

Electronic Supplementary Information

Orienting spins in dually doped monolayer **MoS₂:** From one-sided to double-sided doping

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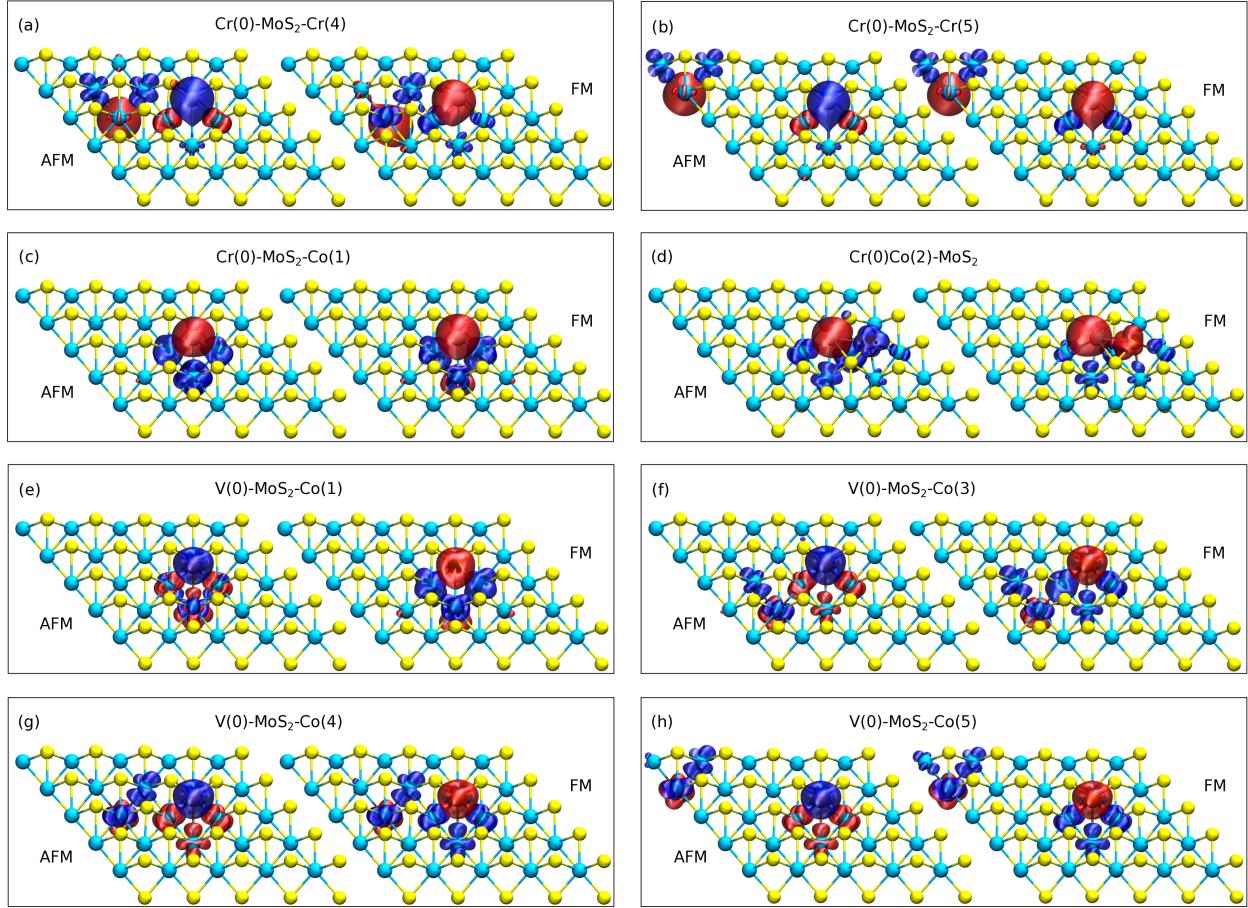


Fig. S1 Spin densities of (a) Cr(0)-MoS₂-Cr(4), (b) Cr(0)-MoS₂-Cr(5), (c) Cr(0)-MoS₂-Co(1), (d) MoS₂-Cr(0)Co(2), (e) V(0)-MoS₂-Co(1), (f) V(0)-MoS₂-Co(3), (g) V(0)-MoS₂-Co(4), and (h) V(0)-MoS₂-Co(5). The left configuration corresponds to the antiferromagnetic and the one on the right to the ferromagnetic state, respectively. The majority (minority) spin density component is shown in red (blue) with the isosurface value of 0.005 (-0.005) $e/\text{\AA}^3$. The 5×5 supercell size was used.

