Supporting Information for

Structural stability and magnetic exchange coupling in Mn-doped monolayer/bilayer MoS_2

Qinglong Fang [a], Xumei Zhao [b], Yuhong Huang [c], Kewei Xu [a,d], Tai Min [a], Paul K. Chu [e,*], Fei Ma [a,e,*]

[a] *State Key Laboratory for Mechanical Behavior of Materials, Xi'an Jiaotong University, Xi'an 710049, Shaanxi, China*

[b] *College of Materials Science and Engineering, Shaanxi Normal University, Xi'an 710062, Shaanxi, China*

[c] *College of Physics and Information Technology, Shaanxi Normal University, Xi'an 710062, Shaanxi, China*

[d] *Department of Physics and Opt–electronic Engineering, Xi'an University of Arts and Science, Xi'an 710065, Shaanxi, China*

[e] *Department of Physics and Materials Science, City University of Hong Kong, Tat Chee Avenue, Kowloon, Hong Kong, China*

[*] Corresponding authors.

E-mail addresses: **paul.chu@cityu.edu.hk** (Paul K. Chu); **mafei@mail.xjtu.edu.cn** (F. Ma).

Table S1 A summary of the results of the optimized structures of Mn-doped monolayer MoS_2 in five doping configurations. The bond length of Mn-S (L_{Mn-S}) and Mn-Mo (L_{Mn-Mo}), the total magnetic moment (μ_{tot}) and local magnetic moment of the Mn dopant (μ_{Mn}) in the optimized structure.

Configuration	L_{Mn-S} (Å)	L_{Mn-Mo} (Å)	μ_{tot} (μ_B)	μ_{Mn} (μ_B)
A_{Mo}	2.391	3.119	5.00	4.0
A_S	2.316	4.395	5.08	4.31
A_H	2.157	3.088	3.00	3.04
Mn_{Mo}	2.311	3.109	1.00	1.04
Mn_S	3.109	2.624	3.00	3.62

