Supplementary Information

Enhancement of single-atom catalytic activity by interlayer charge transfer and magnetic coupling synergistic in electride-based heterostructure

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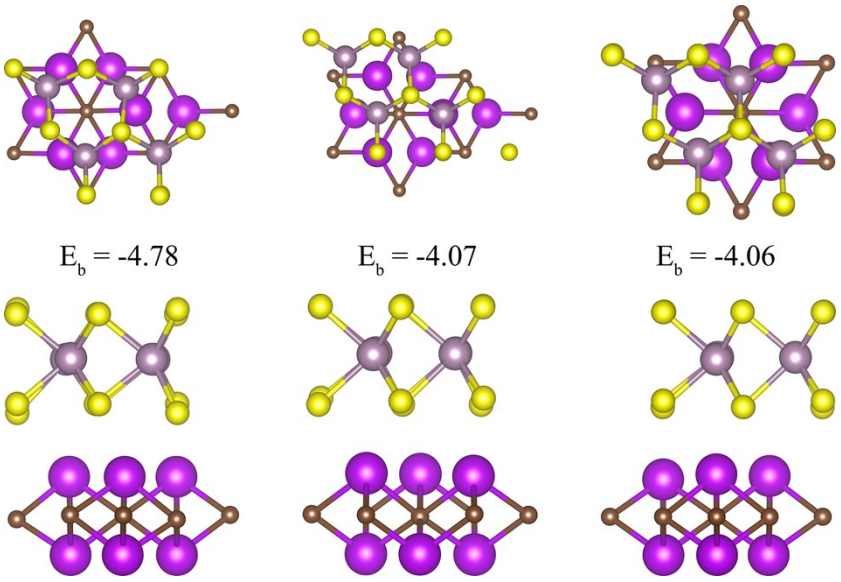

Fig. S1 The structure of MoS_2/Gd_2C heterostructures. E_b is the binding energy of each structure.

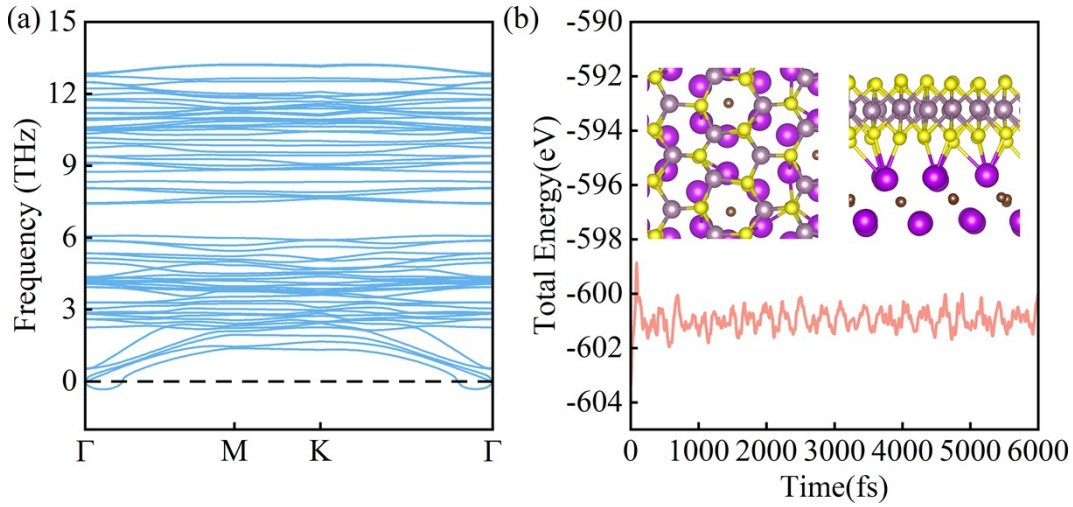

Fig. S2 (a) Phonon dispersions of MoS_2/Gd_2C heterostructure and (b) The fluctuations of energy and the final configuration of MoS_2/Gd_2C heterostructure obtained from AIMD simulation.

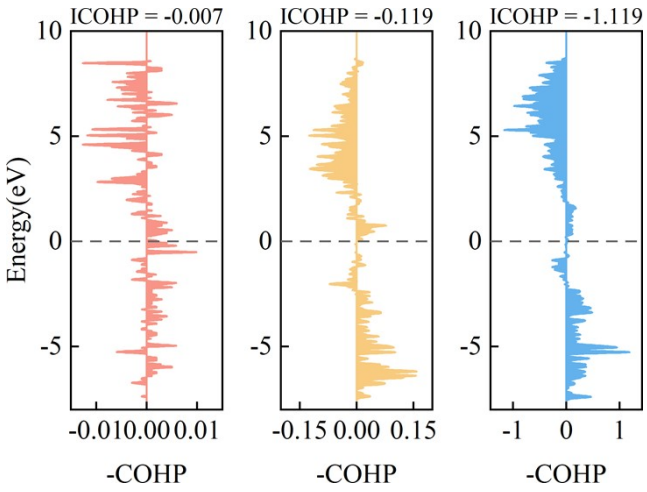

Fig. S3 The –COHP between Gd atom and three nearest-neighbor S atoms at the interface of MoS_2/Ca_2N heterostructure.