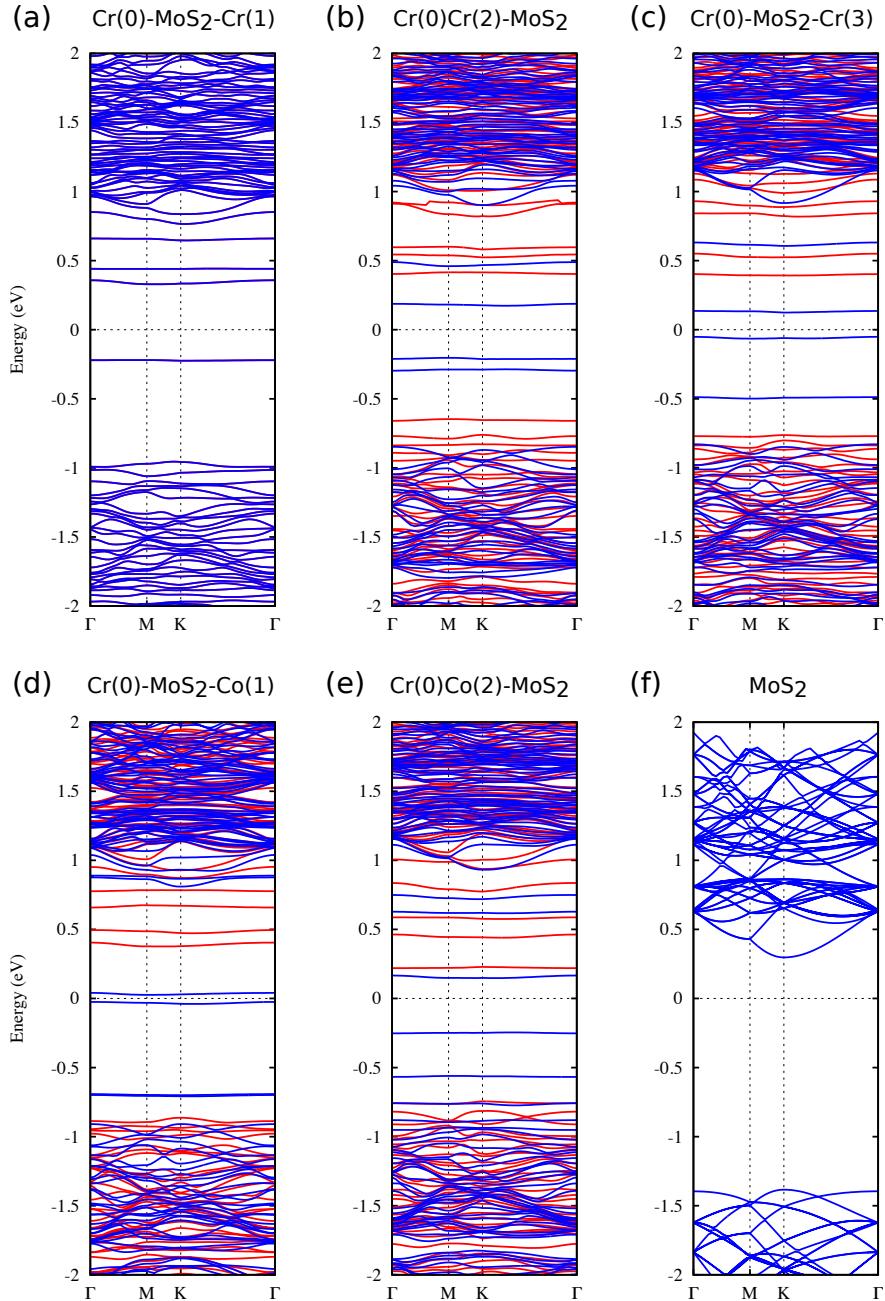


Fig. S2 Spin band structures of (a) Cr(0)-MoS₂-Cr(1), (b) Cr(0)Cr(2)-MoS₂, (c) Cr(0)-MoS₂-Cr(3), (d) Cr(0)-MoS₂-Co(1), and (e) Cr(0)Co(2)-MoS₂ systems. In (f), the band structure of spin-unpolarized MoS₂ is given as a reference. Red (blue) bands correspond to spin-up (spin-down) states.

