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Electronic Supporting Information

A New Phase of Two-Dimensional ReS2 Sheet with Tunable Magnetism

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Table S1 The effect of ENCUT for lattice constant (a and b in the unit of Å, γ in the unit of degree) and relative energy per stoichiometric formula in the unit of eV based on the PBE method.

ENCUT		a/Å	b/Å	γ/°	ΔE
400	DT-1	6.41	6.51	118.84	0.00
	DT-2	5.73	6.39	90.09	0.06
	Tri	5.79	5.79	120	0.54
500	DT-1	6.41	6.51	118.83	0.00
	DT-2	5.73	6.39	90.09	0.06
	Tri	5.79	5.79	120	0.54

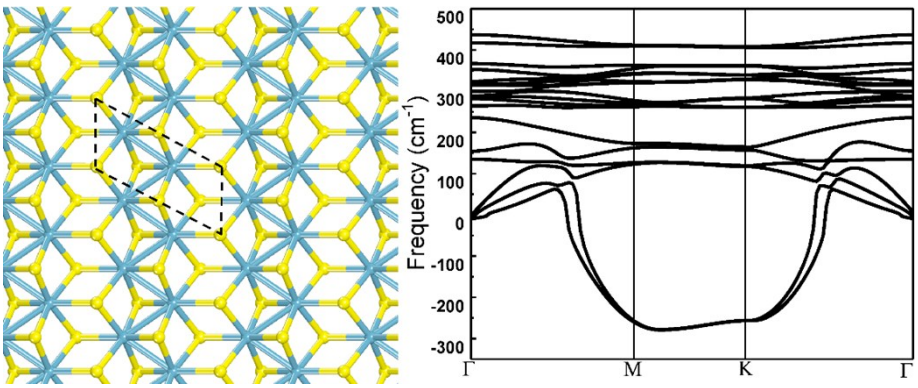

Fig. S1 The structure (a) and phonon spectrum (b) of ZT (zigzag chain direction) phase of ReS_2. Γ (0, 0, 0), K (1/2, 1/2, 0), and M (0, 1/2, 0) refer to the high-symmetric k-points in the first Brillouin zone. The red dashed parallelogram indicates the unit cell.

Table S2 The lattice constant (a and b in the unit of Å, γ in the unit of degree) and relative energy per stoichiometric formula in the unit of eV based on the HSE06 method.

	a/Å	b/Å	γ/°	ΔE
DT-1	6.36	6.46	118.87	0.00
DT-2	5.67	6.34	90.12	0.07
Tri	5.74	5.74	120.00	0.42

Table S3 The elastic constant (C11, C21/12, C22, C66) and Young's modulus Y^{2D} along [100] and [010] directions in the unit of N/m by PBE method.

Phase	C_{11}	C_{22}	$C_{21/12}$	C_{66}	Y_{100}^{2D}	Y_{010}^{2D}
DT-1	159	159	32	58.0	153	153
DT-2	142	141	29	50	136	135
Tri	106	106	26	39	100	100