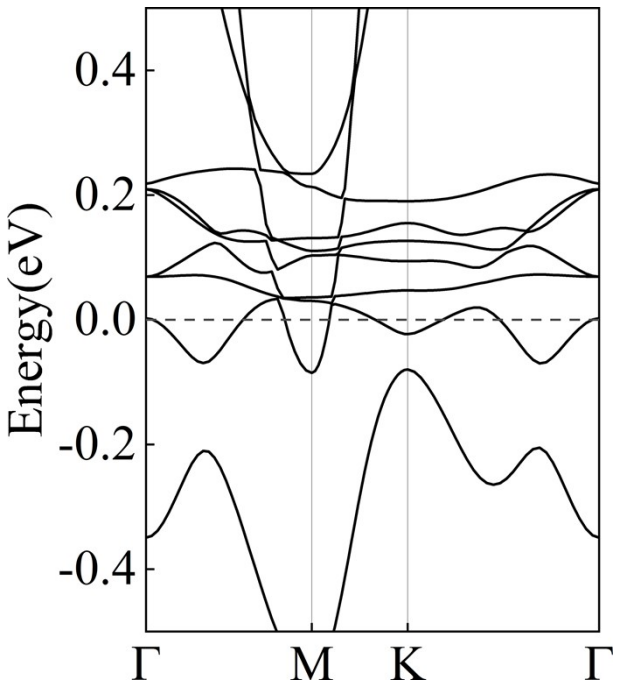

Fig. S4 The band structure of Gd_2C with both correlation effect and SOC.

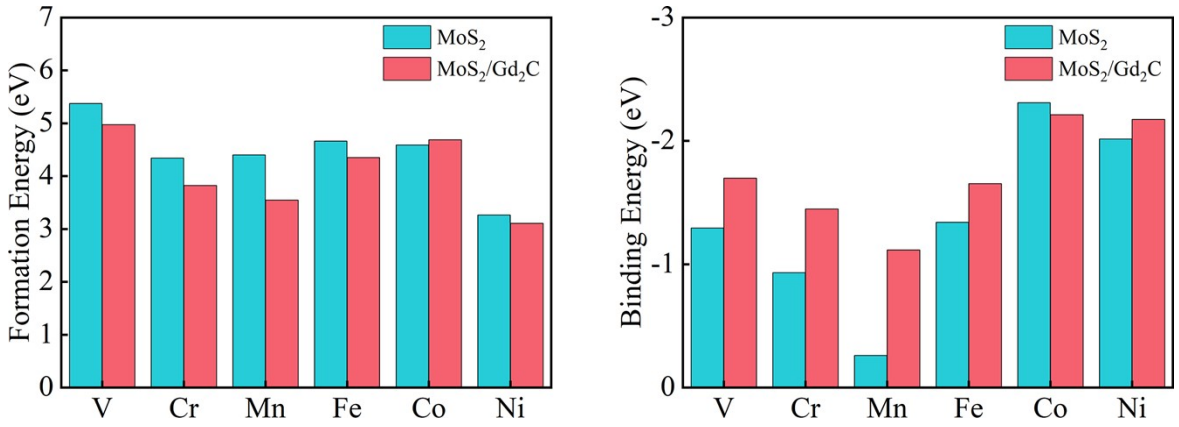

Fig. S5 (a) The formation energy and (b) the binding energy of TM atoms in the catalyst.