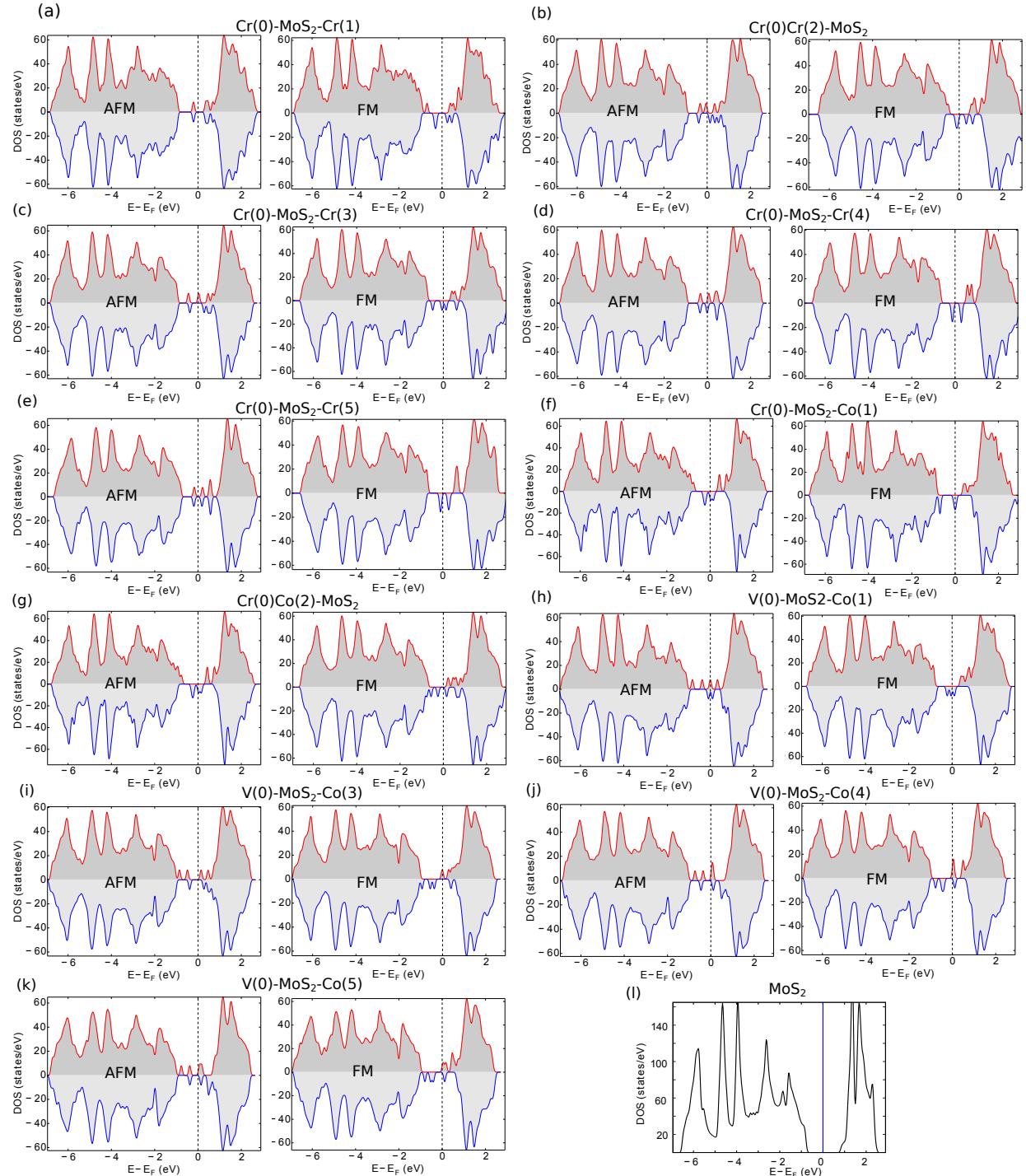


Fig. S3 Total density of states (TDOS) for the dual substitutional TM dopant atoms in the monolayer-MoS₂ systems in (a)-(k). In (l), TDOS for the pure MoS₂ is given as a reference.