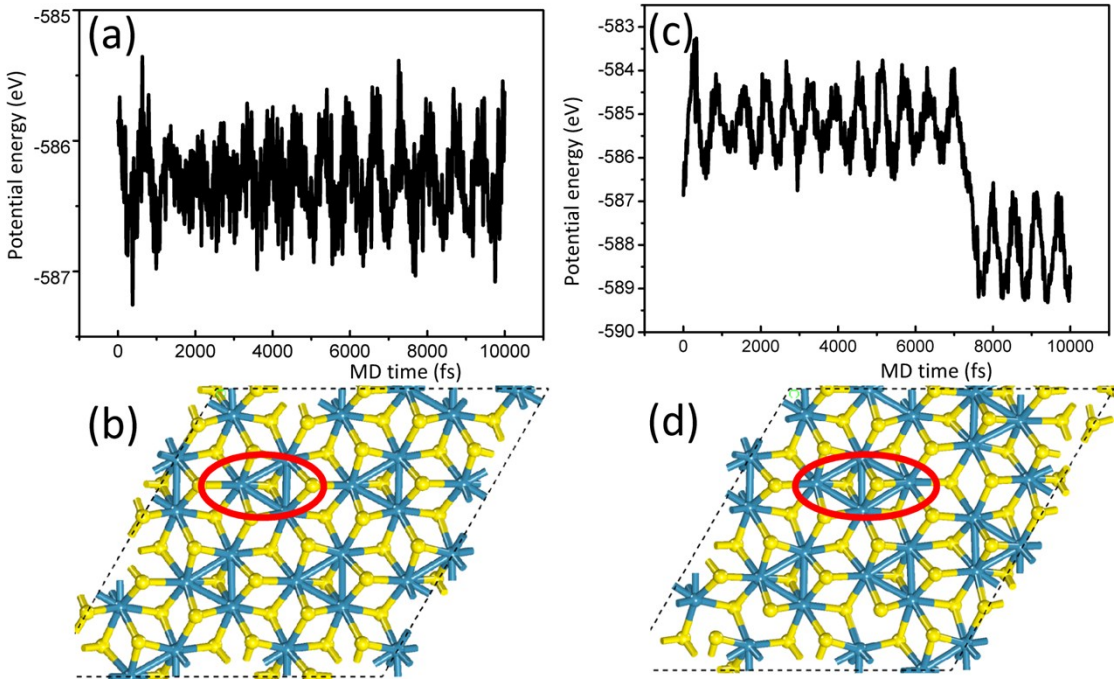

Fig. S2 The thermodynamics of Tri phase by BOMD (a, c) the relationship between total energy and time in 300K and 400K; (b, d) the corresponding structures.

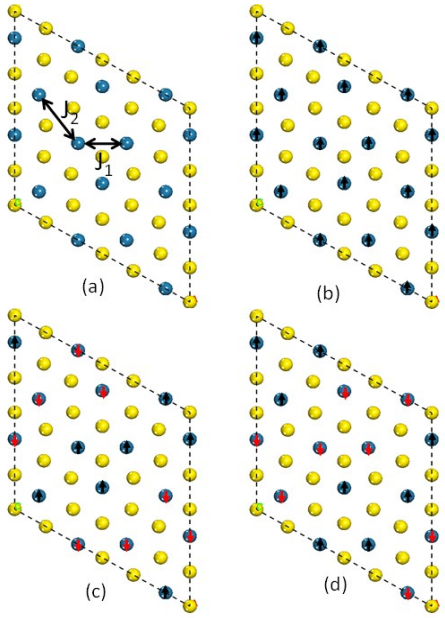

Fig. S3 The Tri phase structure with 2×2×1 super cell: (a) J_1 and J_2 indicate the nearest and next-nearest exchange paremeter; (b, c, d) the ferromagnetic (FM), antiferromagnetic-1(AFM1) and antiferromagnetic-2(AFM2) state, respectively. The black and red arrows show the spin up and down orientation, respectively. Rhenium and Sulfur atoms are in green and yellow, respectively.

Table S4 The magnetic moment (MM) with the unit of μB of Re atoms by removing and adding electrons in the Tri phase of ReS_2 by HSE06 method. GS and FM are the ground state and ferromagnetic state for short, respectively. e and h indicate electron and hole doping per atom, respectively.

