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

Supplemental Material for

Enhanced Curie temperature and conductivity of van der Waals ferromagnet MgV_2S_4 via electrostatic doping

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Table S1. The lattice constants (a), layer thicknesses (L), corresponding energies (E), and symmetrical property for four different atomic structures. The distance between Mg atom and sulfur atom, vertically arranged sulfur atoms and vanadium atoms, and bent vanadium atoms and sulfur atoms are d_1, d_2, and d_3, respectively.

	a	b	c	d
a (Å)	3.73	3.73	3.76	3.77
L (Å)	9.12	9.16	8.87	8.78
E (eV)	-40.90	-40.89	-41.32	-41.33
d_1 (Å)	2.61	2.61	2.56	2.56
d_2 (Å)	2.23	2.23	2.23	2.22
d_3 (Å)	2.31	2.32	2.33	2.33
Point group	D_{3h}	D_{3h}	C_{3v}	C_{3v}

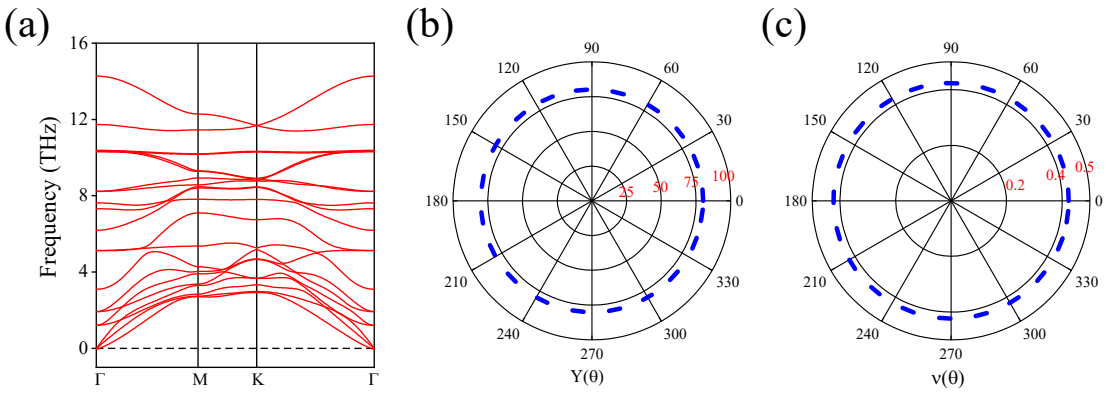

Figure S1. (a) The phonon spectrum, (b) Young's modulus Y(θ), and (c) Poisson's ratio ν(θ) of MgV_2S_4 monolayer.

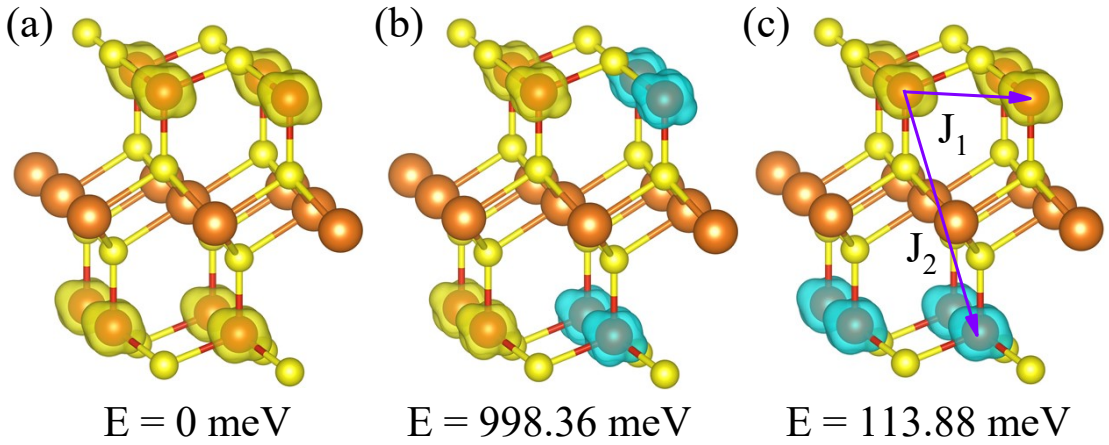

Figure S2. Three different magnetic configurations: (a) FM, (b) AFM-1, (c) AFM-2

are considered in the calculation and the corrosponding energy relative to the FM configuration. The yellow and cyan areas represent the spin up and spin down respectively.

