Br_3$ monolayer with easy-plane anisotropy"

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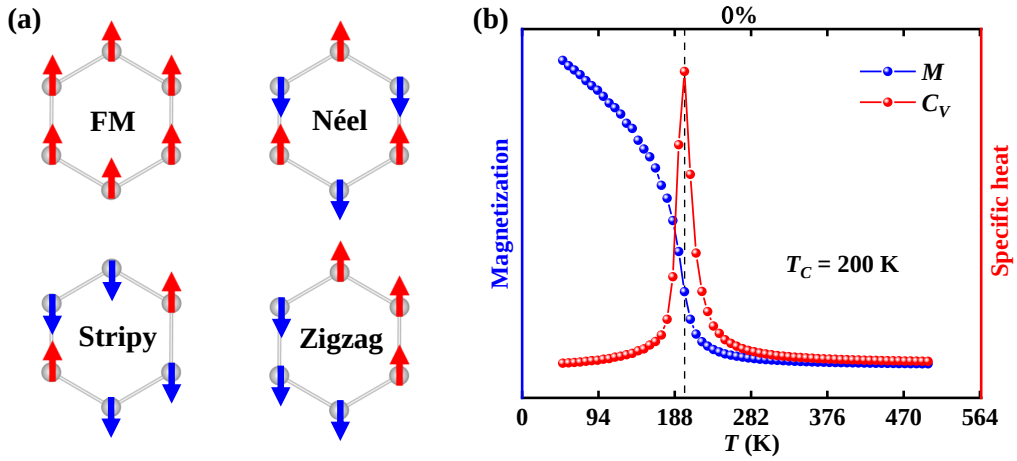

FIG. S1. (a) Schematic diagram of the four spin configurations used to extract the Heisenberg exchange coefficients J_1, J_2 and J_3. Red and blue arrows represent spin up (Z-direction) and down (−Z-direction), respectively. (b) Magnetic moment and specific heat capacity versus temperature of $Mn_2I_3Br_3$ monolayer.

To obtain the Heisenberg exchange coefficients J_1, J_2 and J_3, we choose four spin configurations named FM, $Néel$, $Stripy$ and $Zigzag$ as shown in Fig. S1(a), with the energies being:

$$E_{FM} = -6J_1 - 12J_2 - 6J_3 + 4K(S^z)^2 + E_0, \tag{S1}$$

$$E_{Néel} = 6J_1 - 12J_2 + 6J_3 + 4K(S^z)^2 + E_0, \tag{S2}$$

$$E_{Stripy} = 2J_1 + 4J_2 - 6J_3 + 4K(S^z)^2 + E_0, \tag{S3}$$

$$E_{Zigzag} = -2J_1 + 4J_2 + 6J_3 + 4K(S^z)^2 + E_0, \tag{S4}$$

where J_1, J_2, J_3 are the Heisenberg exchange coefficients of first, second and third nearest neighbor, respectively. K is the single ion anisotropy. S^z is the off-plane component of the normalized spin and E_0 is the spin-independent energy. When the magnetic parameters are obtained, we perform Monte Carlo simulations with the Metropolis algorithm to evaluate the T_C of $Mn_2I_3Br_3$ monolayer. As shown in Fig. S1(b), the T_C of intrinsic $Mn_2I_3Br_3$ monolayer is about 200 K. The T_C with different strains can be found in Fig. S2