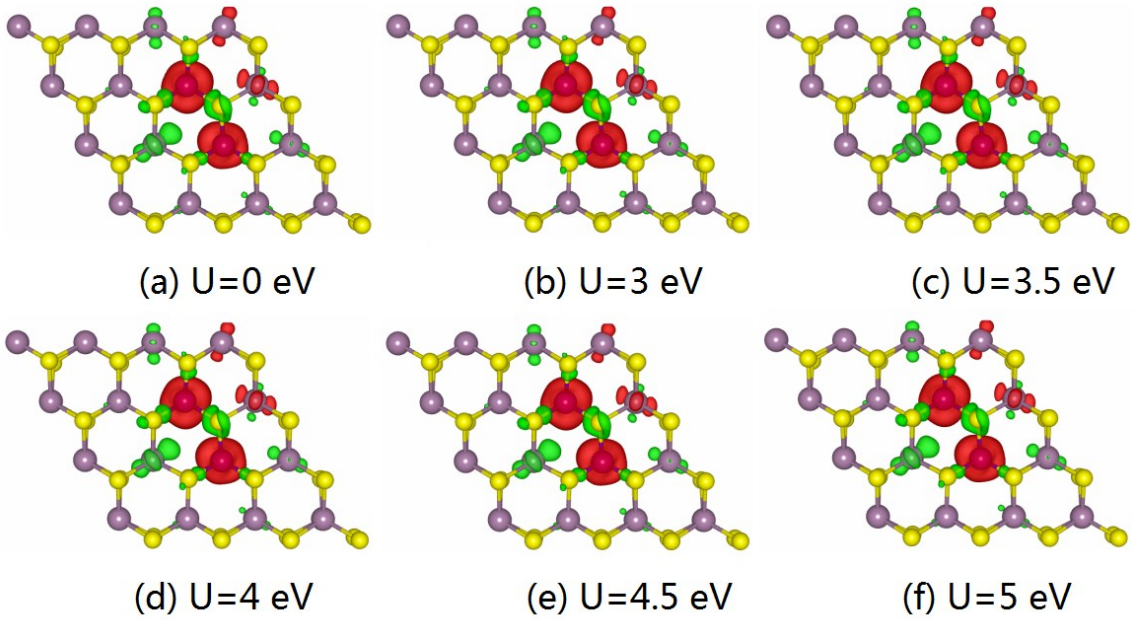

Fig. S1. Spin-density isosurface distributions of Mn-doped monolayer MoS_2 with the $2Mn_{Mo}$ configuration at (a) U= 0 eV, (b) U= 3 eV, (c) 3.5 eV, (d) 4 eV, (e) 4.5 eV, and (f) 5 eV. The red and green isosurfaces represent the positive and negative spin densities, respectively. The isosurface value is 0.003 e/Å^3.

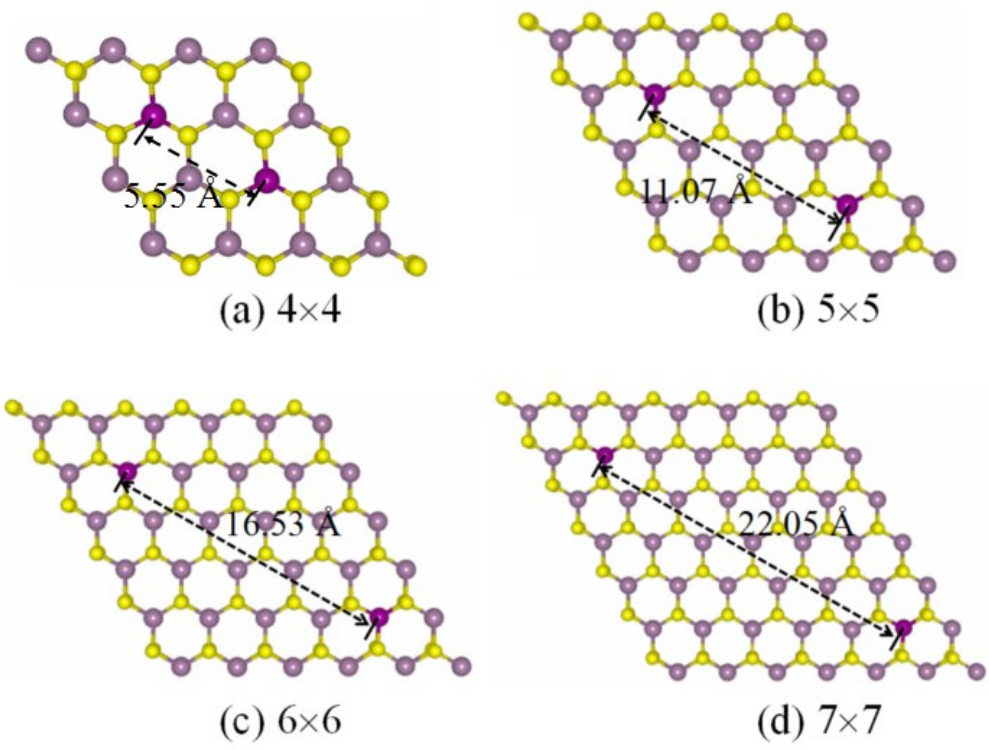

Fig. S2. Atomic models of Mn-doped monolayer MoS_2 with the $2Mn_{Mo}$ configuration in (a) 4′4, (b) 5′5, (c) 6′6, and (d) 7′7 supercell.

