

Supplementary Material: High-throughput calculations of spin Hall conductivity in non-magnetic 2D materials

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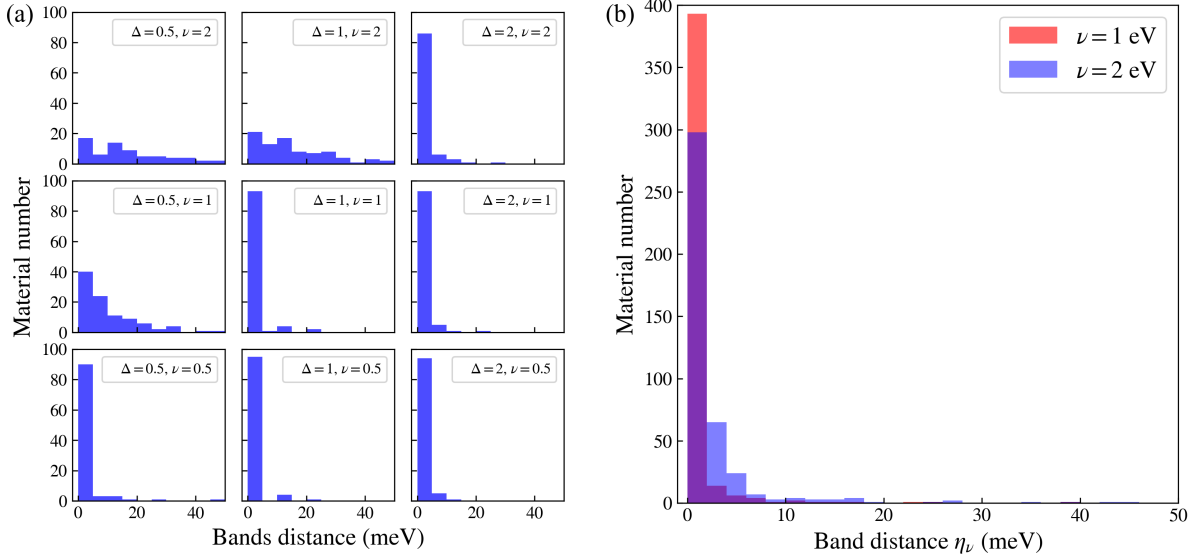
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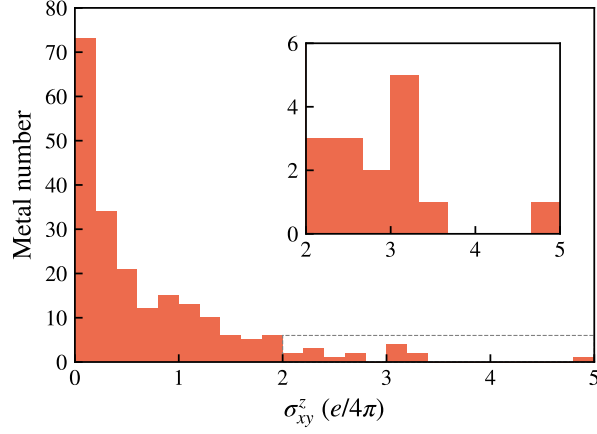
Supplementary Sec. 1. Test on the frozen window maximum

The test of the maximum of frozen window is given based on 100 materials which were arbitrarily chosen, including 60 semiconductors and 40 metals. The DFT bands and Wannier bands generated with different frozen window maxima are compared using the band distance, which is defined by Eq. (4) in the main text. As demonstrated by [Supplementary Fig. 1\(a\)](#), $E_F + 2$ eV gives the optimal result especially for the high-energy bands. The band distance of all the 426 materials is presented in [Supplementary Fig. 1\(b\)](#). Most materials present a small band distance below 2 meV.



Supplementary Fig. 1. **Statistic results of band distance.** (a) Impact of the maximum of frozen window based on 100 randomly selected materials. In the Wannierization, the maximum of the frozen windows is given by $E_F + \Delta$ (unit: eV) where $\Delta = 0.5, 1$, and 2 , respectively. The band distance below $E_F + \nu$ where $\nu = 0.5, 1$, and 2 are respectively evaluated. For insulators, E_F is defined as the conduction band minimum. (b) Distribution of band distances for all the 426 monolayers using a frozen window of $\Delta = 2$ eV.

Supplementary Sec. 2. Metals with high SHC



Supplementary Fig. 2. **Spin Hall conductivities of 210 monolayer metals.** The inset shows the region where 15 materials presents high spin Hall conductivities of $|\sigma_{xy}^z| > 2 e/4\pi$. These 15 materials are reported in [Supplementary Table 1](#).

Supplementary Table 1. **15 monolayer metals with $|\sigma_{xy}^z| > 2 e/4\pi$.**

Monolayer	Space group	$ \sigma_{xy}^z (e/4\pi)$
BiTe ₂	P-3m1	4.88
LiMnTe ₂	P3m1	3.39
NaMnTe ₂	P3m1	3.24
AgCuTe ₂	Pmm2	3.14
Y ₂ Cl ₂	P-3m1	3.14
LiMnSe ₂	P3m1	3.11
Ta ₄ Se ₂	P4/nmm	3.08
Sn ₂ P	P-3m1	2.75
Co ₂ S ₂	P4/nmm	2.71
Sb ₂ Te ₂	P-3m1	2.55
Ba ₂ Ni ₃	P-3m1	2.39
Y ₂ Br ₂	P-3m1	2.38
In ₂ Te ₃	P-3m1	2.31
CoBr ₂	P-3m1	2.01
Tl ₂ I ₂ S ₂	P4/nmm	2.00

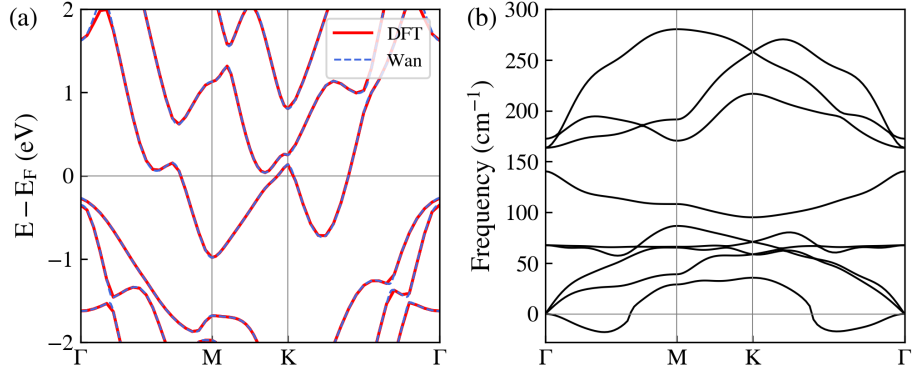
Supplementary Sec. 3. Electronic structures and phonon dispersions of promising materials

Electronic structures and phonon dispersions of 8 materials with superior SHC properties are detailed here. For all the materials, the reproduction of DFT results illustrates the high-quality Wannierization. The absence of imaginary frequency suggests mechanical stability. The Sn_2P and Y_2Br_2 monolayers are discussed in more depths since they required special treatments.

The Sn_2P monolayer with the P-3m1 space group presents negative frequencies around Γ as shown in [Supplementary Fig. 3](#), which is consistent with Ref. [1]. To eliminate the negative frequencies, the structure of Sn_2P monolayer was relaxed without any symmetry. To obtain initial structures for relaxation, 10 structures randomly displaced for the equilibrium position were generated with the P1 space group. Then their total energies were calculated, 3 of them present the same total energy minima, and only 1 structure presents a positive phonon dispersion. This relaxed structure is given in [Supplementary Table 2](#). Electronic structure and phonon dispersion of this low-symmetry structure are depicted in [Supplementary Fig. 4](#). The comparison between [Supplementary Fig. 3\(a\)](#) and [Supplementary Fig. 4\(a\)](#) illustrates that the change of electronic structure is negligible. However, the broken symmetry is critical to obtain a positive phonon dispersion as shown in [Supplementary Fig. 4\(b\)](#). This low-symmetry Sn_2P monolayer is the material reported in the main text.

We now focus on Y_2Br_2 . The possible magnetism of Y_2Br_2 has been considered since Y_2Cl_2 was predicted to be a ferromagnet [2]. Due to the self-interaction error in exchange-correlation functional, a Hubbard correction was included with magnetism. The *ab initio* Hubbard parameter $U = 1.16$ eV computing using density-functional perturbation theory and atomic projectors [3]. As shown in [Supplementary Fig. 5](#) and [Supplementary Fig. 6](#), both non-magnetic and ferromagnetic phases present positive phonon dispersions. However, apart from the mechanical stability, the thermal stability of magnetism should also be checked. The magnetic anisotropy is critical for the ferromagnetism in 2D material since it could stabilize the long-range magnetic order by opening up an excitation gap to resist the thermal agitation [4]. Defined as the energy difference between in-plane and out-of-plane magnetic configurations considering spin-orbital coupling, the magnetic anisotropic energy (MAE) of ferromagnetic Y_2Br_2 was calculated to be 0.03 meV. Two well-known 2D magnets can be taken as reference: CrI_3 monolayer with an MAE of 0.65 meV and a Curie temperature of 45 K [5], and MnBi_2Te_4 monolayer with an MAE of 0.15 meV with a Curie temperature of 20 K [6, 7]. With a much smaller MAE, the ferromagnetism of Y_2Br_2 is easily vulnerable to the thermal agitation, illustrating the difficulty of maintaining ferromagnetism at room temperature. As a result, Y_2Br_2 is expected to be non-magnetic at room temperature, and the non-magnetic configuration has been employed in the main text discussion.