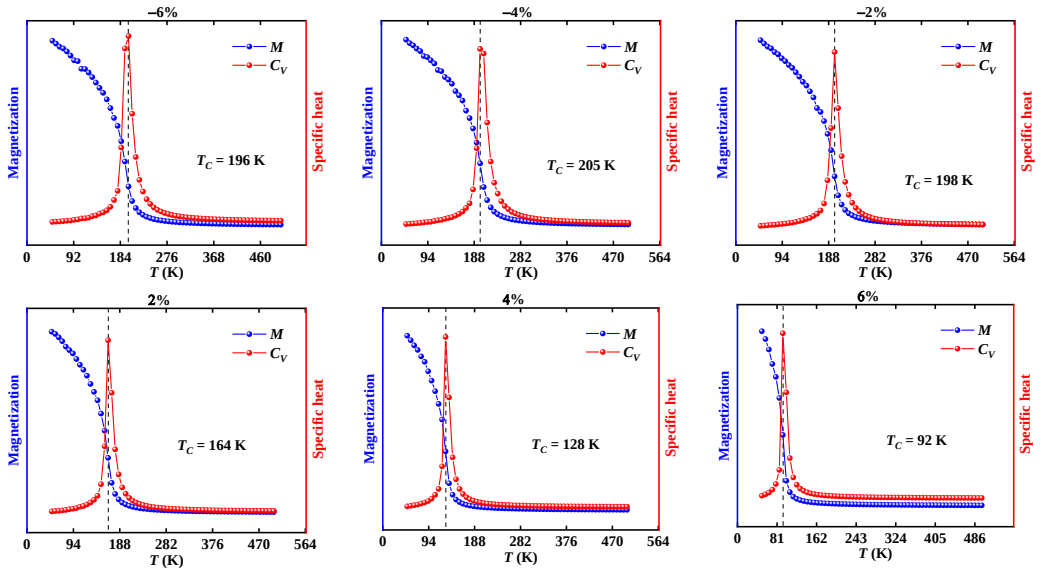

FIG. S2. Magnetic moment and specific heat capacity versus temperature of $Mn_2I_3Br_3$ monolayer with different biaxial strains.

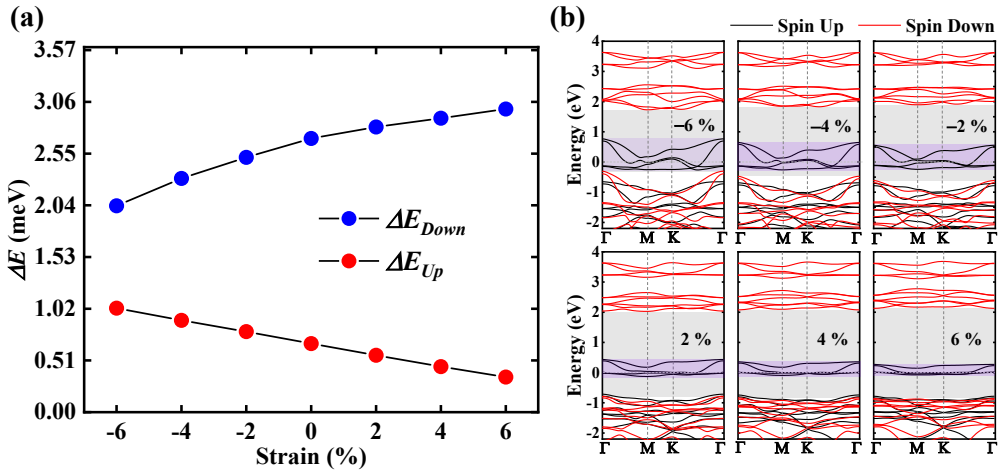

FIG. S3. (a) The band gap of spin down electrons (ΔE_{Down}) and band width of spin up electrons around fermi level (ΔE_{Up}), and (b) band structures of $Mn_2I_3Br_3$ with different strains.

As shown in Fig. S3(a), we illustrate the band gap of spin down electrons (ΔE_{Down}) and band width of spin up electrons around Fermi level (ΔE_{Up}) as functions of strains in $Mn_2I_3Br_3$ monolayer. The band structures of $Mn_2I_3Br_3$ with different strains are illustrated in Fig. S3(b) and ΔE_{Up} (ΔE_{Down}) is marked by the purple (gray) shaded areas. It's clearly

seen that the half-metallic feature can be further enhanced by tensile strain due to the more localized spin-up channel around Fermi level and larger band gap of the spin-down channel.

TABLE S1. The optimized lattice constant a, Heisenberg exchange coefficients (J_1, J_2, J_3), in-plane ($d_{//}$) and off-plane (d_z) components of DMI, single ion anisotropy (K), the magnetic moments of Mn atoms and the T_C of $Mn_2I_3Br_3$ monolayer with different strains.

Strain (%)	−6	−4	−2	0	2	4	6
a (Å)	6.24	6.38	6.51	6.64	6.77	6.91	7.04
J_1 (meV)	−13.37	−16.16	−15.42	−14.28	−8.40	−4.57	−1.68
J_2 (meV)	−1.05	−1.36	−2.10	−2.60	−3.46	−3.38	−3.04
J_3 (meV)	−7.43	−5.74	−4.33	−3.85	−2.23	−1.51	−0.98
$d_{//}$ (meV)	1.68	1.91	2.38	3.00	3.71	3.99	3.83
d_z (meV)	1.07	1.20	1.46	1.81	2.19	2.31	2.18
K (meV)	−13.15	−12.47	−10.88	−9.95	−10.02	−9.33	−7.75
M_{Mn} (μ_B)	4.00	4.01	4.01	4.02	4.04	4.07	4.12
T_C (K)	196	205	198	200	164	128	93