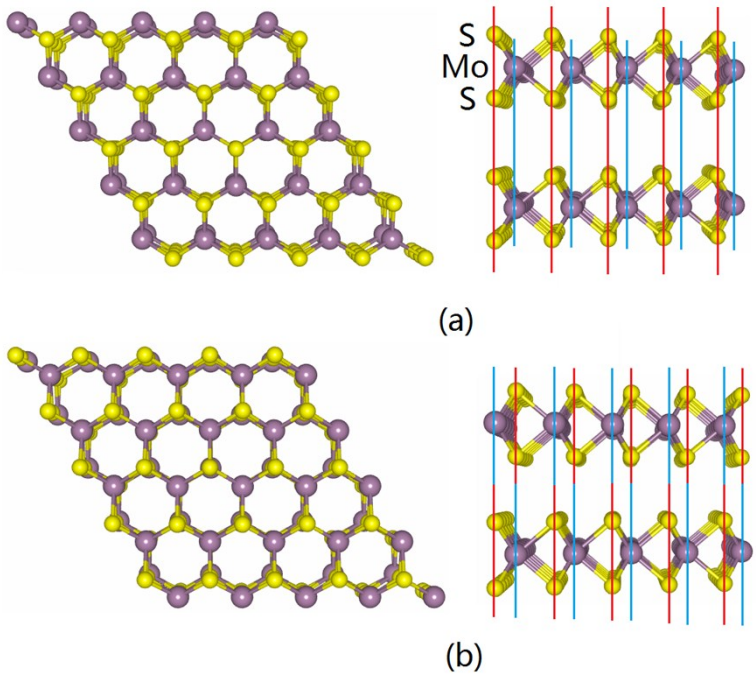

Fig. S3. Top and side views of the (a) AA and (b) AB configurations of the bilayer MoS_2.

Table S2 Summary of the results of the optimized structures of the single transition metal (Cr, Mn, Fe, and Co) doping in bilayer MoS_2 with the AA and AB configurations. The interlayer distance (d), bond length of Mn-S (L_{Mn-S}) , the total magnetic moment (μ_{tot}) and local magnetic moment of the transition metal dopant (μ_{TM}) in the optimized structure.

				d (Å)	L_{TM-S} (Å)	μ_{tot} (μ_B)	μ_{TM} (μ_B)
Bilayer MoS_2	AA	H	Cr	3.663	2.574	5.82	4.05
			Mn	3.665	2.563	5.07	4.05
			Fe	3.631	2.521	3.55	2.89
			Co	3.588	2.482	1.16	1.40
		S	Cr	3.866	2.233	5.06	4.16
			Mn	3.872	2.113	3.03	3.20
			Fe	3.826	2.085	2.10	2.45
			Co	3.785	2.037	1.00	1.22
	AB	H	Cr	3.124	2.439	5.44	3.77
			Mn	3.112	2.421	3.87	3.59
			Fe	3.100	2.383	2.34	2.34
			Co	3.074	2.355	1.02	1.04
		S	Cr	3.252	2.204, 2.257	4.02	3.21
			Mn	3.121	2.155, 2.220	3.00	2.83
			Fe	3.099	2.114, 2.162	2.00	1.64
			Co	3.087	2.097, 2.127	0.99	0.56

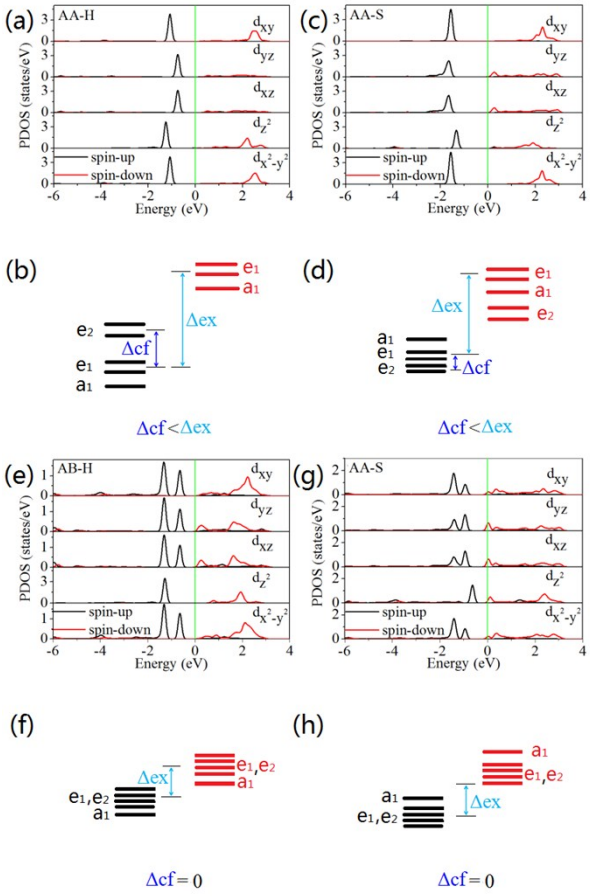

Fig. S4. Orbital decomposed density of states and the splitting of Cr 3*d* orbitals for single Cr dopant at the different sites in the bilayer MoS_2.