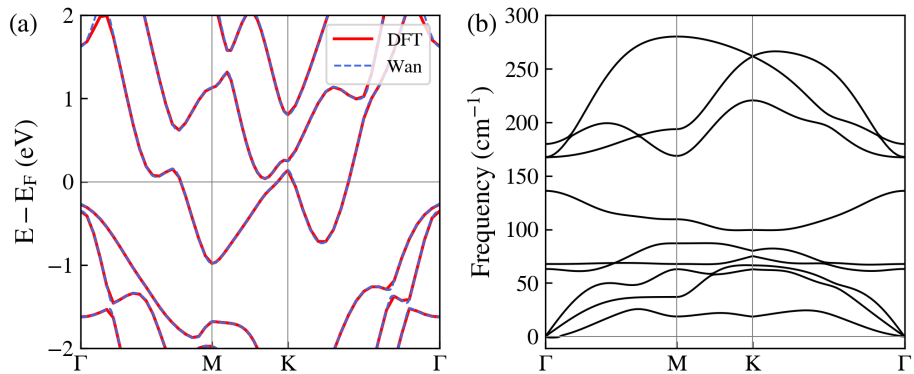
Finally, [Supplementary Fig. 7-Supplementary Fig. 12](#) show the electronic structures with SOC including DFT and Wannier bands and phonon dispersion without SOC.



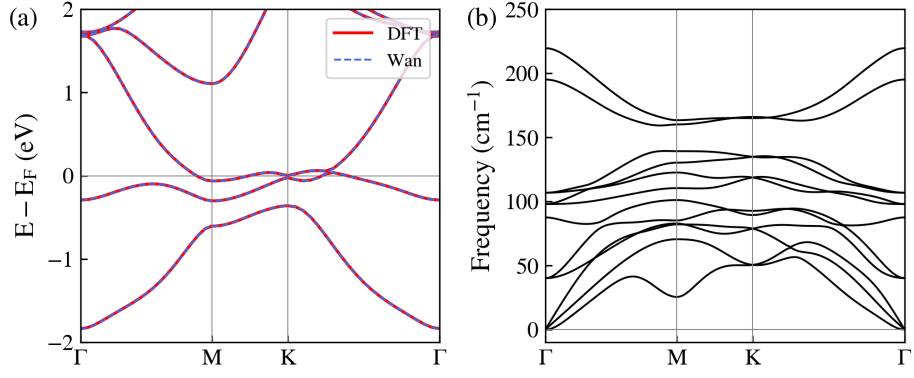
Supplementary Fig. 3. **Electronic structure and phonon dispersion of the high-symmetry Sn_2P monolayer.**

Supplementary Table 2. **Lattice vectors and atomic positions of the stable low-symmetry Sn_2P monolayer.**

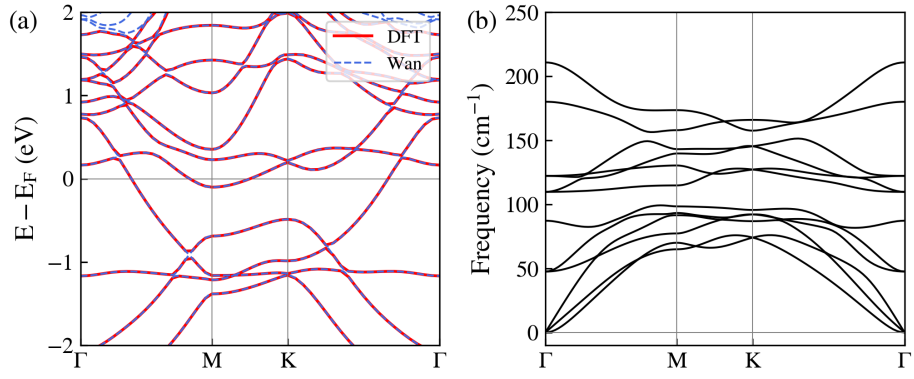
Lattice vectors	x (Å)	y (Å)	z (Å)
$\mathbf{a}_1$	3.6891078804627813	-0.0000036894473096	0.0000000000000000
$\mathbf{a}_2$	-1.8445281141002232	3.1948732083140299	0.0000000000000000
$\mathbf{a}_3$	0.0000000000000000	0.0000000000000000	23.1653976932423795
Atomic positions	x (Å)	y (Å)	z (Å)
Sn	0.1155841121498199	2.0834497119210194	2.1794277321228130
P	1.9600579913745917	1.0184036750440661	0.4381801612843048
Sn	0.1154896622986938	-0.0464530931859039	-1.3030736816146300



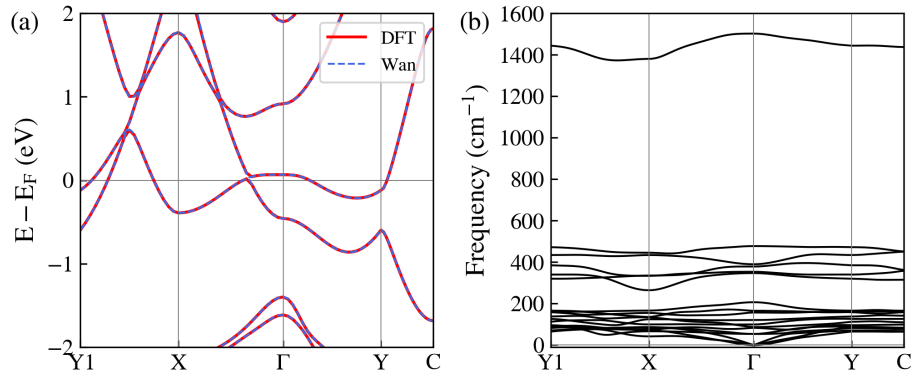
Supplementary Fig. 4. **Electronic structure and phonon dispersion of the low-symmetry Sn_2P monolayer.**



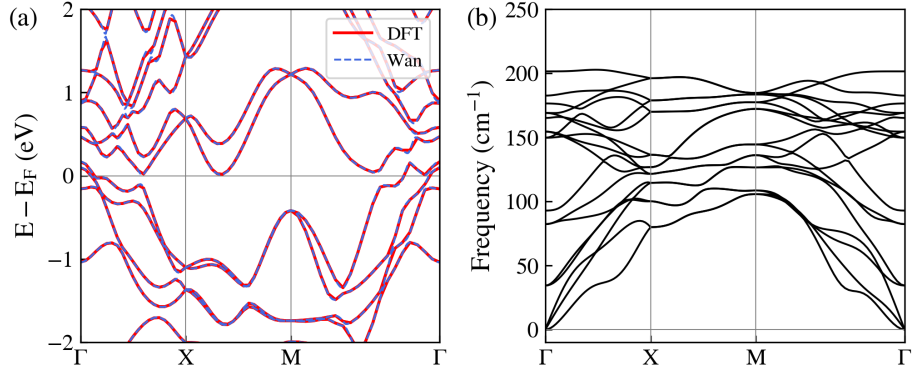
Supplementary Fig. 5. **Electronic structure and phonon dispersion of the non-magnetic Y_2Br_2 monolayer.**



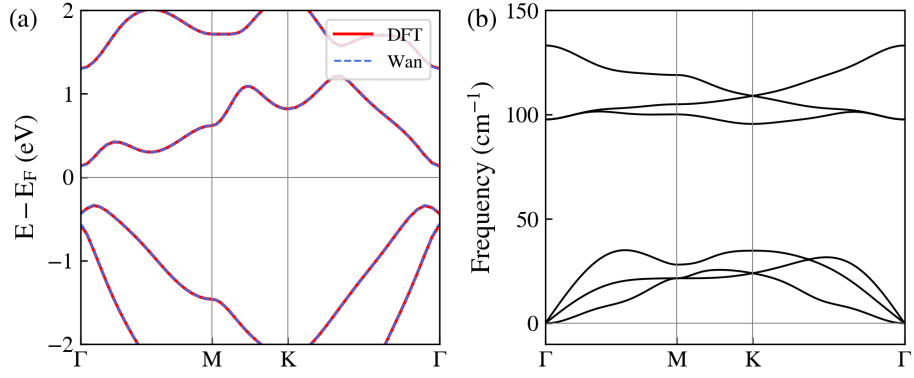
Supplementary Fig. 6. **Electronic structure and phonon dispersion of the ferromagnetic Y_2Br_2 monolayer.**