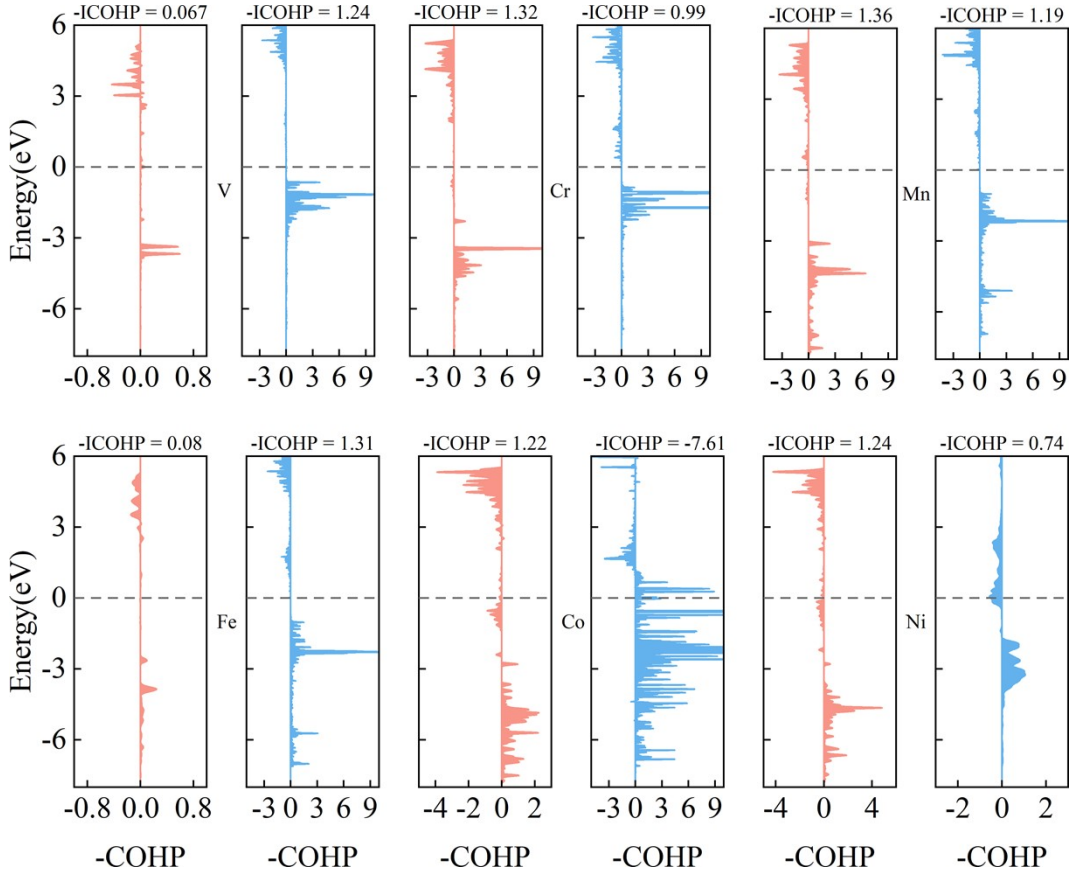

Fig. S6 The −COHP between the TM atom on MoS_2 and MoS_2/Gd_2C heterostructure and H atom.

Table S1 Adsorption energy and catalytic performance of MoS_2/Gd_2C with and without SOC.

	total	slab	E_{ads}	ΔG	ΔG_H
MoS_2/Gd_2C	-617.146	-615.100	1.250	0.390	1.641
MoS_2/Gd_2C +SOC	-626.627	-624.502	1.260	0.385	1.645

Table. S2 The energy corresponds to different magnetic moment orientations. The magnetic moment orientation of Co and Ni is not taken into consideration in this work due to their almost negligible magnetic moments.

orientation	V	Cr	Mn	Fe	Co	Ni
Parallel	-619.55	-621.08	-620.76	-619.20	-617.66	-617.69
Antiparallel	-619.56	-621.09	-620.74	-619.21	——	——

Table. S3 (a) The d-band center of TM atoms in the catalyst.

d-band center	V	Cr	Mn	Fe	Co	Ni
TM-MoS_2	-0.483	-1.226	-1.565	-1.539	-1.592	-1.237
TM-MoS_2/Gd_2C	-0.895	-1.571	-2.274	-1.346	-1.696	-2.331

Table. S4 The energy consumptions of each reaction step and overpotential in MoS_2.

atom	ΔG_1	ΔG_2	ΔG_3	ΔG_4	Overpotential (eV)
V	-0.649	0.230	3.064	2.275	1.834
Cr	0.077	1.609	0.489	2.745	1.515
Mn	1.401	-0.044	1.227	2.335	1.105
Fe	-0.083	1.238	1.910	1.855	0.680
Co	-0.175	1.340	2.087	1.67	0.857
Ni	0.762	1.439	1.744	0.976	0.514

Table. S5 The energy consumptions of each reaction step and overpotential in MoS_2/Gd_2C.

atom	ΔG_1	ΔG_2	ΔG_3	ΔG_4	Overpotential (eV)
V	-0.898	0.654	3.294	1.870	2.064
Cr	0.027	1.228	2.261	1.404	1.031
Mn	-0.670	1.763	1.772	2.054	0.824
Fe	-0.539	1.692	2.315	1.452	1.085
Co	-0.348	1.372	2.045	1.851	0.815
Ni	0.847	1.407	1.752	0.914	0.522