	0.0556h	0.0417h	0.0278h	0.0139h	0	0.0139e	0.0278e	0.0417e	0.0556e
1	0.014	0.881	0.975	0.911	0.925	0.924	0.695	0.907	0.907
2	0.92	0.91	0.924	0.664	0.925	0.914	0.92	0.862	0.877
3	0.945	0.013	0.92	0.917	0.925	0.82	0.921	0.869	0.89
4	0.973	0.856	0.934	0.912	0.925	0.902	0.919	0.254	0.233
5	0.86	0.962	0.925	0.921	0.925	0.792	0.881	0.836	0.89
6	0.916	0.921	0.934	0.924	0.925	0.897	0.92	0.9	0.876
7	0.92	0.901	0.869	0.868	0.925	0.922	0.88	0.896	0.907
8	0.973	0.873	0.004	0.922	0.925	0.898	0.678	0.894	0.877
9	0.92	0.921	0.869	0.904	0.925	0.904	0.878	0.883	0.89
10	0.86	0.971	0.92	0.913	0.925	0.863	0.92	0.737	0.233
11	0.014	0.914	0.924	0.921	0.925	0.9	0.919	0.875	0.89
12	0.92	0.634	0.975	0.912	0.925	0.916	0.721	0.874	0.877
average size of MM	0.770	0.813	0.848	0.891	0.925	0.888	0.854	0.816	0.779
GS	FM	FM	FM	FM	FM	FM	FM	FM	FM

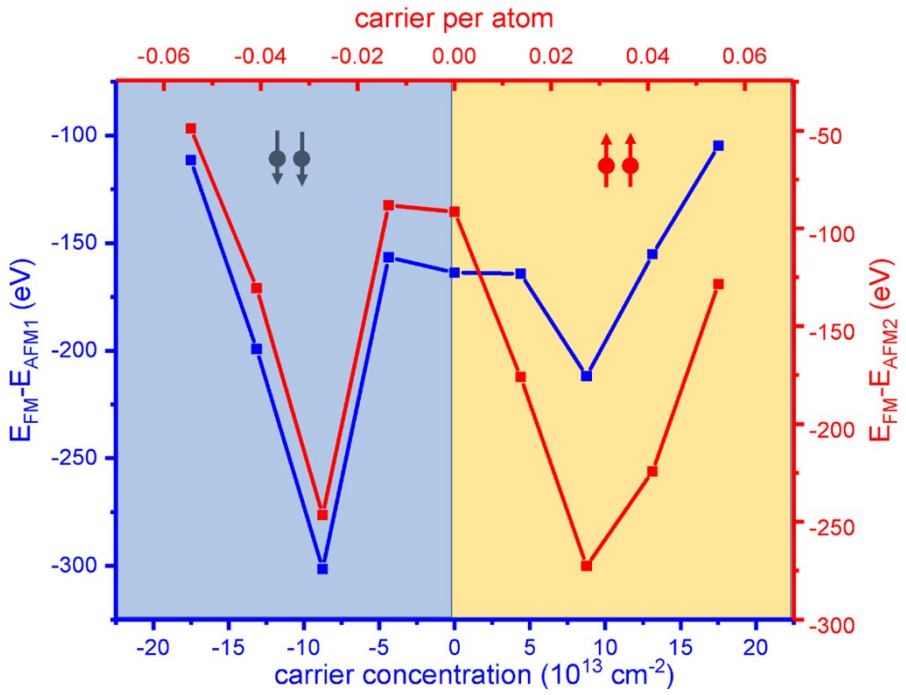

Fig. S4 Relative energy of FM and AFM1 state, FM and AFM2 state with the variation of carrier concentration for 2D Tri phase of ReS_2 by HSE06 method. The positive and negative value indicate electron and hole doping in the neutral system, respectively. The up and down arrow indicates the up-spin and down-spin direction, respectively. The light yellow and blue region are all the FM GS with different spin orientations.