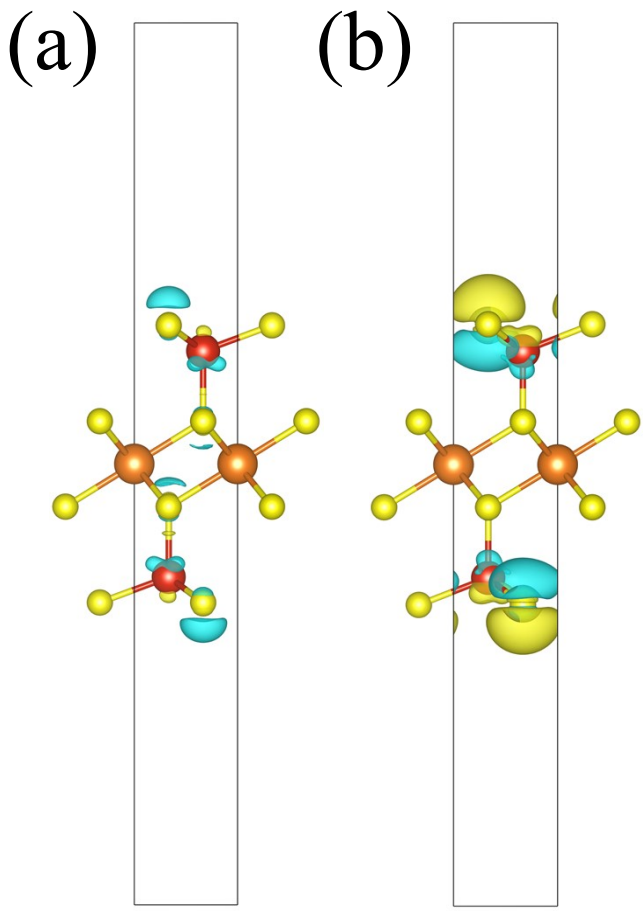

Figure S3. Charge density difference of the MgV_2S_4 monolayer with (a) 0.2 holes and (b) 0.2 electrons doping at an iso-surface of 0.002 e/Å^3, respectively. Yellow and blue represent charge accumulation and depletion, respectively.

According to the formula of MAE, the energy level difference between occupied and unoccupied states near the Fermi level and spin-orbit coupling matrix element determines MAE. In addition，the total MAE may also be influenced by the intensity of DOS because it is the integral of all occupied and unoccupied states. It can be seen from the Fig. S4(a) that the states near the Fermi level are mainly occupied spin up states and unoccupied spin up states d_{xy} ($d_{x^2-y^2}$) and d_{yz} (d_{xz}). For 0.2 electrons doping, the unoccupied spin up states move towards the Fermi level, while the occupied spin up states are far away from the Fermi level, making the electronic states near the

Fermi level smaller than that undoped case. Thus the contribution of the (d_{xy}, $d_{x^2-y^2}$) and (d_{yz} , d_{xz}) orbitals to MAE decreases. When 0.2 holes are doped, the unoccupied spin up states basically do not move, but the occupied spin up states moves towards and passes through the Fermi level. The occupied states near the Fermi level increase and the energy level difference decreases compared with that without doping, so the contribution of (d_{xy}, $d_{x^2-y^2}$) and (d_{yz} , d_{xz}) orbitals to MAE increases.