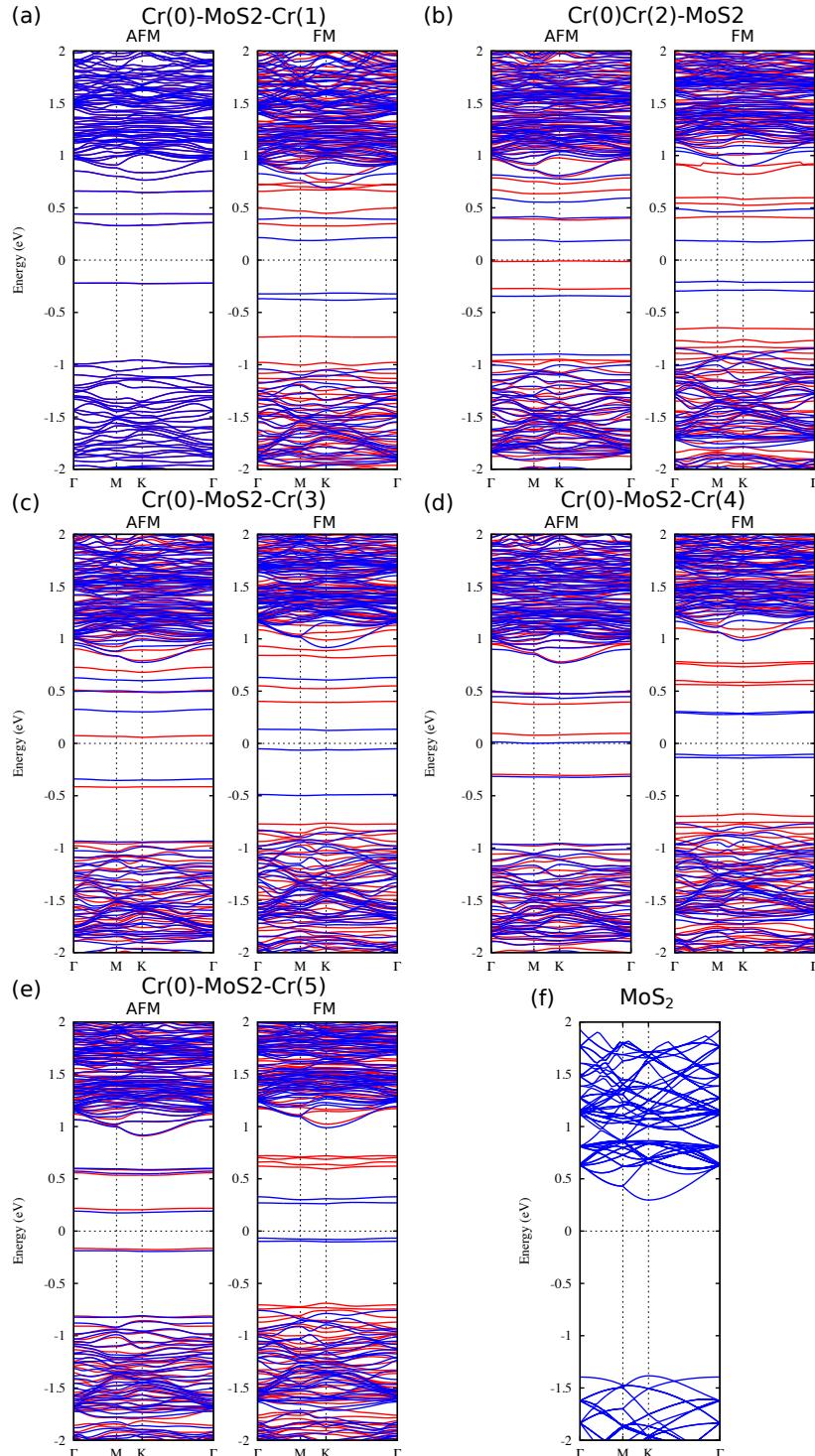


Fig.S4 Spin band structures of (a) Cr(0)-MoS₂-Cr(1), (b) Cr(0)Cr(2)-MoS₂, (c) Cr(0)-MoS₂-Cr(3), (d) Cr(0)-MoS₂-Cr(4), and (e) Cr(0)-MoS₂-Cr(5). In (f), the band structure of undoped MoS₂ is given as a reference.

