Theoretical Study of Lifshitz Transition for 2H-TaS_2

Wenxuan Wang[1], Zhenyi Jiang[1]*, Xiaodong Zhang[1]*, Jiming Zheng[1], Hongwei Du[1], Zhiyong Zhang[2]

[1]Shaanxi Key Laboratory for Theoretical Physics Frontiers, Institute of Modern Physics, Northwest University, Xi'an 710069

[2]Stanford Research Computing Center, Stanford University, Stanford, CA 94305, United States

Table S1-1 Lattice parameters (Å) of metal phase for bulk 2H-TaS_2 with various vdW corrections.

	a	*b*	*c*
Tkatchenko-Scheffler	3.310	3.310	12.025
DFT-D2	3.350	3.350	12.025
DFT-D3	3.320	3.320	12.025
OPT-B86b	3.316	3.316	12.025
Exp.	3.316	3.316	12.070

Table S1-2 Lattice parameters (Å) of metal phase for bulk 2H-TaS_2 with various *U* values.

U	*a*	*b*	*c*
0	3.316	3.316	12.025
1	3.338	3.338	12.124
2	3.346	3.346	12.147
3	3.355	3.355	12.170
Exp.	3.316	3.316	12.070

Table S1-3 Lattice parameters *a*/*b*/*c* (Å), in-plane area *S* (Å^2), volume *V* (Å^3), and total energy *E* (eV) of metal and CDW phases for 2H-TaS_2 (Ta_xS_{2x}) and 1H-TaS_2 (Ta_xS_{2x}).

	a	*b*	*c*	*S*	*V*	*E*
bulk metal 2H	9.948	9.948	12.025	85.704	1031.039	-269.662
bulk T-Ts 2H	9.952	9.952	12.013	85.773	1030.484	-269.726
monolayer metal 2H	9.953	9.953		85.790		-321.022
monolayer T-Ts 2H	9.957	9.957		85.859		-321.071
Metal 1H	9.953	9.953		85.790		-159.335
T 1H	9.958	9.958		85.885		-159.382

Table S1-4 Geometrical parameters (Å) of metal and CDW phases for bulk and mono-layer 2H-TaS_2

	d_1	d_2	d_3	d_4	d_5	d_6
bulk metal	1.569	1.569	2.872	1.569	1.569	6.011
bulk T-Ts	1.567	1.567	2.876	1.563	1.563	6.006
monolayer metal	1.572	1.563	2.877	1.563	1.572	6.005
monolayer T-Ts	1.570	1.560	2.882	1.558	1.567	6.001

Table S1-5 Formation energy ΔE_{CDW} (meV/TaS_2) of various possible CDW images for mono-layer 1H-TaS_2

Structure	$H^{(3,5,8,9,10)}$	$S^{(3,6,10)}$	$Ts^{(4,5,7,10)}$	$T^{(4\text{-}7,10)}$	$C^{(1,4)}$	$R^{(3)}$	$T'^{(1,2,4)}$
ZPE(eV)	1.100	1.112	1.099	1.094	1.099	1.099	1.094
ΔE_{CDW} (meV) without ZPE	-3.500	-1.700	-3.700	-4.400	-3.700	-3.700	-4.400
ΔE_{CDW} (meV) with ZPE	-2.500	-1.400	-2.800	-4.100	-2.800	-2.800	-4.100

[1] Physical Review B, 105(2022), L180505.

[2] Physical Review B, 107(2023), 045431.

[3] ACS Nano, 15(2021), 18297-18304.

[4] Nano Letters, 19(2019), 3027-3032.

[5] Physical Review B, 98(2018), 195419.

[6] Physical Review B, 97(2018), 081101.

[7] Nano Letters, 18(2018), 2924-2929.

[8] Physical Review B, 104(2021), 115136.

[9] Physical Review B, 87(2013), 094502.

[10] Nano Letters, 22(2022) 1858-1865.