The states near the Fermi level are mainly occupied spin up (down) states and unoccupied spin up states p_x(p_y) as seen from the Fig. S4(b). For 0.2 electron doping, the energy difference between the occupied spin up states and the spin down states is reduced. The difference between the two denominators (the energy difference between the occupied spin up (down) states and the unoccupied spin up states) also decreases when states are integrated superposition. The spin orbit coupling matrix elements of different occupied spin states and unoccupied spin up states are opposite, which means that the molecules of the two fractions are opposite to each other. So the contribution of (p_x,p_y) orbitals to MAE decreases. The unoccupied spin up states and the occupied spin up and down states move towards the Fermi level during 0.2 hole doping, which increases the electronic states near the Fermi level, so the contribution of (p_x,p_y) orbitals to MAE increases.

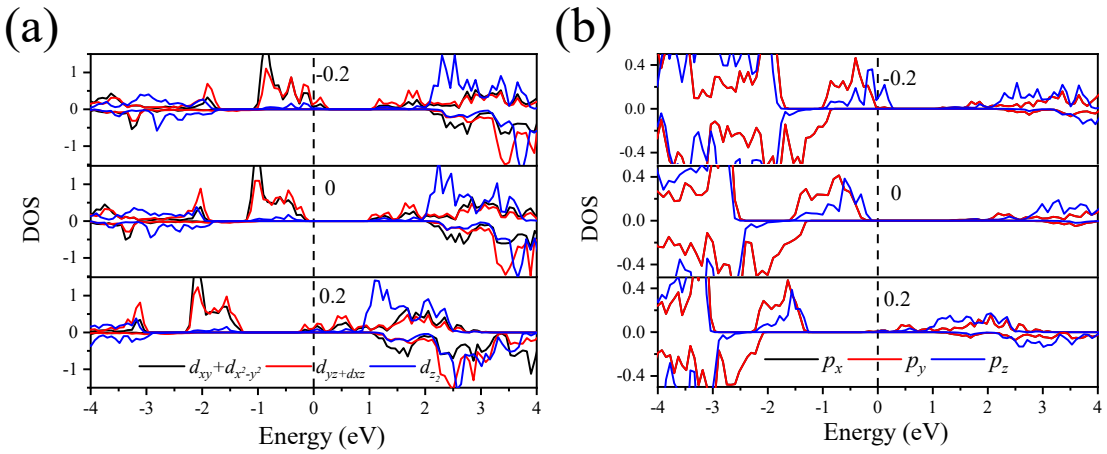

Figure S4. Projected DOS of (a) the *d* orbitals of the vanadium atom and (b) the *p* orbitals of the sulfur atom in the MgV_2S_4 monolayer under different doping. The Fermi level is indicated by the vertical dashed line. The 0.2 and -0.2 represent 0.2 electrons and holes per unit cell, respectively.