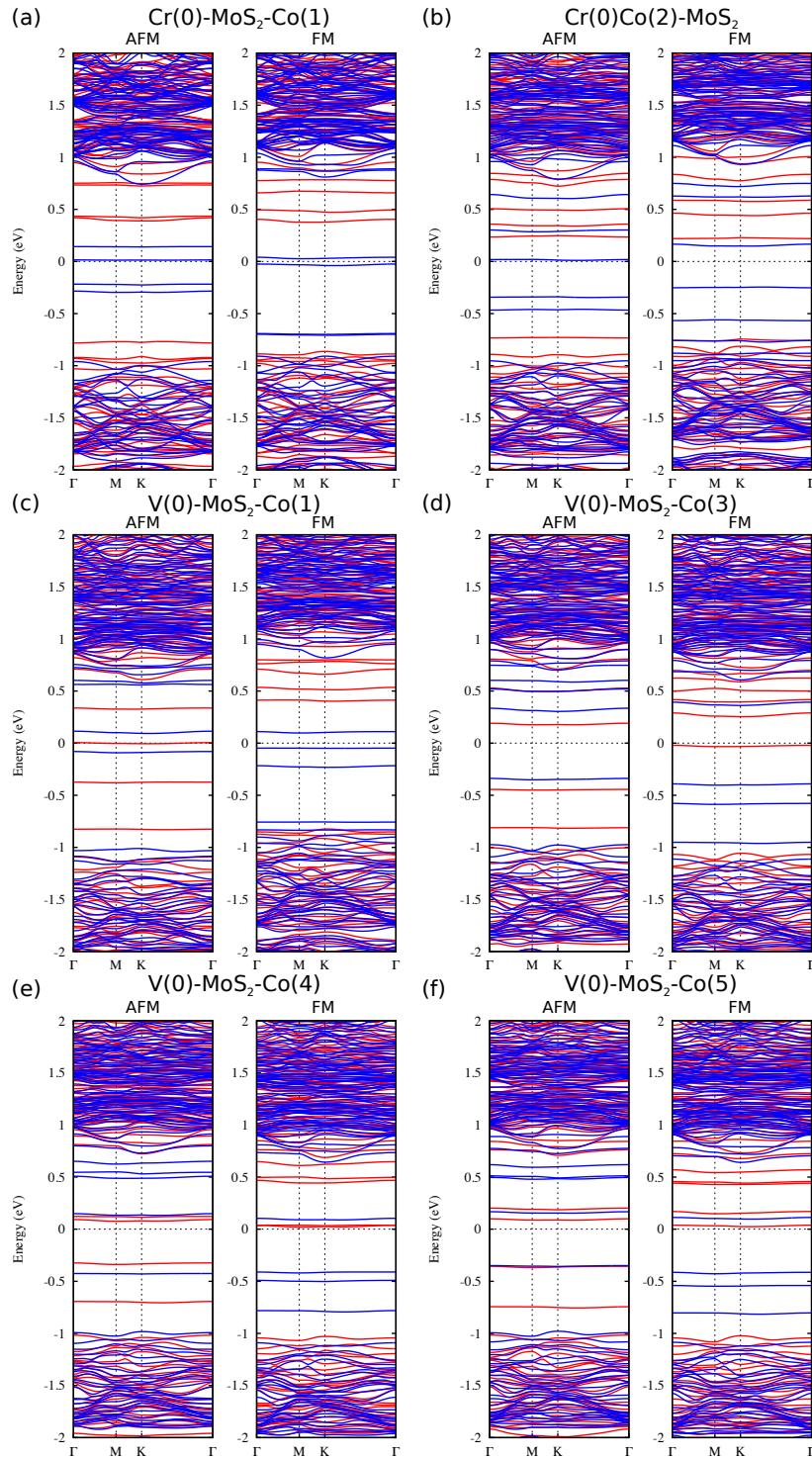


Fig. S5 As Fig. S3, but for (a) Cr(0)-MoS₂-Co(1), (b) Cr(0)Co(2)-MoS₂, (c) V(0)-MoS₂-Co(1), (d) V(0)-MoS₂-Co(4), and (e) V(0)-MoS₂-Co(5).