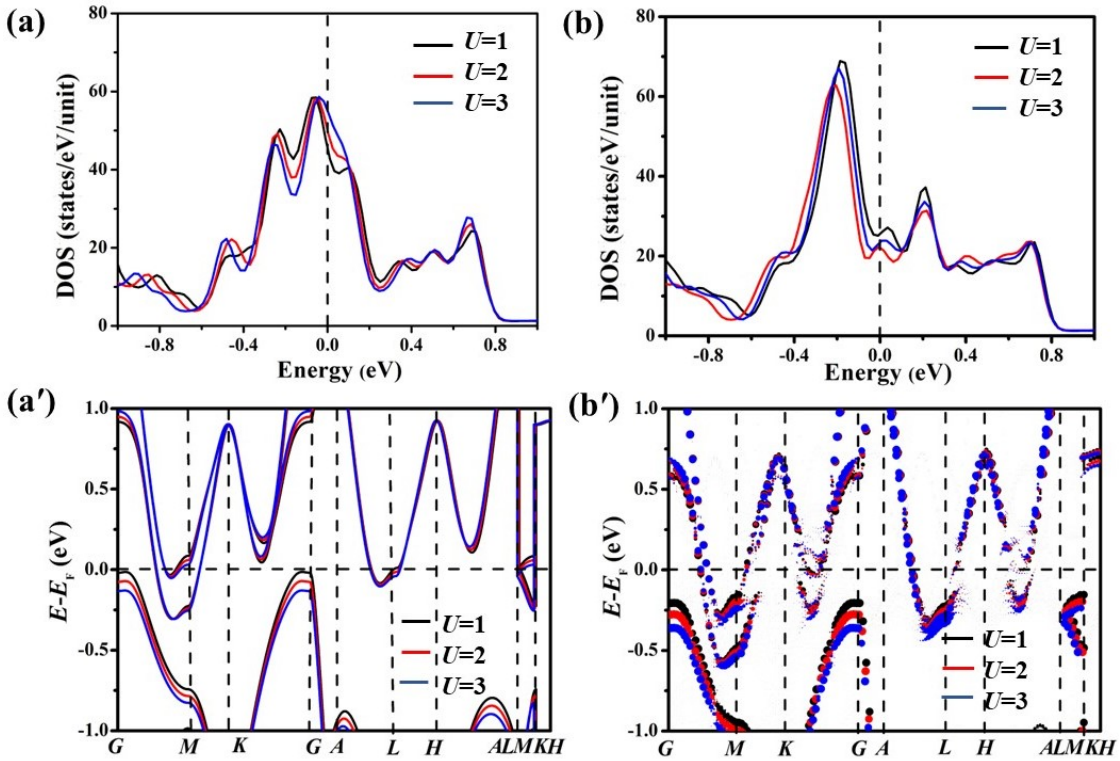

Figure S1-1 DOS and Energy band with various U value for bulk 2H-TaS_2. (a-a′) for metal phase, and (b-b′) for CDW phase.

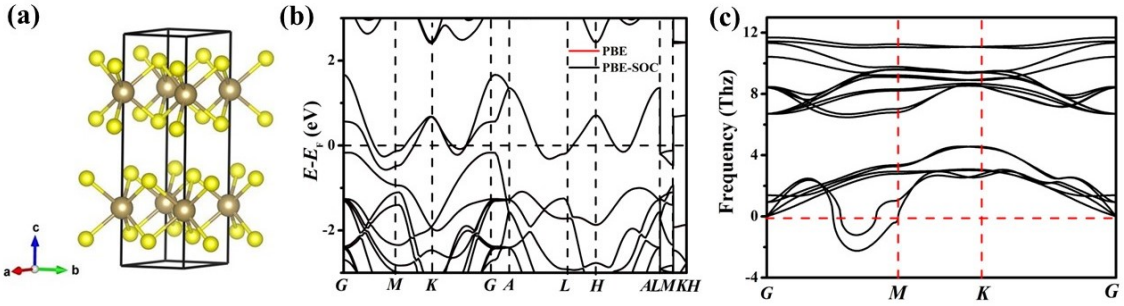

Figure S1-2 (a) Unit cell of metal phase for bulk 2H-TaS_2. Brown spheres represent tantalum atoms while the others represent oxygen atoms. (b) Energy band of metal phase for bulk 2H-TaS_2 with SOC and without SOC. (c) Phonon spectrum of metal phase for bulk 2H-TaS_2.