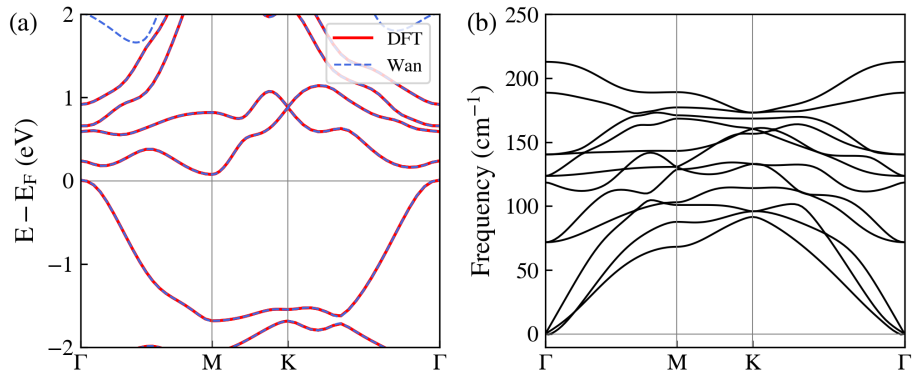
Supplementary Fig. 7. **Electronic structure and phonon dispersion of the $\text{Y}_2\text{C}_2\text{I}_2$ monolayer.**



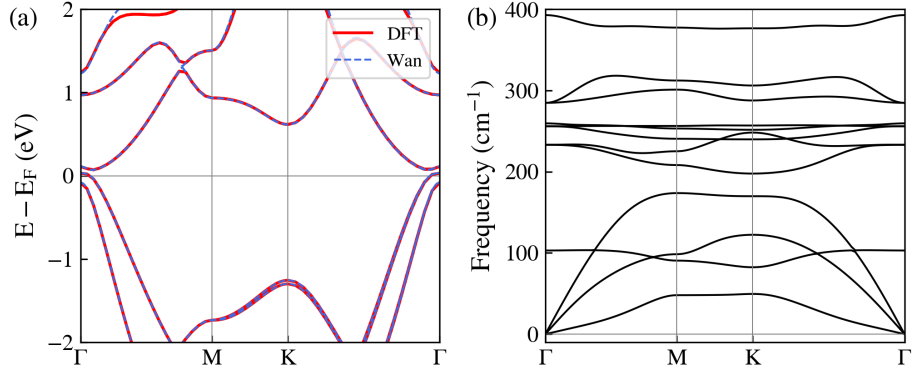
Supplementary Fig. 8. **Electronic structure and phonon dispersion of the Ta_4Se_2 monolayer.**



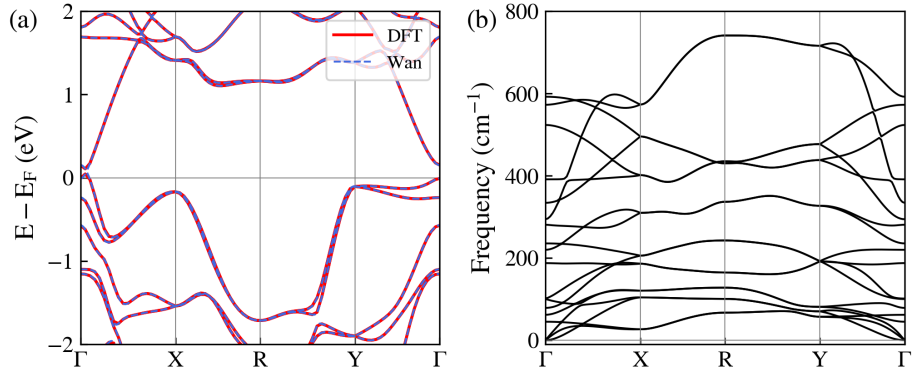
Supplementary Fig. 9. **Electronic structure and phonon dispersion of the Bi_2 monolayer.**



Supplementary Fig. 10. **Electronic structure and phonon dispersion of the Hf_2Br_2 monolayer.**

