Supplementary Material

Giant valley splitting in the $MoTe_2/MnSe_2$ van der Waals heterostructure with room-temperature ferromagnetism

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1. Band structure and spin-polarized density of states of monolayer $MnSe_2$.

Fig. S1 shows the band structure and spin-polarized density of states (DOS) of $MnSe_2$. The asymmetry of the spin-up and spin-down states indicates that it is the ferromagnetic, and the fermi-level crosses the conduction bands, thus the $MnSe_2$ monolayer is a ferromagnetic metal. It is found that the total magnetic moments for per unit cell is 3.25 μ_B, and the magnetic moments of each Mn and Se atoms are 3.99 and -0.37 μ_B, respectively, so the Mn and Se are antiferromagnetic coupled. The bands near Fermi level are almost composed by Mn-*d* orbitals and Se-*p* orbitals, and the conduction band edge at K/K' valleys is mainly composed of Mn-d_{xz} and Se-p_z.

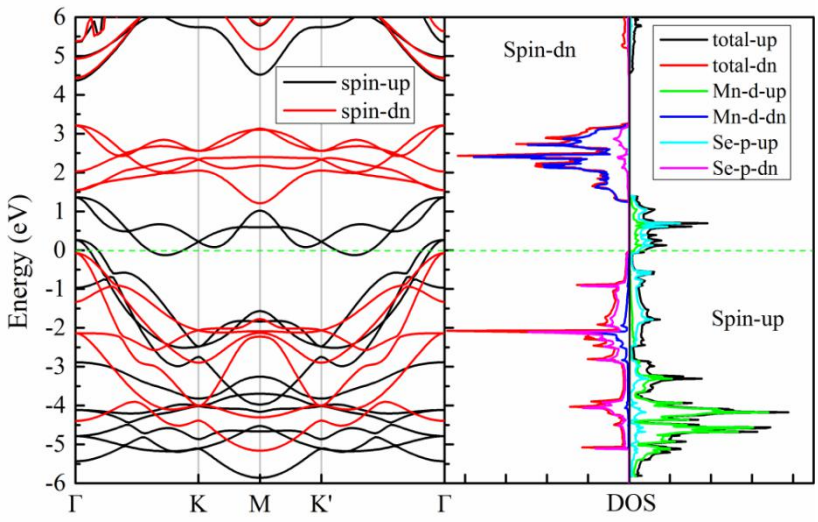

Fig. S1. Band structure and spin- polarized density of states of monolayer $MnSe_2$

2. Comparison of valley splitting and Curie temperature of the $MoTe_2/MnSe_2$ van der Waals heterostructure with other typical heterostructures.

TABLE S1. Comparison of intrinsic Curie temperature (K) and size of valley splitting (meV) of the $MoTe_2/MnSe_2$ heterobilayer with several typical heterostructures reported by Refs.

Systems	Curie temperature (K)	Valley splitting (meV)
$MoTe_2$/EuO [Ref. 16]	69	44
WS_2/MnO(111) [Ref. 18]	118	214
MoS_2/CoO(111) [Ref. 40]	298	103
WSe_2/CrI_3 [Refs. 20, 21]	45	3
MoS_2/h-VN [Ref. 23]	768	376
MoS_2/Cr_2O_3 [Ref. 41]	307	24
$MoTe_2/NiCl_2$ [Ref. 22]	120	32
$WSe_2/CrSnSe_3$ [Ref. 42]	73	9
$MoTe_2/MnSe_2$	**266**	**116**

3. Interfacial charge transfer in $MoTe_2/MnSe_2$ vdW heterostructure in six stackings.

As shown in Fig. S2, since the electronegativity of Se is larger than that of Te, the interlayer charge transfer is from the monolayer $MoTe_2$ to the $MnSe_2$ layer in the heterobilayer. Noted that although the $MnSe_2$ is a ferromagnetic metal, it forms a vdW heterostructure with the monolayers $MoTe_2$, so the charge transfer between the layers is a kind of spacial charge transfer rather than a chemical bond transfer, so the charge transfer between layers is not strong.