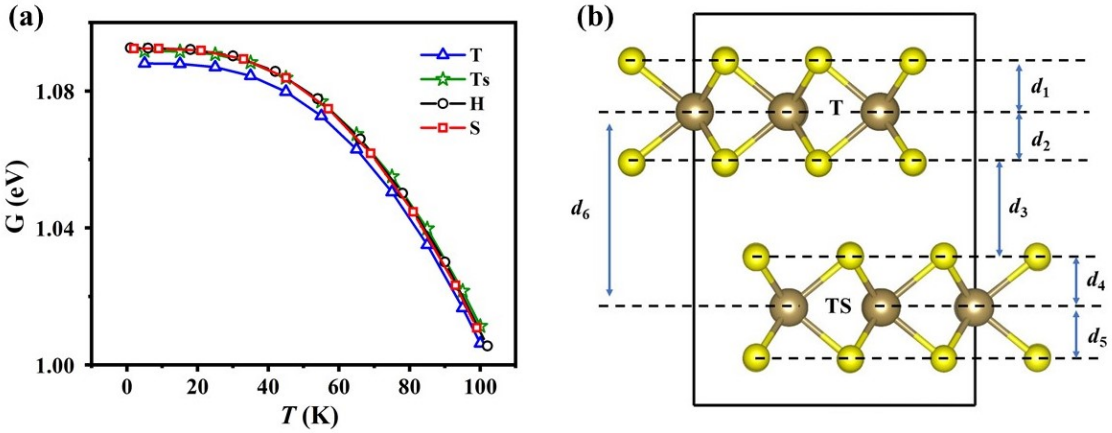

Figure S1-3 (a) Temperature dependence of Gibbs free energy for H, S, T, and Ts configurations, (b) Schematic diagram of atomic separation for CDW and metal phase for two-layer and bulk 2H-TaS_2,

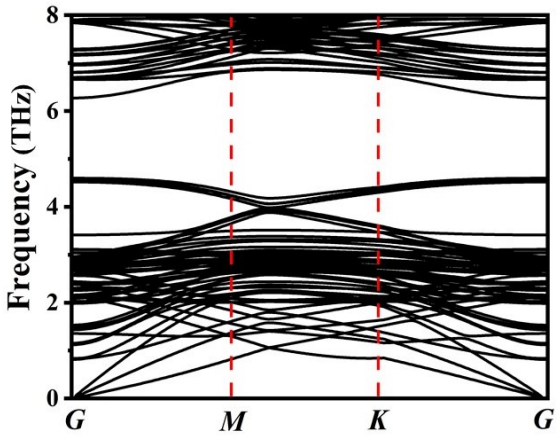

Figure S1-4 (a) Phonon dispersion of CDW phase.

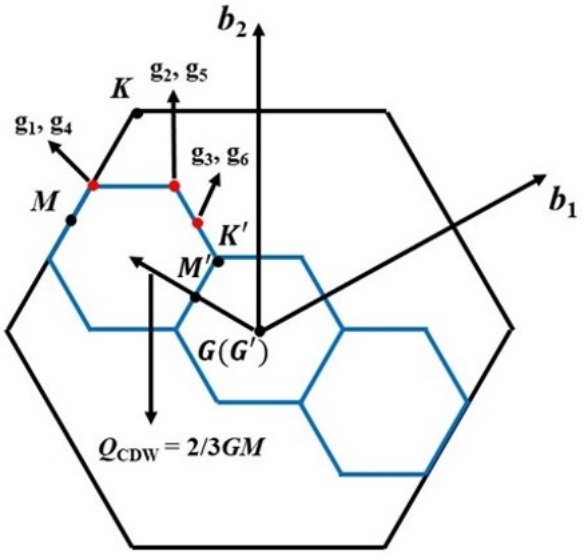

Figure S1-5 BZ of hexagonal 2H-TaS_2 lattice. The black line represents the undistorted BZ and bule line represents the BZ of CDW phase.