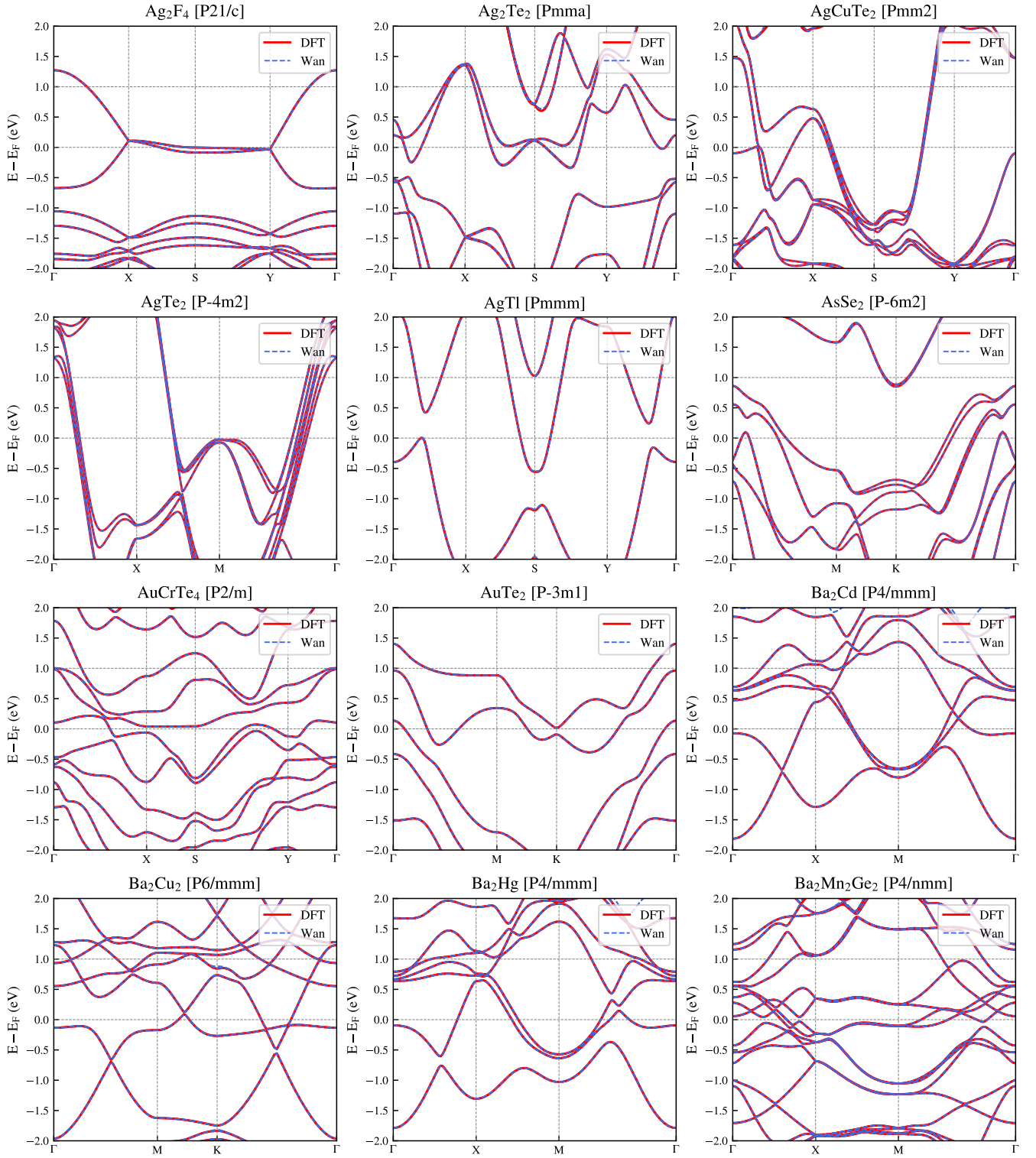


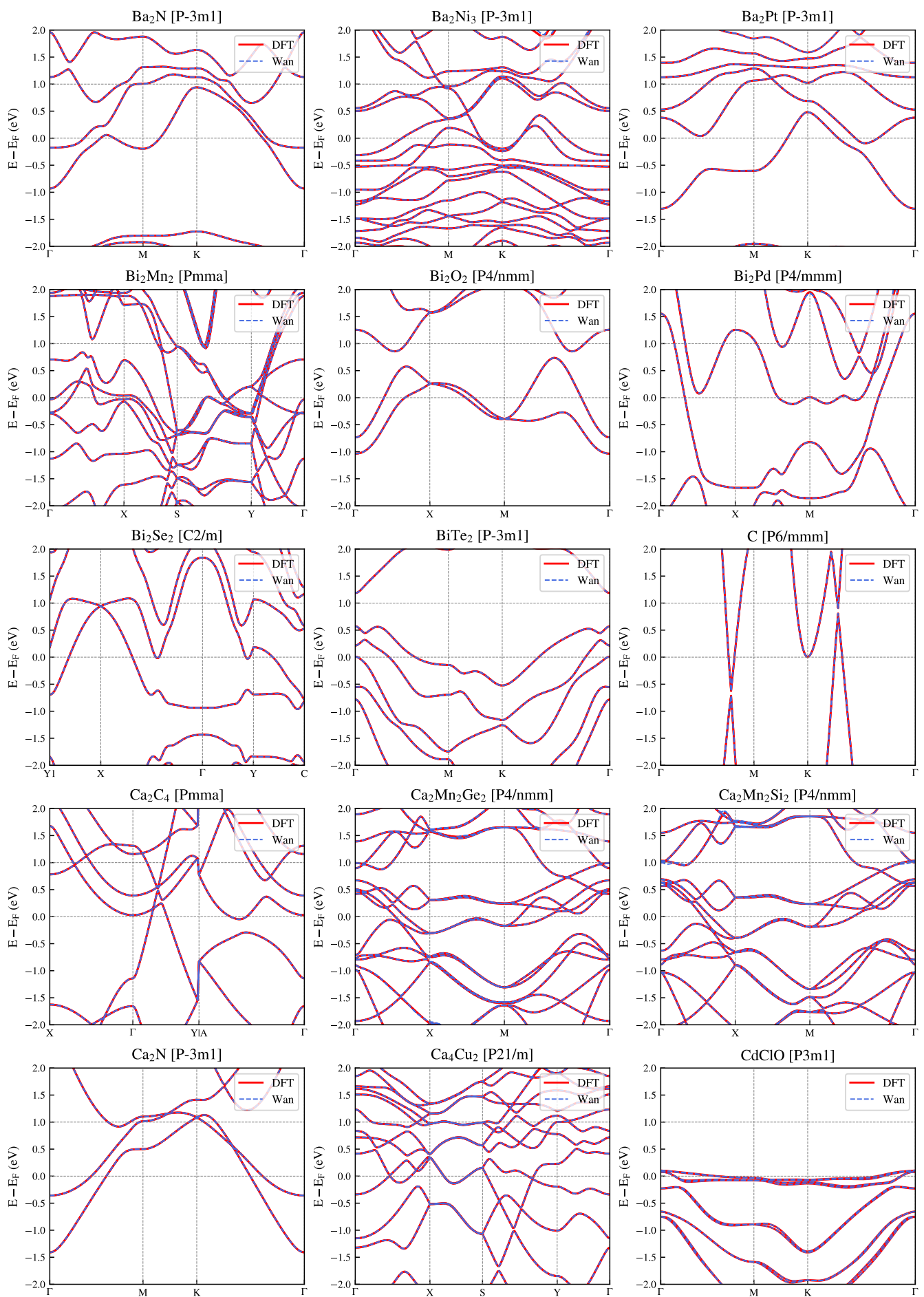
Supplementary Fig. 11. **Electronic structure and phonon dispersion of the CuLi₂As monolayer.**

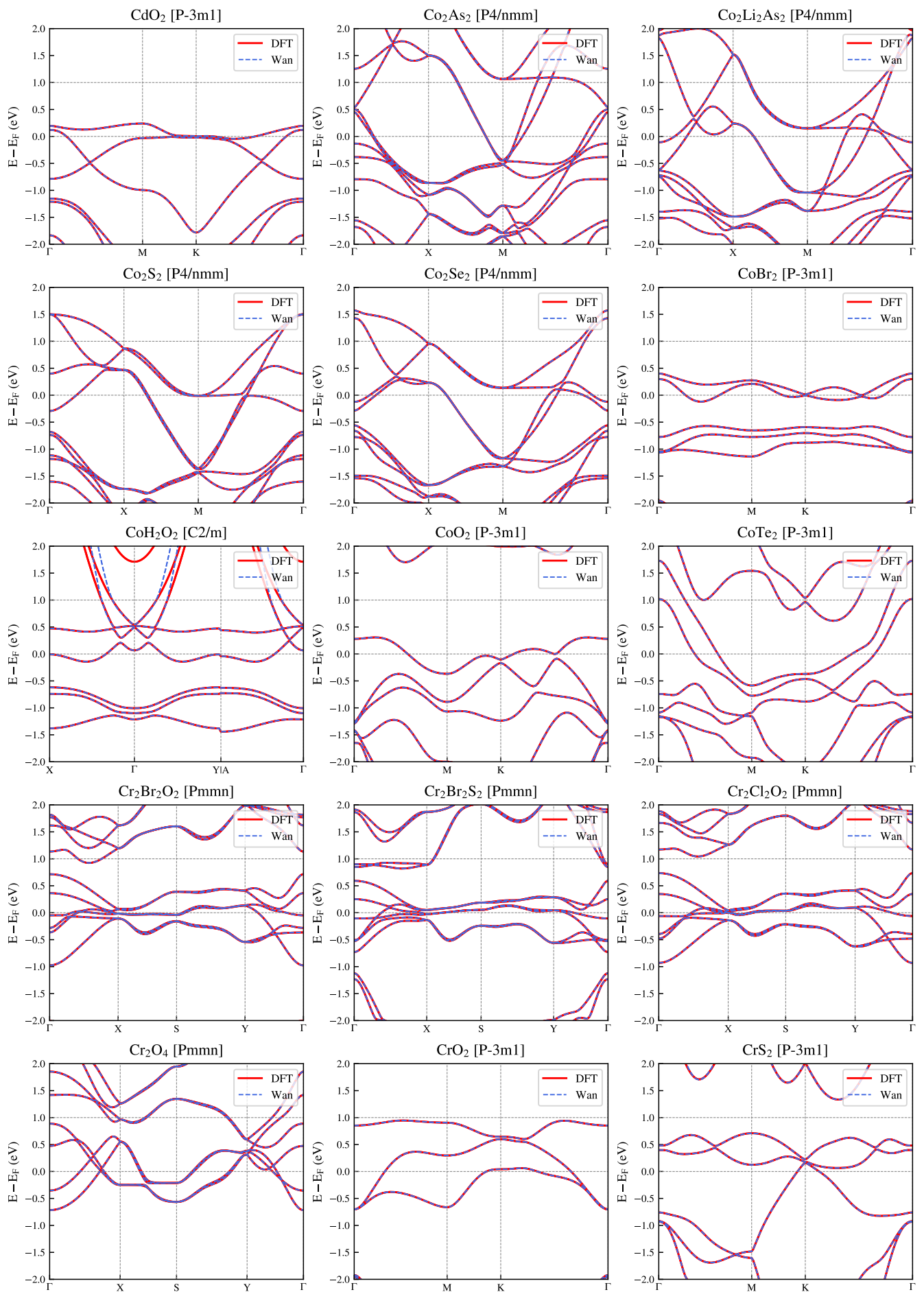


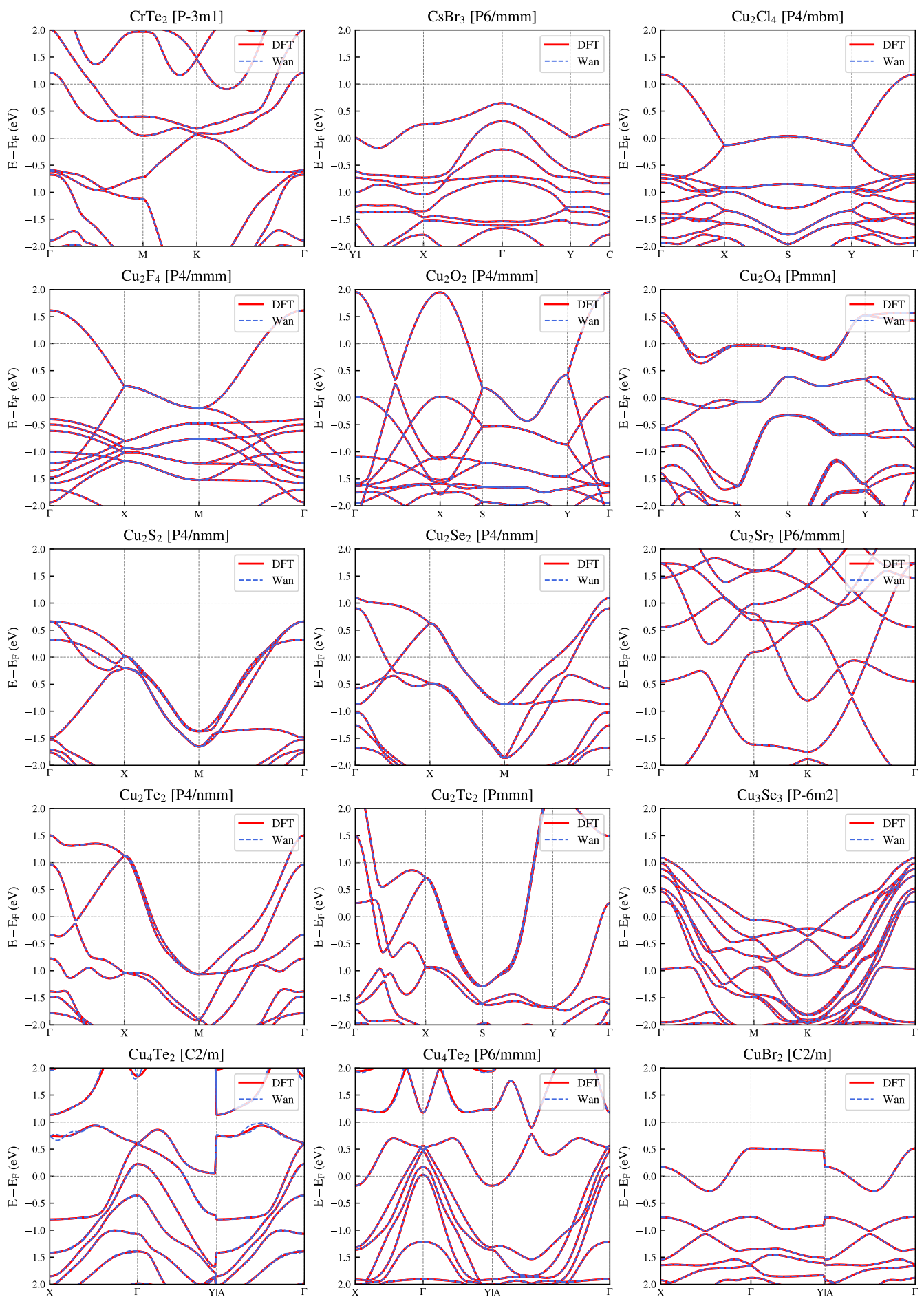
Supplementary Fig. 12. **Electronic structure and phonon dispersion of the Ti₂N₂I₂ monolayer.**