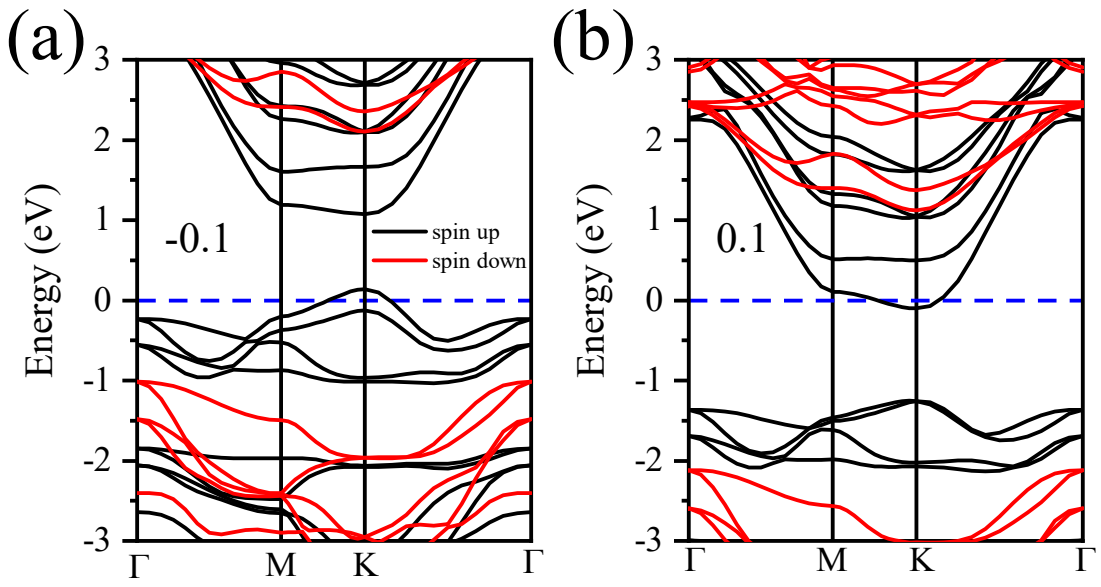

Figure S5. The band structure at doping concentration of 8×10^{13} cm^{-2}. 0.1 and -0.1 represents 0.1 electrons and holes per unit cell, respectively.

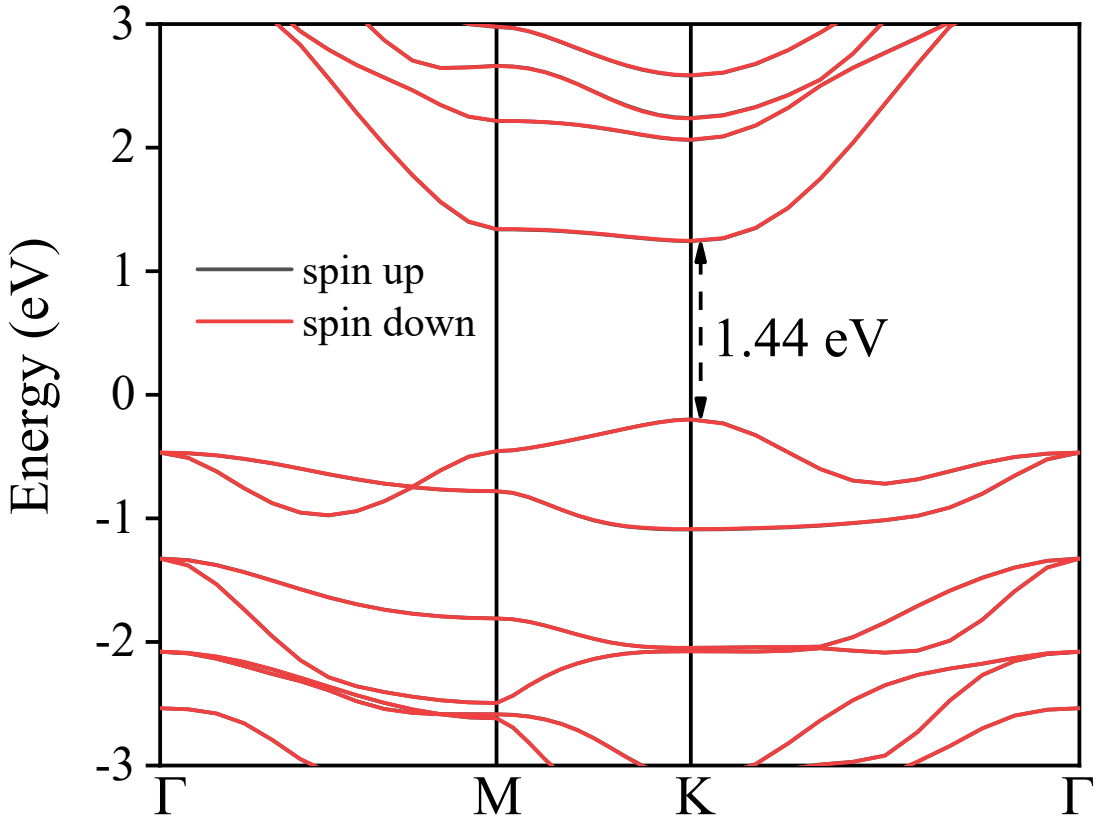

Figure S6. The band structure of MgV_2S_4 monolayer when the two layers are magnetized anti-parallel.

Table S2. The evolution of σ/τ with temperature without doping.

	σ/τ (S/m•s)	
	parallel	antiparallel
300	0	0
400	2.81×10^{10}	0
500	4.79×10^{12}	1.02×10^{11}
600	6.50×10^{13}	4.44×10^{12}