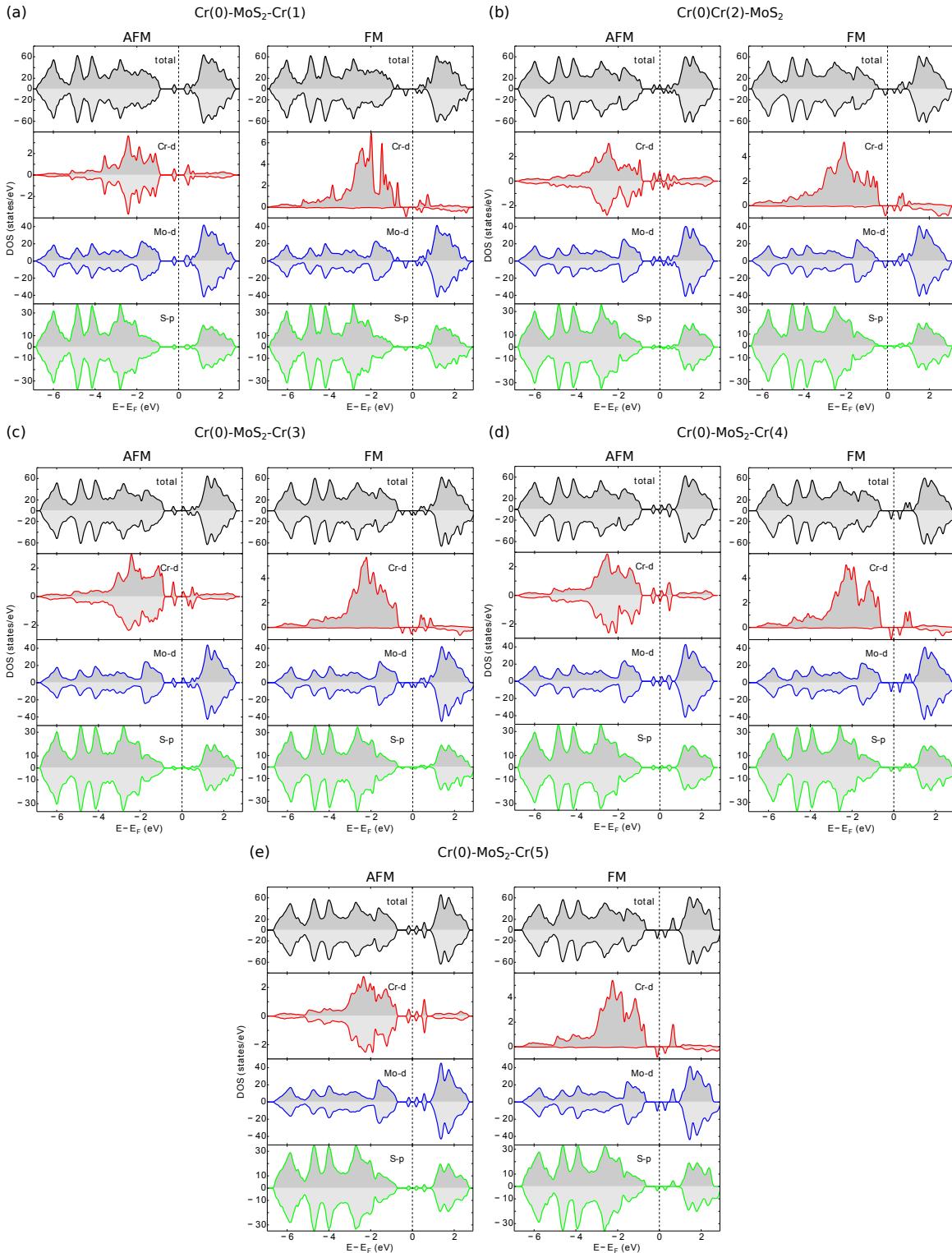


Fig. S6 Partial density of states (PDOS) for (a) Cr(0)-MoS₂-Cr(1), (b) Cr(0)-Cr(2)-MoS₂, (c) Cr(0)-MoS₂-Cr(3), (d) Cr(0)-MoS₂-Cr(4), and (e) Cr(0)-MoS₂-Cr(5) systems.