Supplementary Sec. 4. Fully relativistic electronic structures of 210 monolayer metals

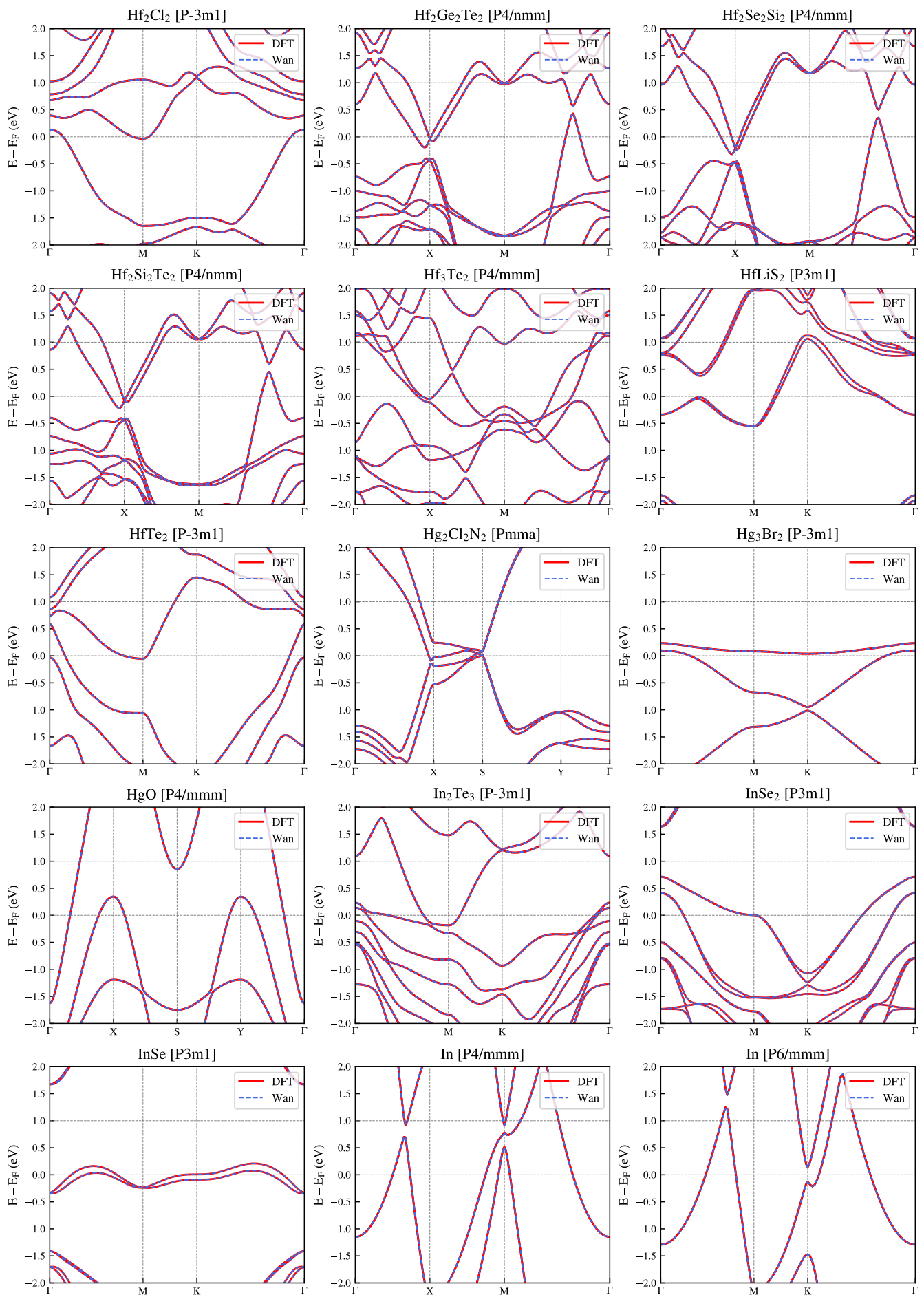