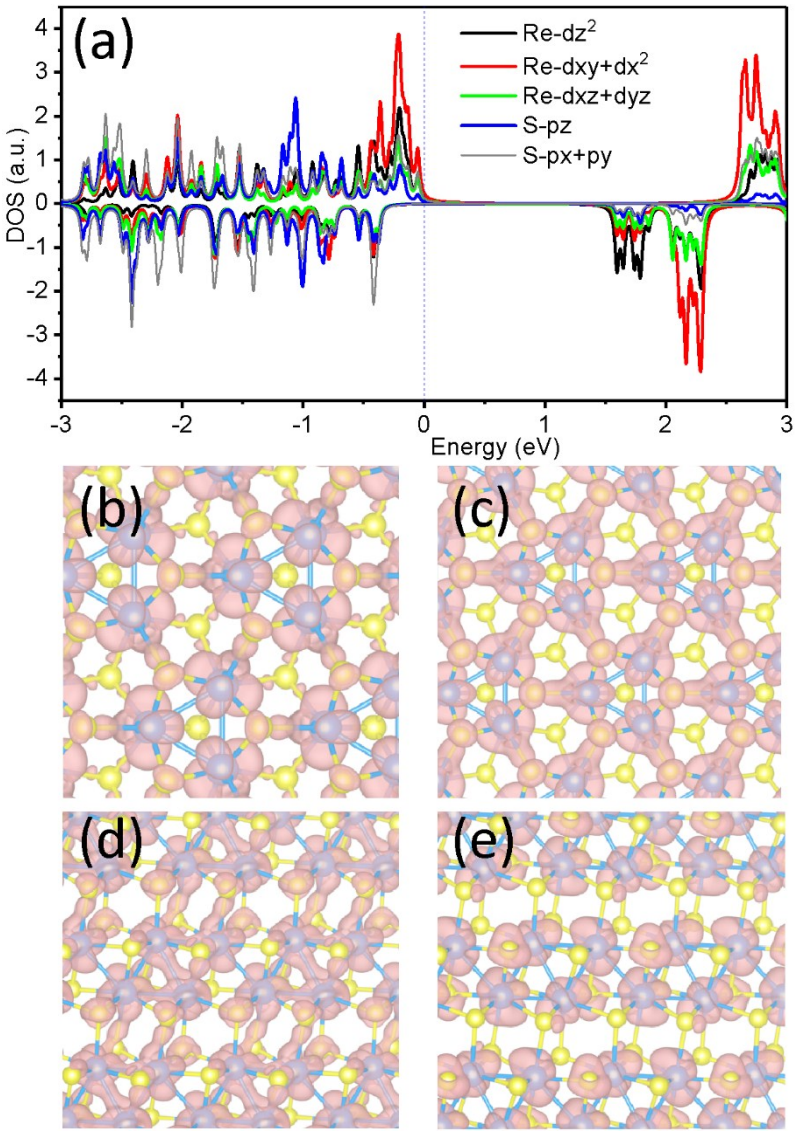

Fig. S5 Calculated spin-resolved density of state (pDOS) of Tri phase of 2D ReS_2 nanosheet (a); bonding (b, d) and antibonding (c, e) state of Tri and DT-1 phase of ReS_2 nanosheet with the isosurface value of 0.003e/bohr^3, respectively. Blue dashed line in (a) is the Fermi level.

The adsorption sites of Li atom on the surface of 2×2 unit cell DT-1 and Tri structure in Fig. S6. For DT-1, five adsorption sites including Re1, Re2, Re3, Re4, and S5 are considered, such as Re '1-4' site, they are from diamond-shaped (DS) Re4 cluster, according to clockwise sense, named as Re1, Re2, Re3 and Re4, the Li atom adsorbs on the above of every Re atom connected with three S atoms. The S5 site is on the non-Re-chain site, the adsorption configuration is similar to the Re site. For the Tri, three adsorption sites are taken into account, "3Re", "Re" and "S" sites are the adsorption sites on the top of cluster center including three Re atoms, one Re atom and one bottom S atom, respectively. Thus, the adsorption configurations and adsorption energy of Li atom on the DT-1 and Tri phase are in Fig. S6 and Table S5. Where the formula of adsorption energy of Li atom showed as $E_{ads} = E_{total} - E_{surf} - nE_{Li}$, E_{toal} and E_{surf} are the total energy of surf with and without Li atom, E_{Li} is the energy of Li atom, n is the number of Li atom. For the DT-1, Re1 site owns the lowest adsorption energy with -1.95eV in Table S5, the relative size: Re1< Re4< Re2< S5< Re3. While the lowest energy adsorption configuration of Li atom in the Tri phase is 3Re site of -2.72eV, it is much lower than DT-1 by 0.77eV. They are all lower than the binding energy of bulk metal Li with -1.59eV, It means that Li atom can intercalate in DT-1 and Tri structure, without aggregations.