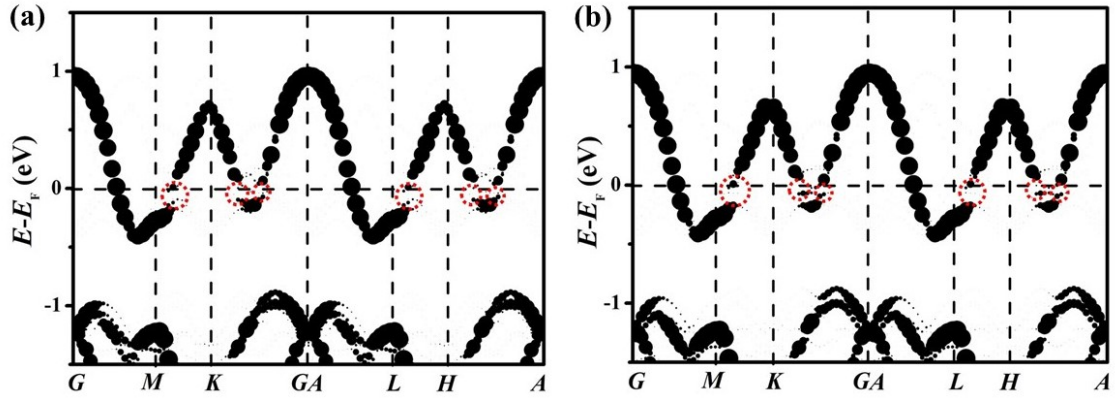

Figure S1-6 Unfolding energy band T (a) and Ts (b) configuration for mono-layer 1H-TaS_2.

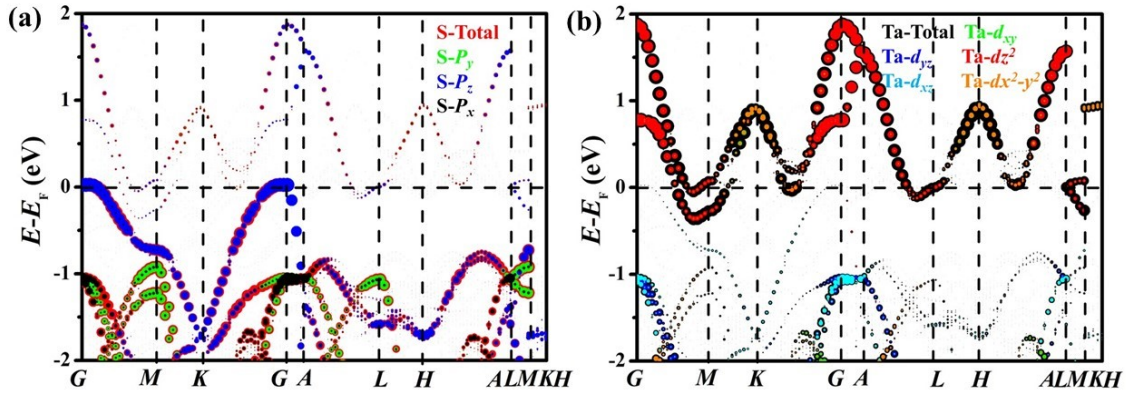

Figure S1-7 Schematic diagram of the contribution of sulfur atoms to various energy bands (a), and the contribution of tantalum atoms to various energy bands (b).