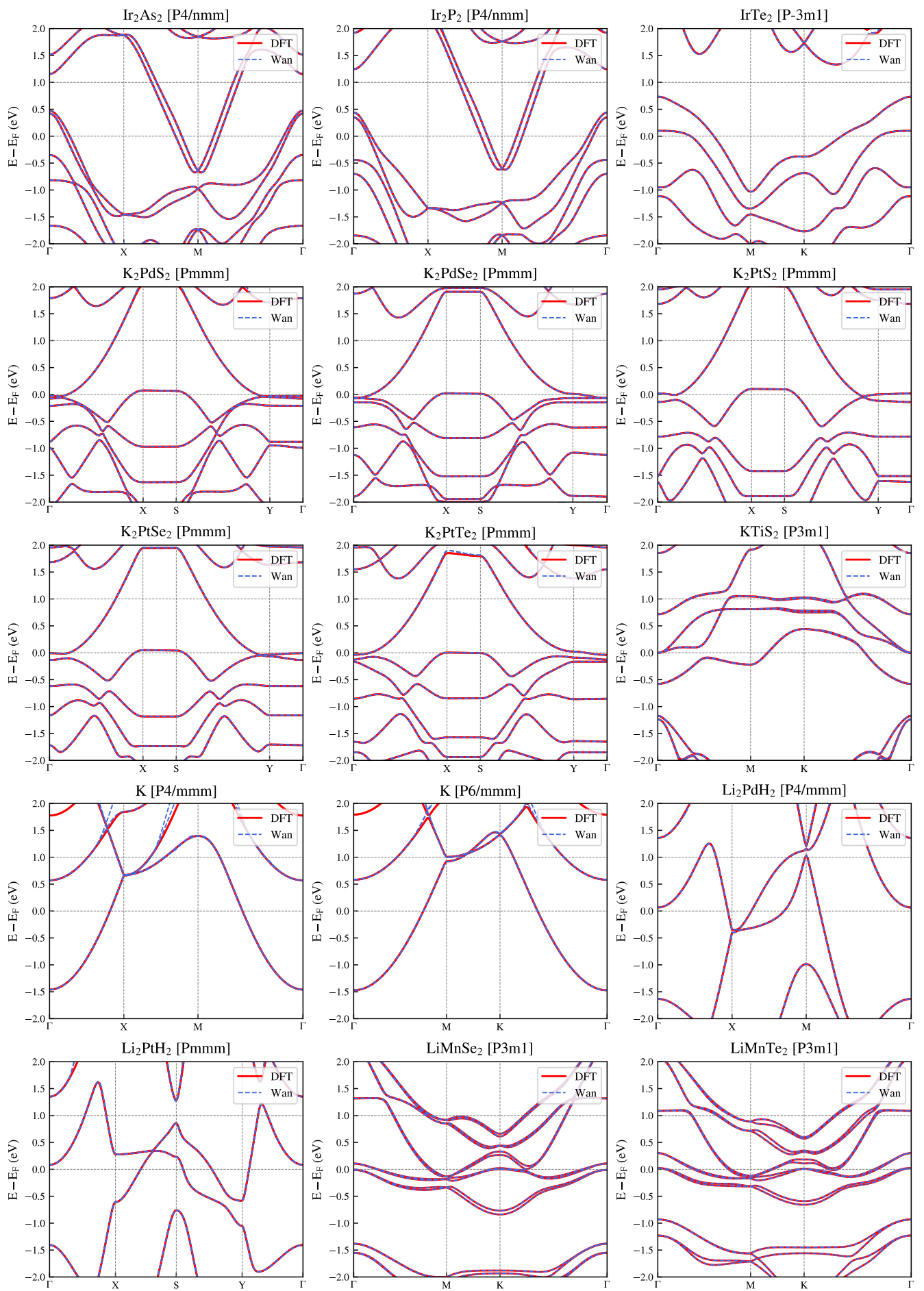


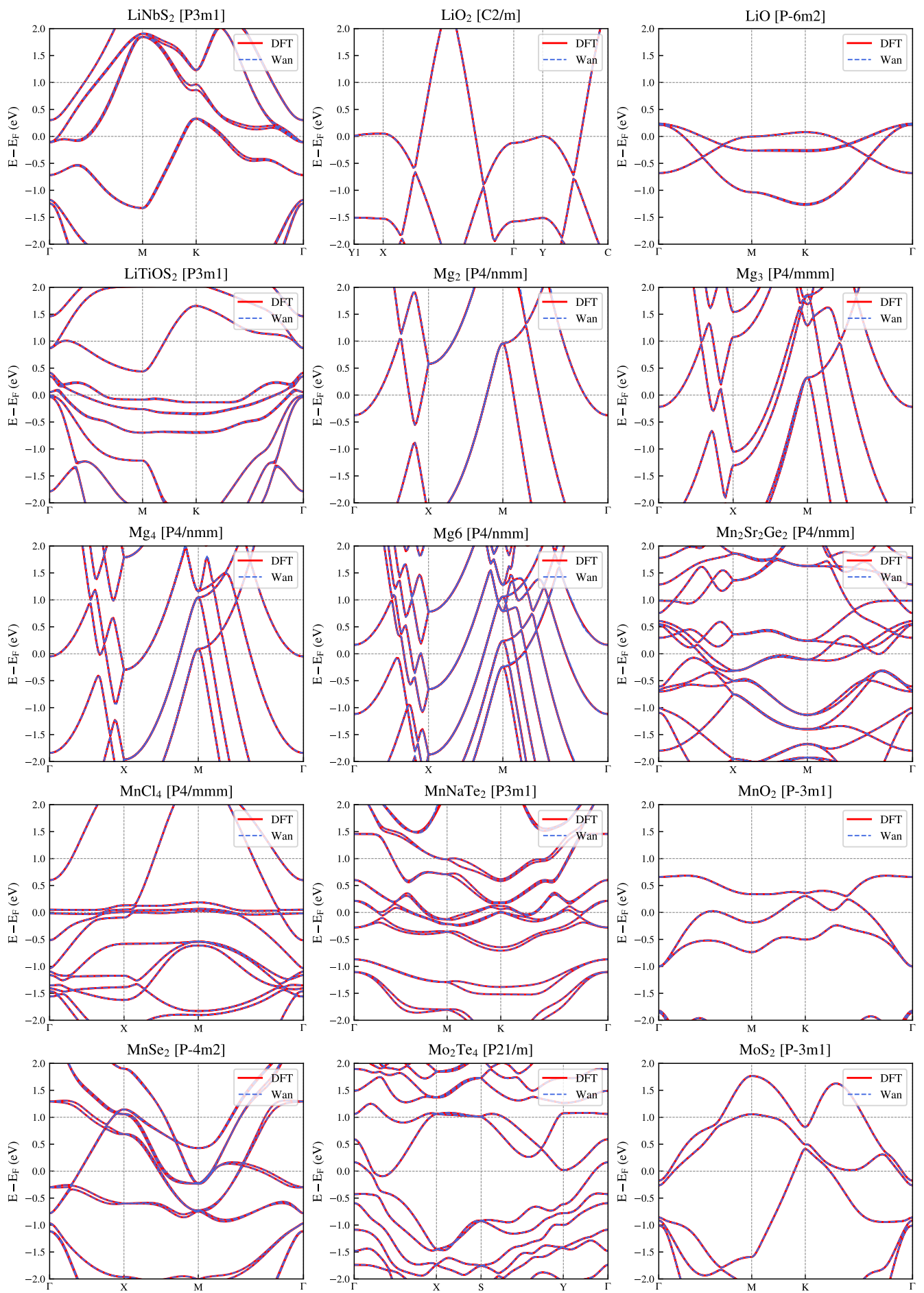


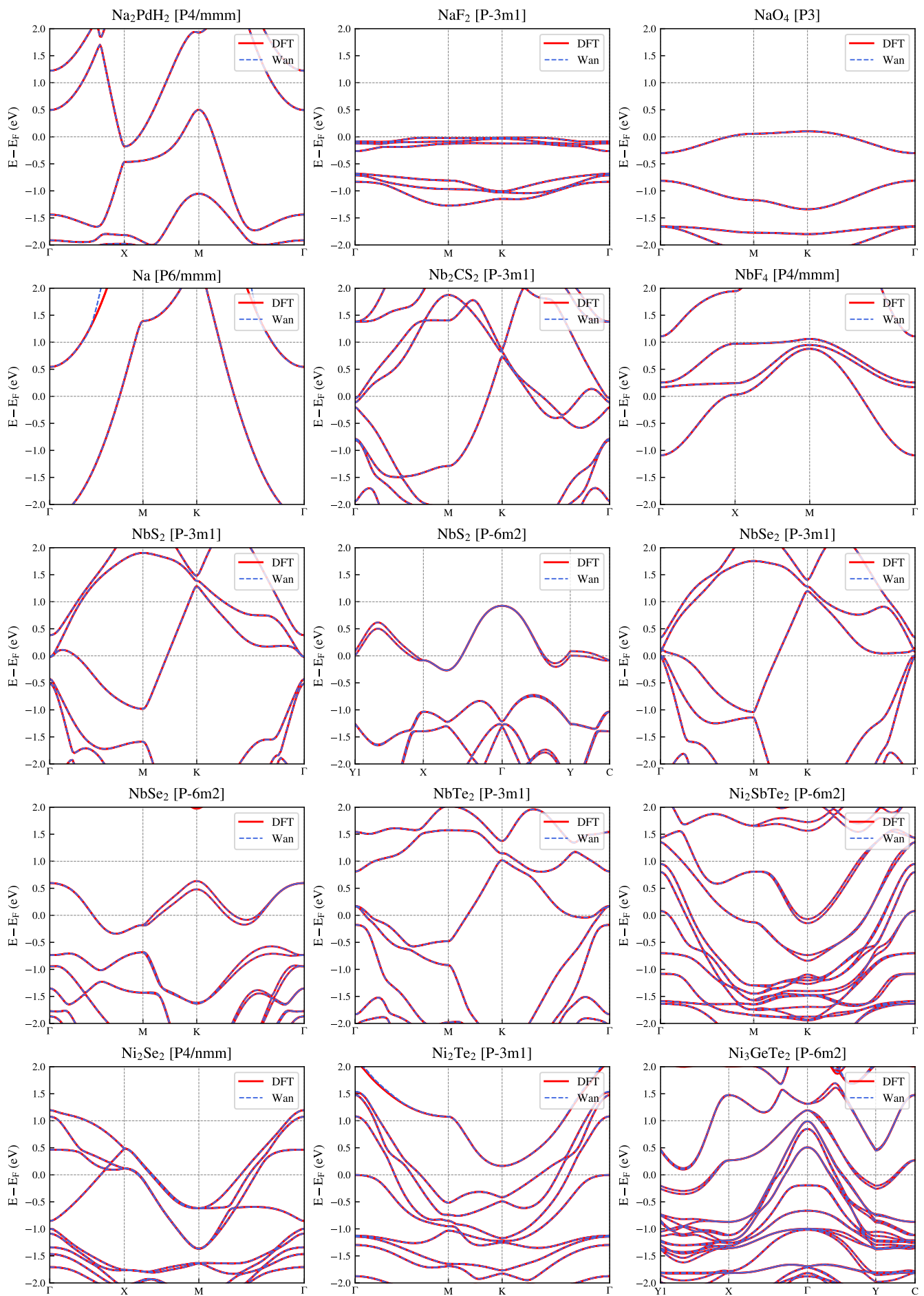