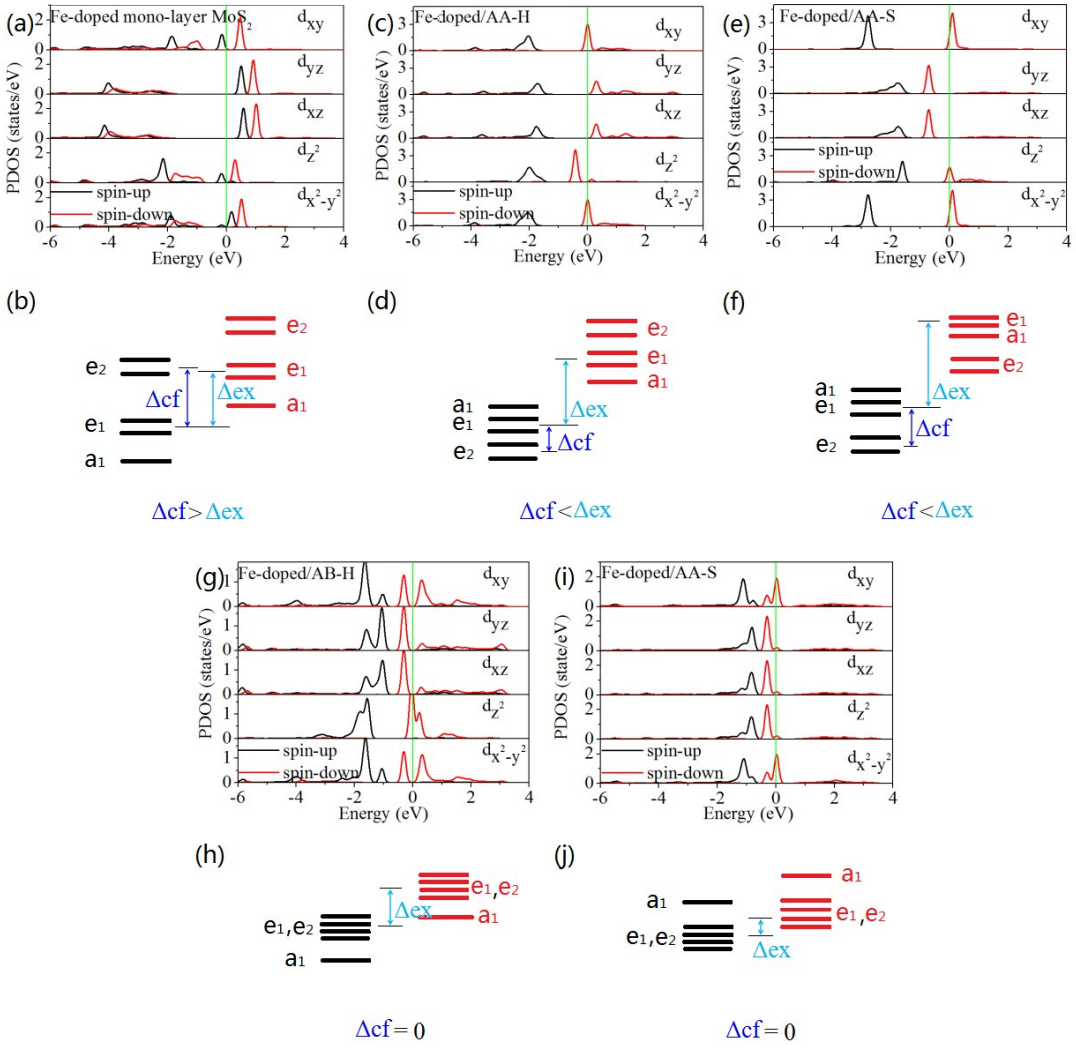

Fig. S5. Orbital decomposed density of states and the splitting of Fe 3*d* orbitals for single Fe dopant at the different sites in the monolayer and bilayer MoS_2.