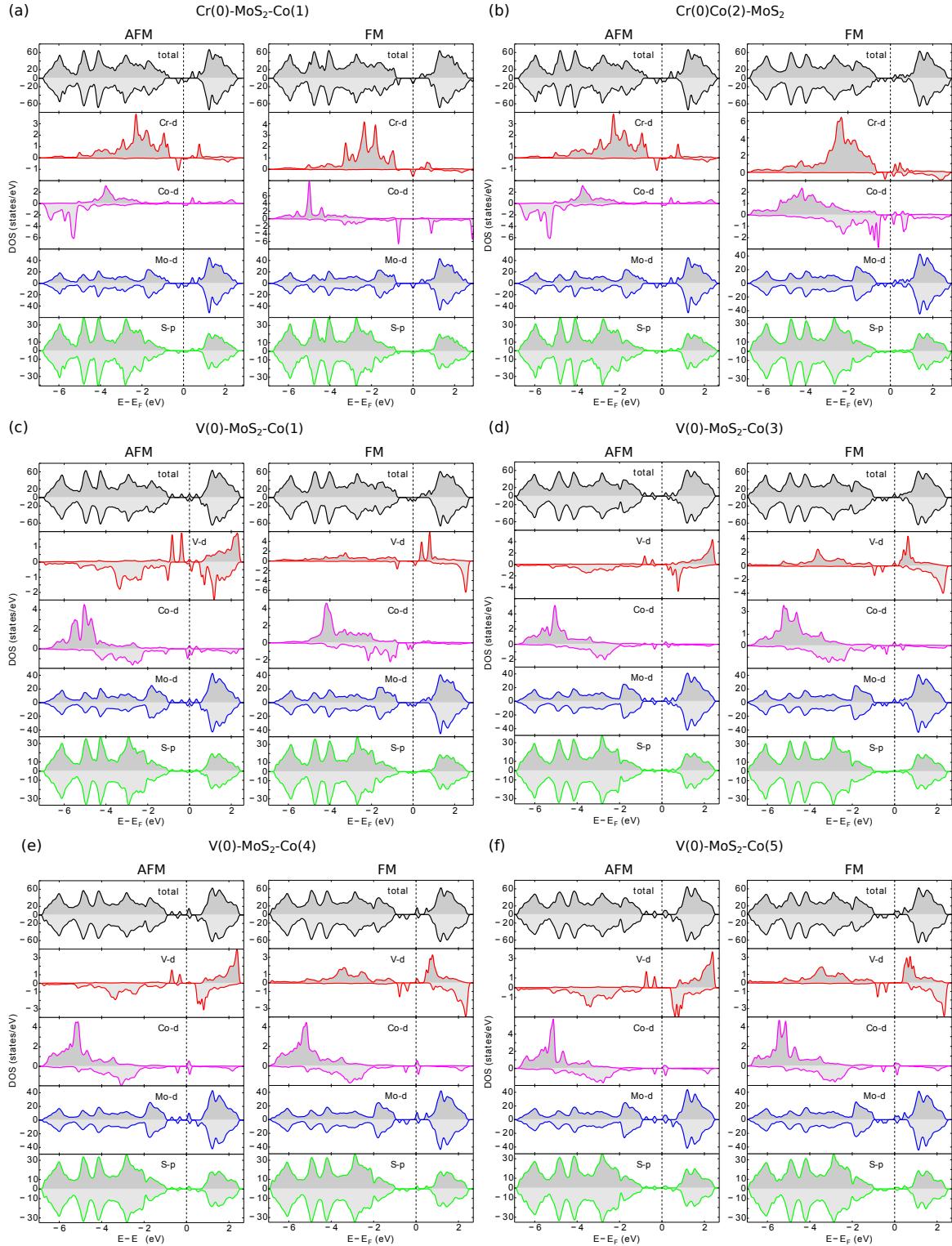


Fig. S7 Partial density of states (PDOS) for (a) Cr(0)-MoS₂-Co(1), (b) Cr(0)Co(2)-MoS₂, (c) V(0)-MoS₂-Co(1), (d) V(0)-MoS₂-Co(3), (e) V(0)-MoS₂-Co(4), and (f) V(0)-MoS₂-Co(5) systems.