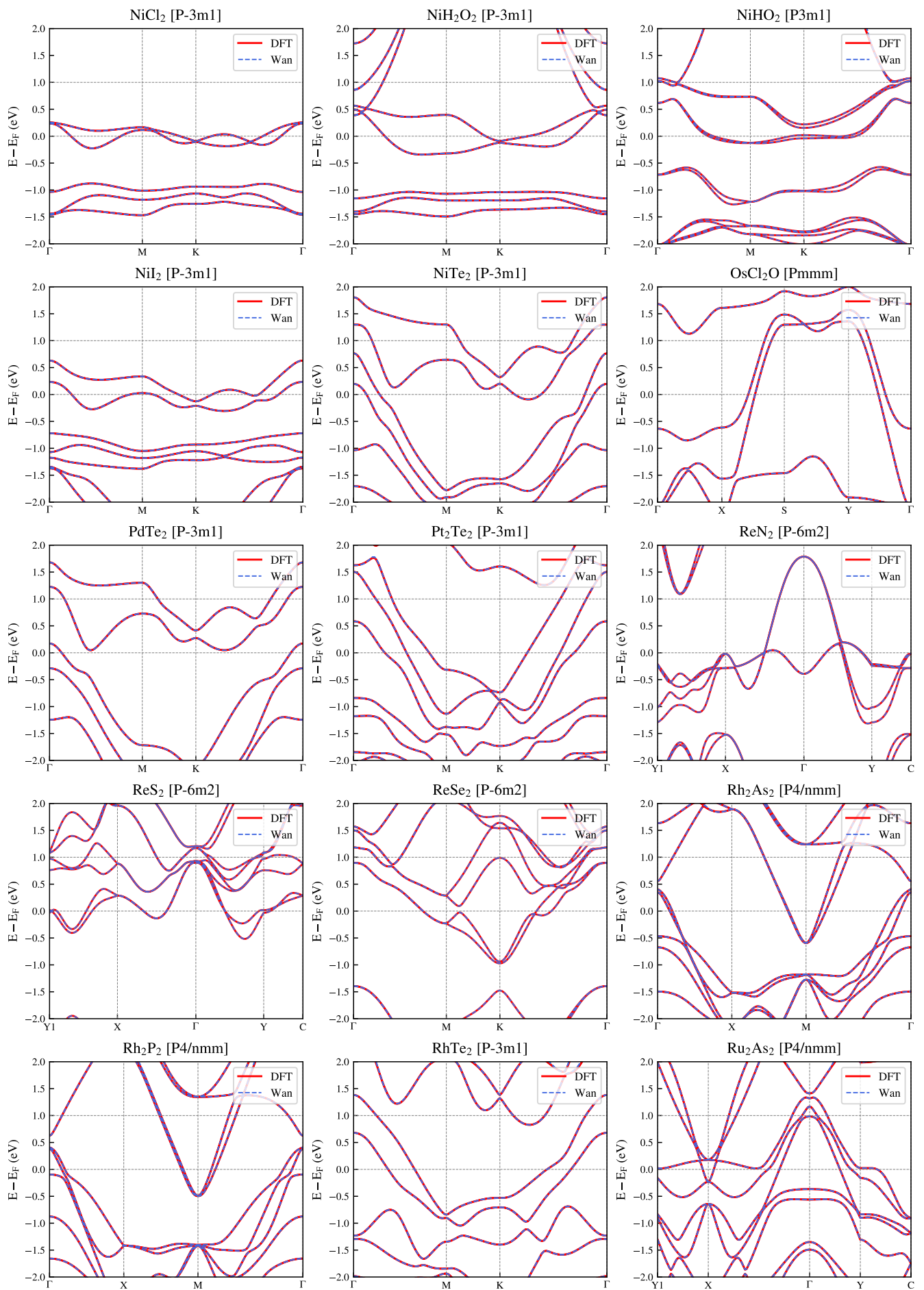


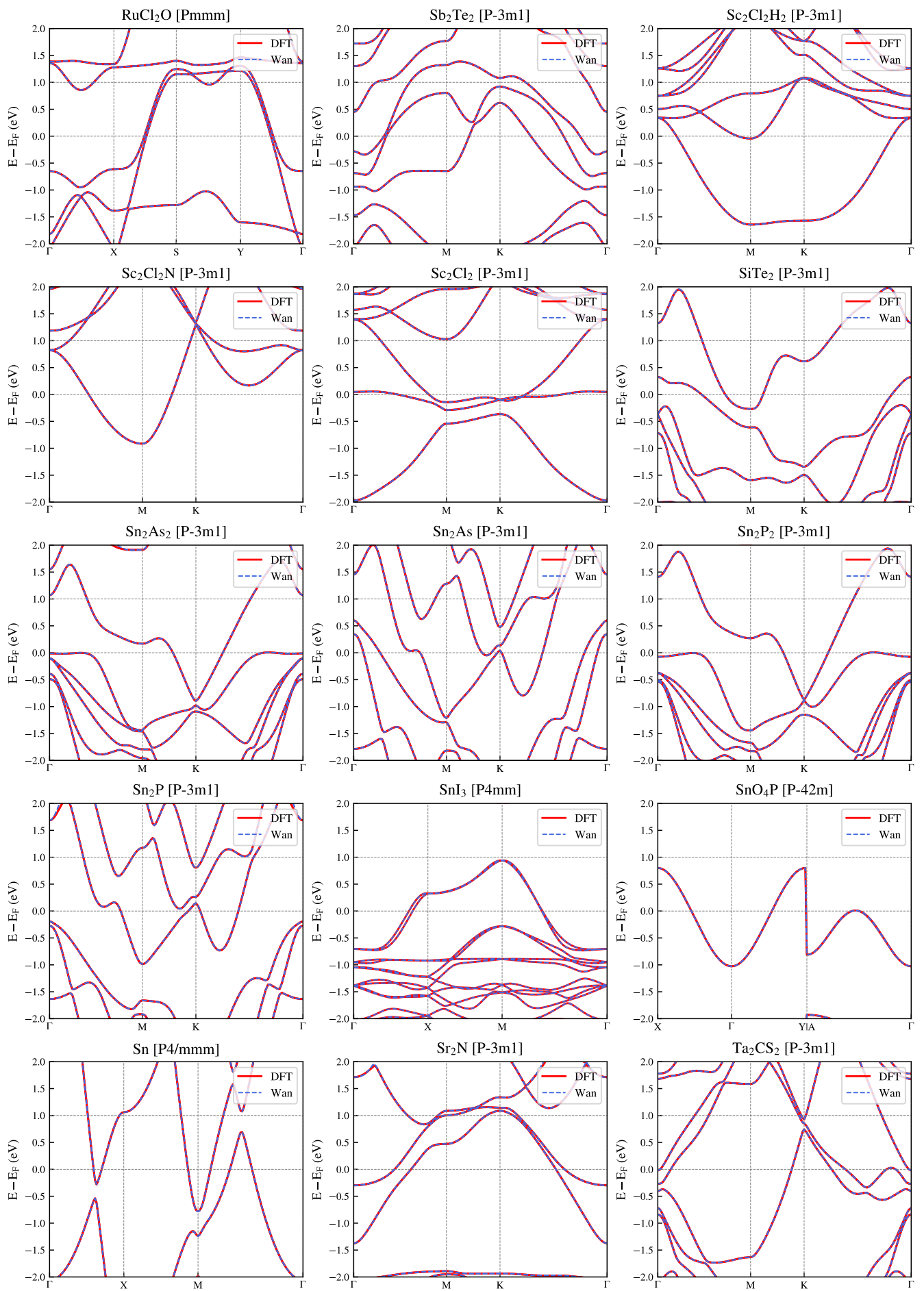


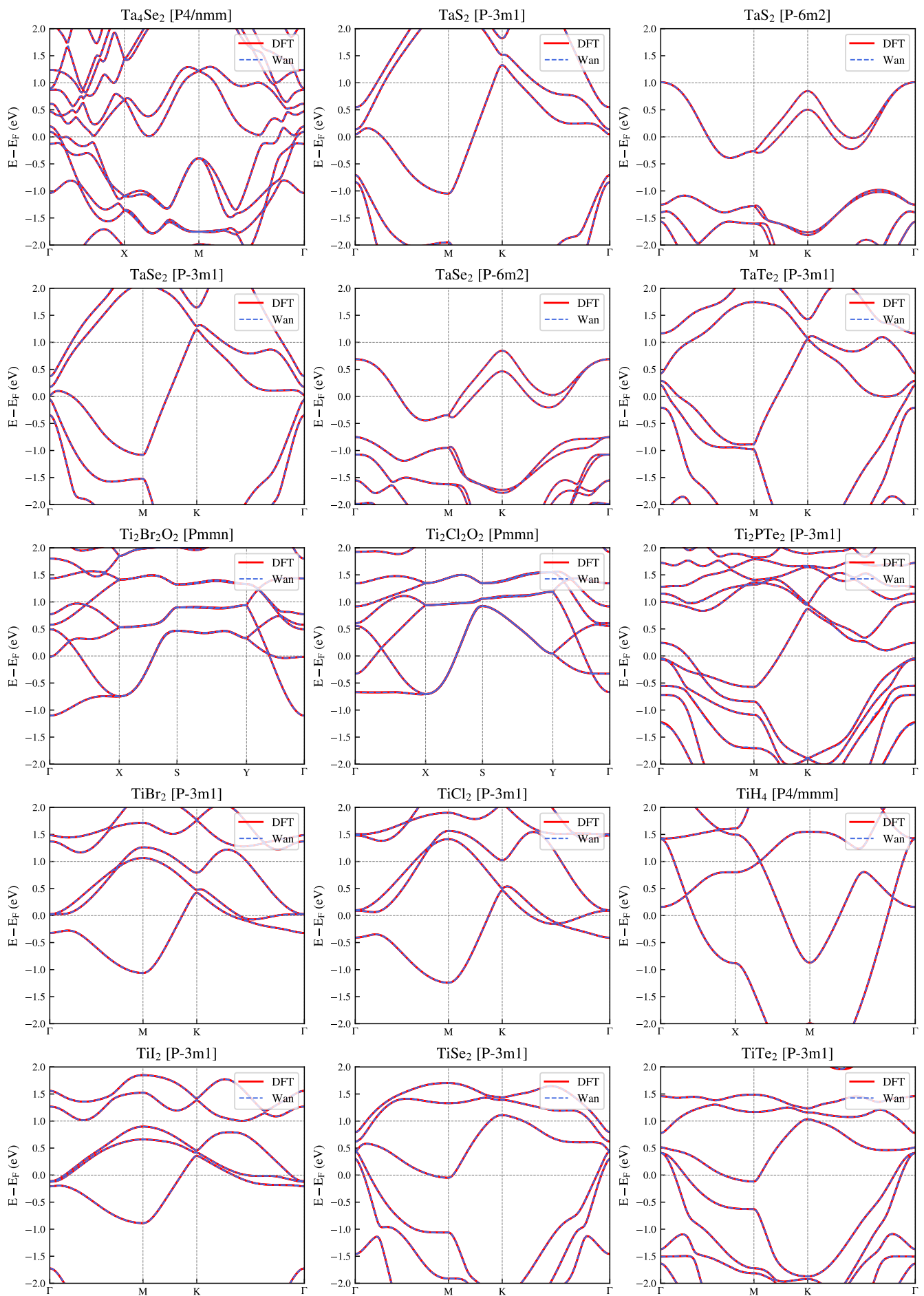