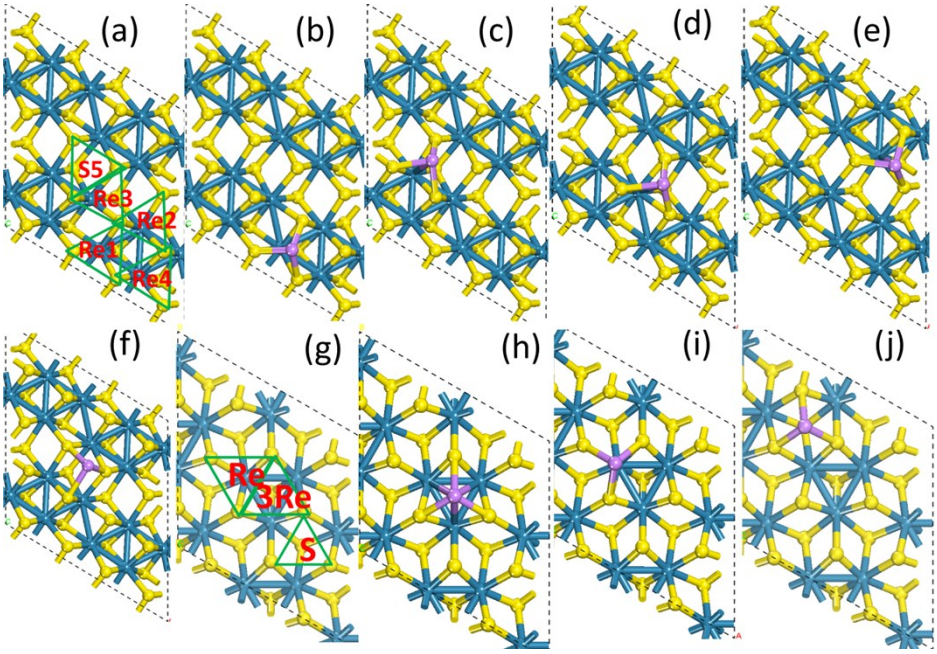

Fig. S6 The possible adsorption sites of Li atom on the 2×2 unit cell DT-1 (a) and Tri (g) phase. the corresponding adsorbed structure of DT-1 shows Re1(b), Re2(c), Re3(d), Re4(e), and S5(f); the possible adsorbed structure of Tri phase indicates 3Re(h), Re(i), and S(j).

Table S5 The possible adsorption energy (E_{ads}) and relative energy (ΔE) of Li atom on the 2×2 unit cell DT-1 and Tri phase. The binding energy per Li atom in the bulk metal Li with the body-center cubic (BCC) symmetry is -1.59eV. The formula of adsorption energy showed as: $E_{ads}=E_{total}-E_{surf}-E_{Li}$, among E_{total} and E_{surf} are the total energy of surf with and without Li atom, respectively. E_{Li} is the energy of Li atom.