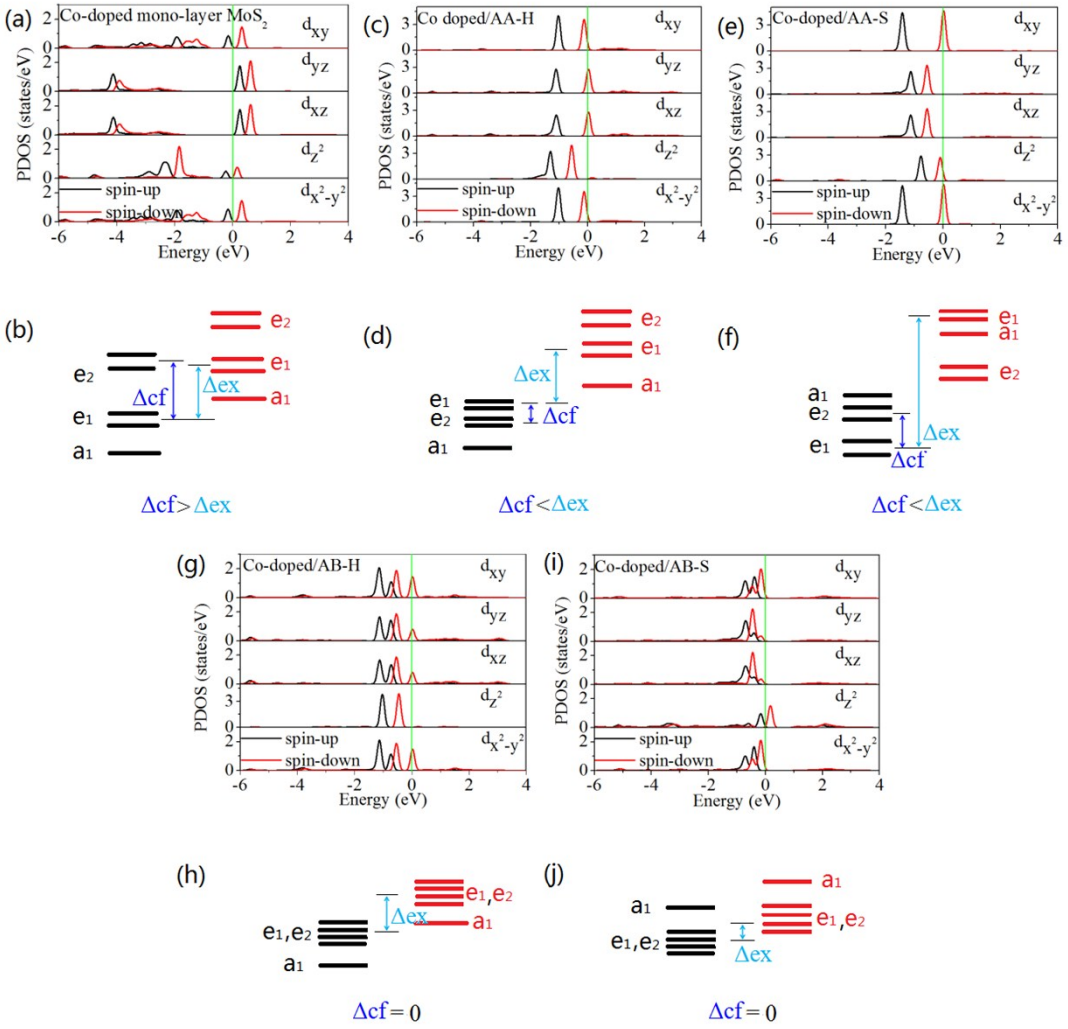

Fig. S6. Orbital decomposed density of states and the splitting of Co 3*d* orbitals for single Co dopant at the different sites in the monolayer and bilayer MoS_2.