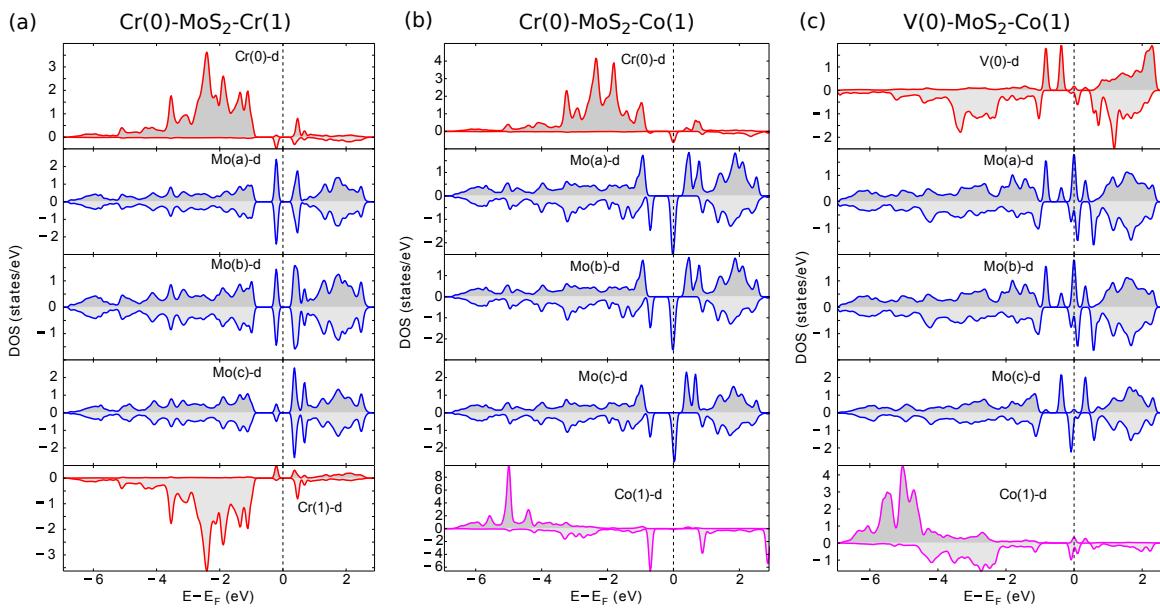


Fig. S8 Partial density of states (PDOS) for dopant atoms and nearest three Mo atoms in (a) Cr(0)-MoS₂-Cr(1), (b) Cr(0)-MoS₂-Co(1), and (c) V(0)-MoS₂-Co(1) systems in their AFM, FM, and AFM ground states, respectively.

Table S1 Hubbard U parameters for the dual substitutional TM dopant atoms in the monolayer-MoS₂ systems at uniform concentration $\theta = 2/75$. For each system, the assigned U value, from linear response determination, is tabulated for the TM atoms i and j at both possible magnetic configurations (AFM/FM). Due to the calculation of the exchange energy value ($J = E_{\text{AFM}} - E_{\text{FM}}$) of these systems, we used the AFM-FM-averaged (bolded) values in the all electronic structure property calculations.

System	AFM i	FM i	aver. i	AFM j	FM j	aver. j
Cr(0)-MoS ₂ -Cr(1)	3.25	3.26	3.26	3.25	3.26	3.26
Cr(0)Cr(2)-MoS ₂	3.11	3.08	3.10	3.11	3.10	3.11
Cr(0)-MoS ₂ -Cr(3)	3.32	3.32	3.32	3.29	3.25	3.27
Cr(0)-MoS ₂ -Cr(4)	3.48	3.34	3.41	3.26	3.26	3.26
Cr(0)-MoS ₂ -Cr(5)	3.30	3.29	3.30	3.30	3.29	3.30
Cr(0)-MoS ₂ -Co(1)	3.02	3.15	3.09	5.25	5.85	5.55
Cr(0)Co(2)-MoS ₂	3.24	3.25	3.25	5.64	5.63	5.64
V(0)-MoS ₂ -Co(1)	4.22	4.19	4.21	5.25	5.74	5.50
V(0)-MoS ₂ -Co(3)	4.01	3.94	3.98	5.68	6.12	5.90
V(0)-MoS ₂ -Co(4)	3.99	3.96	3.98	5.96	6.05	6.01
V(0)-MoS ₂ -Co(5)	3.98	3.98	3.98	6.13	6.12	6.13

Table S2 Relative energies of the ground state and the first excited state for two strongly correlated heteronuclear dimer molecules VMo and CrMo. For both systems, the ground state is $S = 5/2$ and the first excited state is $S = 1/2$, and $d\sigma \rightarrow d\delta$ is the first excitation that takes place. Here, spin-orbit splitting is not considered. The assigned U values, from the linear response U determination, are communicated in the main text.

Molecule	Method	ΔE (eV)	ΔE (cm^{-1})
VCr	DFT	-	-
	DFT+U	0.6995	5666
	CASPT2 ¹	0.7005	5674
VMo	DFT	-0.0717	-581
	DFT+U	0.8912	7218
	CASPT2 ¹	0.6063	4911

¹ Ruiperez, F.; Ugalde, J. M.; Infante, I. *Inorg. Chem.* **2011**, 50, 9219–9229.