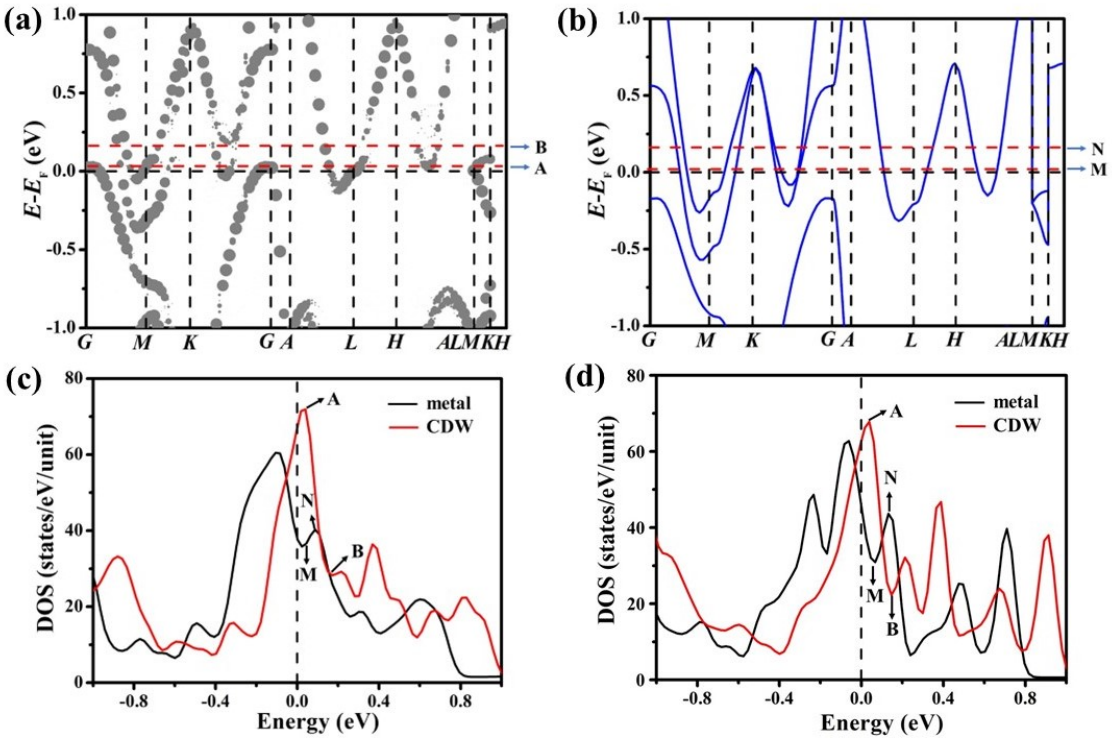

Figure S1-8 Energy band and DOS of metal and CDW phase for bulk 2H-TaS_2. (a) Unfolding energy band of CDW phase. (b) Energy band of metal phase. (c) Projected in-plane DOS. (d) Projected out-of-plane DOS.

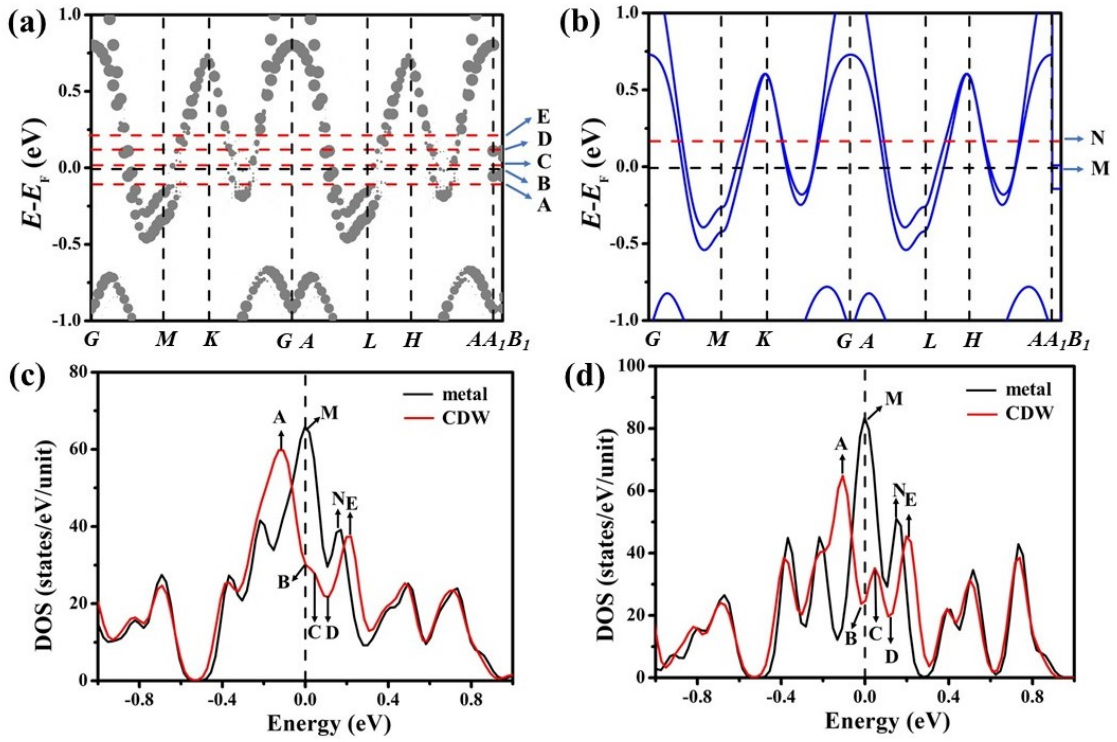

Figure S1-9 Energy band and DOS of metal and CDW phase for mono-layer 2H-TaS_2. (a) Unfolding energy band of CDW phase. (b) Energy band of metal phase. (c) Projected in-plane DOS. (d) Projected out-of-plane DOS.