	Conf.	Eads/eV	ΔE/eV
DT-1	Re1	-1.95	0.00
	Re2	-1.76	0.19
	Re3	-1.60	0.35
	Re4	-1.85	0.10
	S5	-1.70	0.25
Tri	3Re	-2.72	0.00
	Re	-2.64	0.08
	S	-2.47	0.25

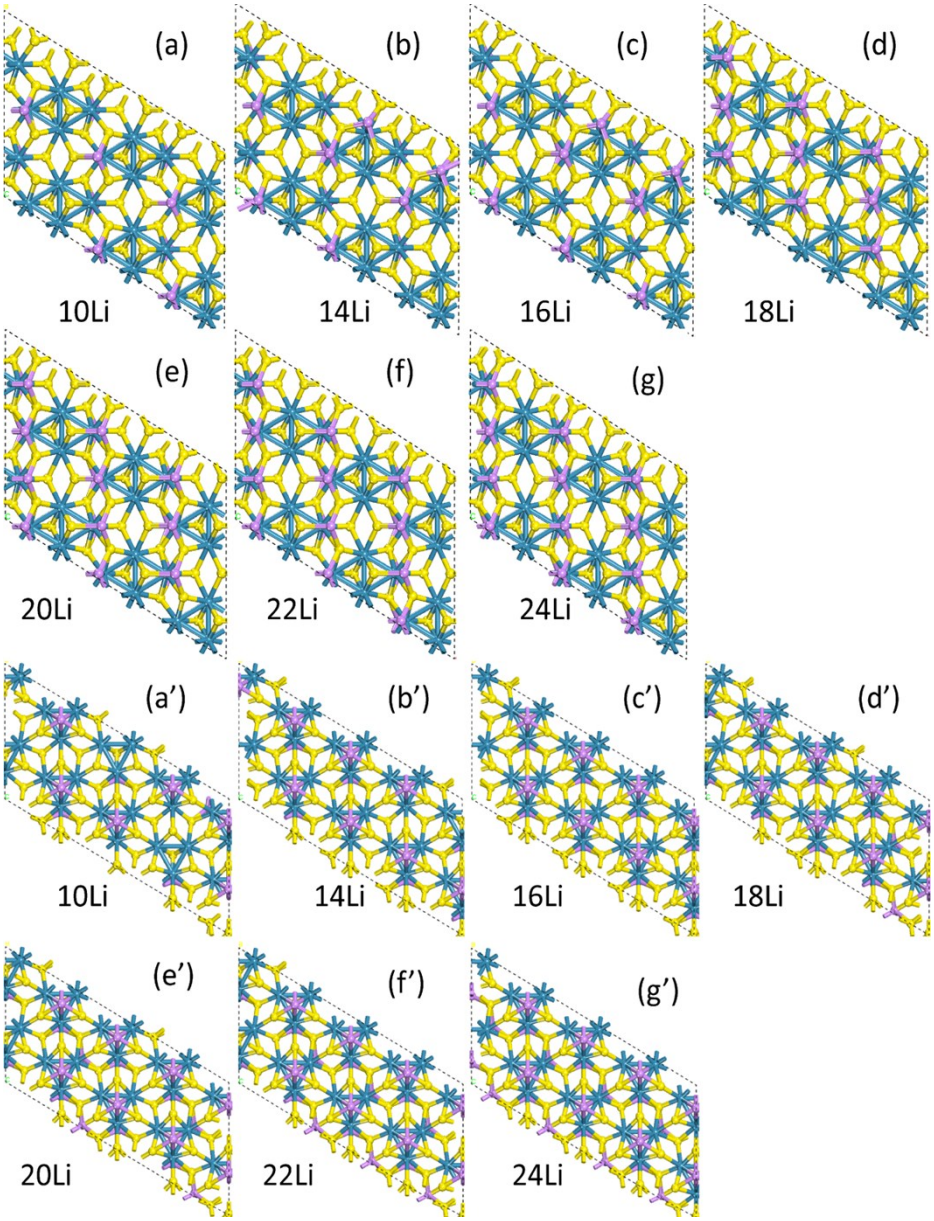

Fig. S7 The adsorption configurations of Li atoms on the 2×3 unit cell DT-1 and 2× 4 unit cell Tri structure about 10Li (a, a'), 14Li(b, b'), 16Li(c, c'), 18Li(d, d'), 20Li(e, e'), 22Li(f, f'), 24Li atoms (g, g')