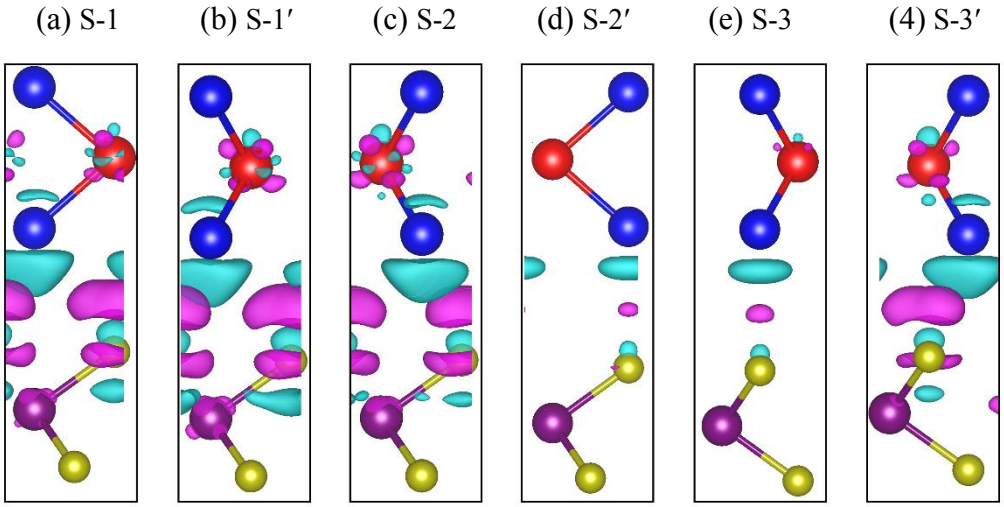

Fig. S2. Interfacial charge transfer and charge redistribution of $MoTe_2/MnSe_2$ in six stackings. The isovalue chosen to plot the isosurfaces is 0.0004 $eÅ^{-3}$. Losses and gains of electrons are colored aqua and pink, respectively.

4. Modulations of Curie temperature of monolayer $MnSe_2$.

The Curie temperature of ferromagnetic system is realized in open-source mcsolver,[1] and which is based on classical Ising model and Wolf algorithm to accurately describe the equilibrium magnetic system at various temperatures. In the calculations, 80000 iterations were used at each temperature to fully thermalize the system to reach equilibrium, and the output results were calculated from 640000 iterations after thermal equilibrium. In the Monte Carlo (MC) simulations, we use a (32×32) super cell to reduce the periodic constraints. The exchange parameter (J) was defined as $J = (E_{FM} - E_{AFM})/4N\mu_{unit}^2$, where E_{FM} and E_{AFM} are the total energy of the

ground state of ferromagnetic (FM) and antiferromagnetic (AFM) respectively, N is the number of Mn atoms in the supercell, and μ_{unit} is the magnetic moment of the primary cell. The exchange energy of per unit cell is $E_{ex} = (E_{FM}-E_{AFM})/N$. The total energy of monolayer $MnSe_2$ was obtained by DFT calculations with (2x2) supercell, and the exchange parameter is -2.63 meV at zero strain. The magnetic moment direction of the two Mn atoms in the supercell is reversed as an antiferromagnetic system. TABLE S1 lists the calculated Curie temperature (Tc) for $MnSe_2$ under different biaxial strains. Fig. S3 shows the corresponding magnetic moment (red) and heat capacity (blue) of $MnSe_2$ changes with the temperature under several different biaxial strains.

TABLE S2. Calculated total energy of AFM state (E_{AFM}) and FM state (E_{FM}), exchange energy per unit cell (E_{ex}), exchange parameter (J), and Curie temperature (T_c) for $MnSe_2$.

Strains	E_{FM}(eV)	E_{AFM}(eV)	E_{ex}(meV)	J(meV)	T_c(K)
-5%	-60.50	-60.37	-32.11	-0.89	86
-4%	-60.60	-60.41	-46.81	-1.30	125
-3%	-60.67	-60.44	-57.85	-1.61	154
-2%	-60.73	-60.45	-69.33	-1.93	184
-1%	-60.77	-60.44	-83.75	-2.33	222
0	-60.80	-60.40	-99.88	-2.63	266
1%	-60.80	-60.34	-115.31	-3.20	306
2%	-60.78	-60.27	-129.06	-3.58	342
2.3%	-60.77	-60.24	-132.94	-3.69	353
3%	-60.74	-60.17	-141.64	-3.93	377
4%	-60.68	-60.07	--154.04	-4.27	409
5%	-60.61	-59.95	-166.2	-4.62	443

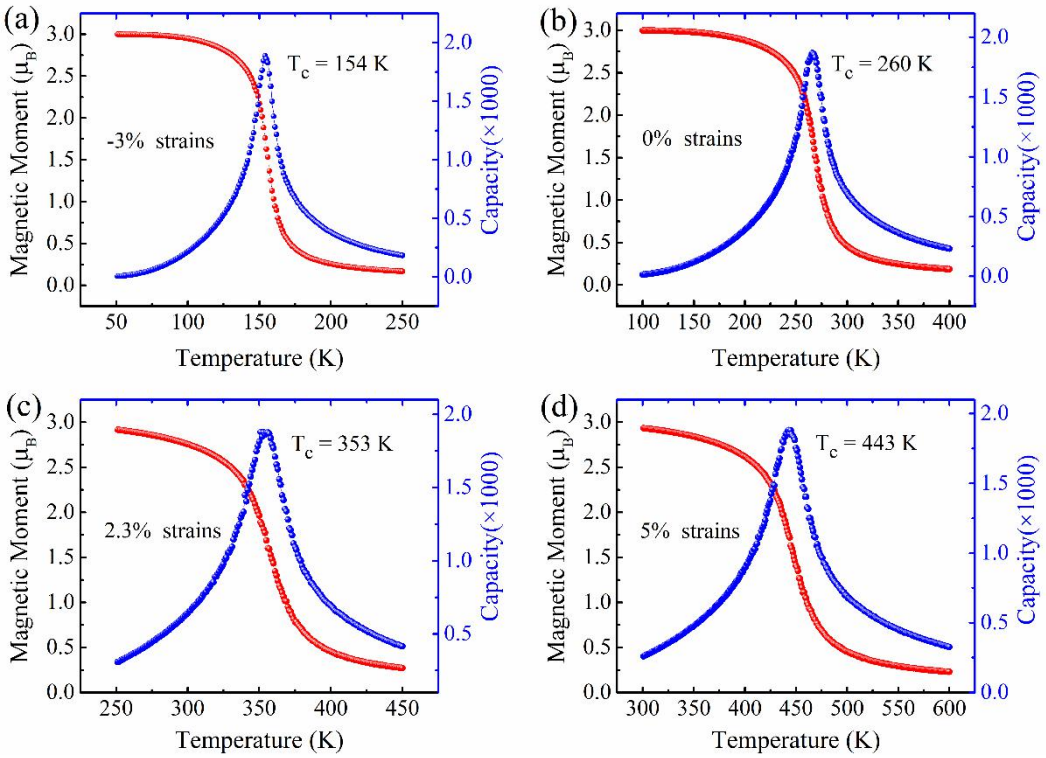

Fig. S3. The magnetic moment (red) and heat capacity (blue) changes with the temperature under (a) -3% biaxial compressive strains, (b) 0% strains, (c) 2.3% biaxial tensile strains, and (d) 5% biaxial tensile strains, respectively.

Reference

[1] L. Liu, X. Zhang, https://github.com/golddoushi/mcsolver.