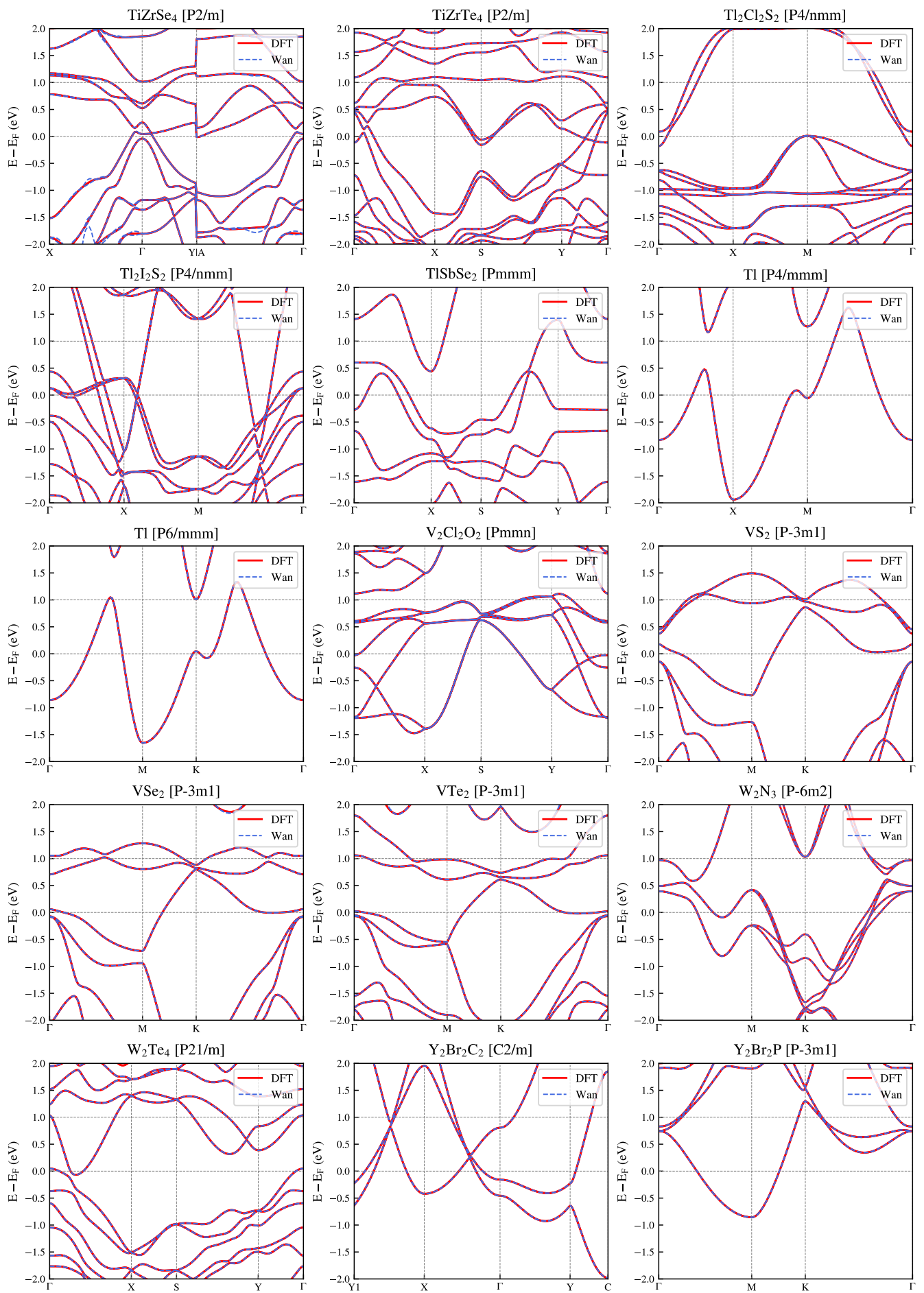


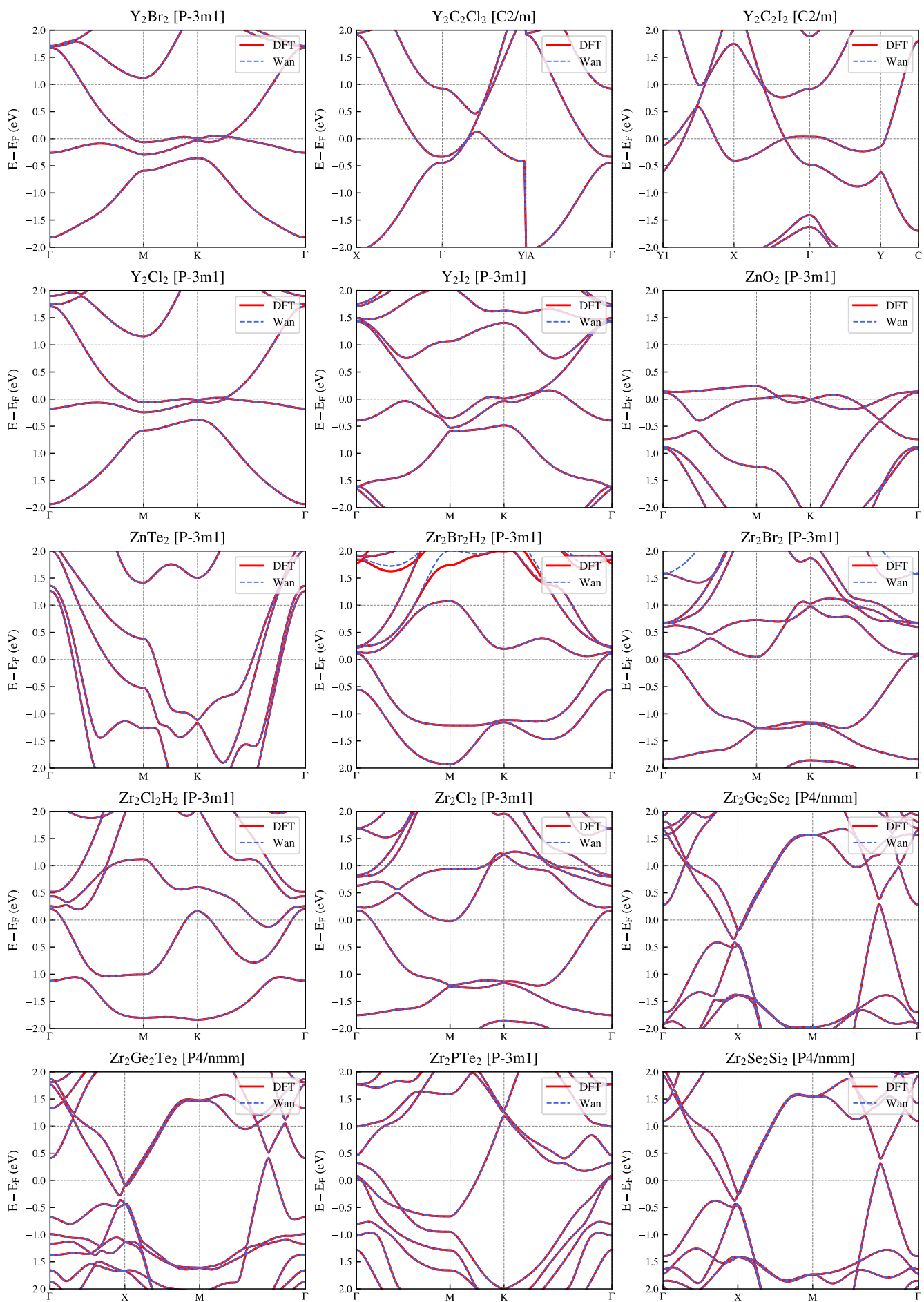


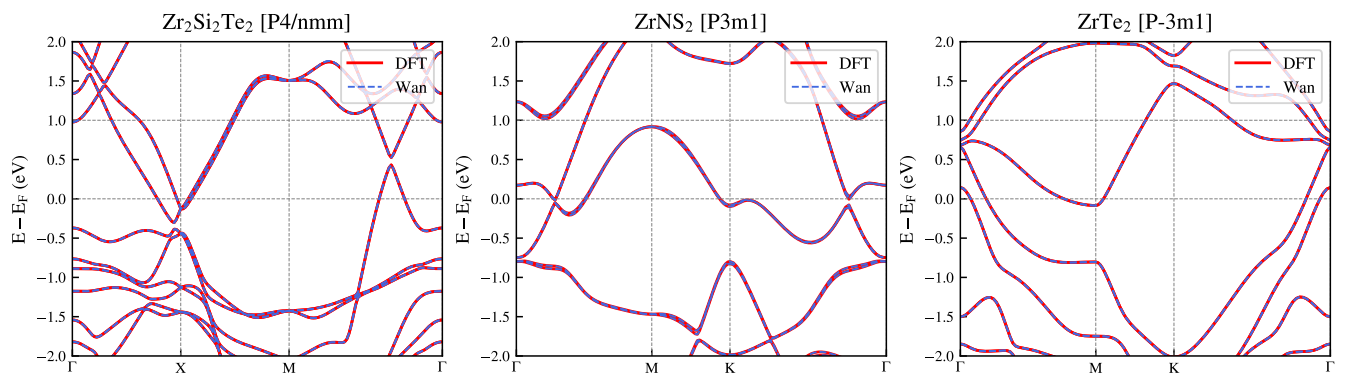




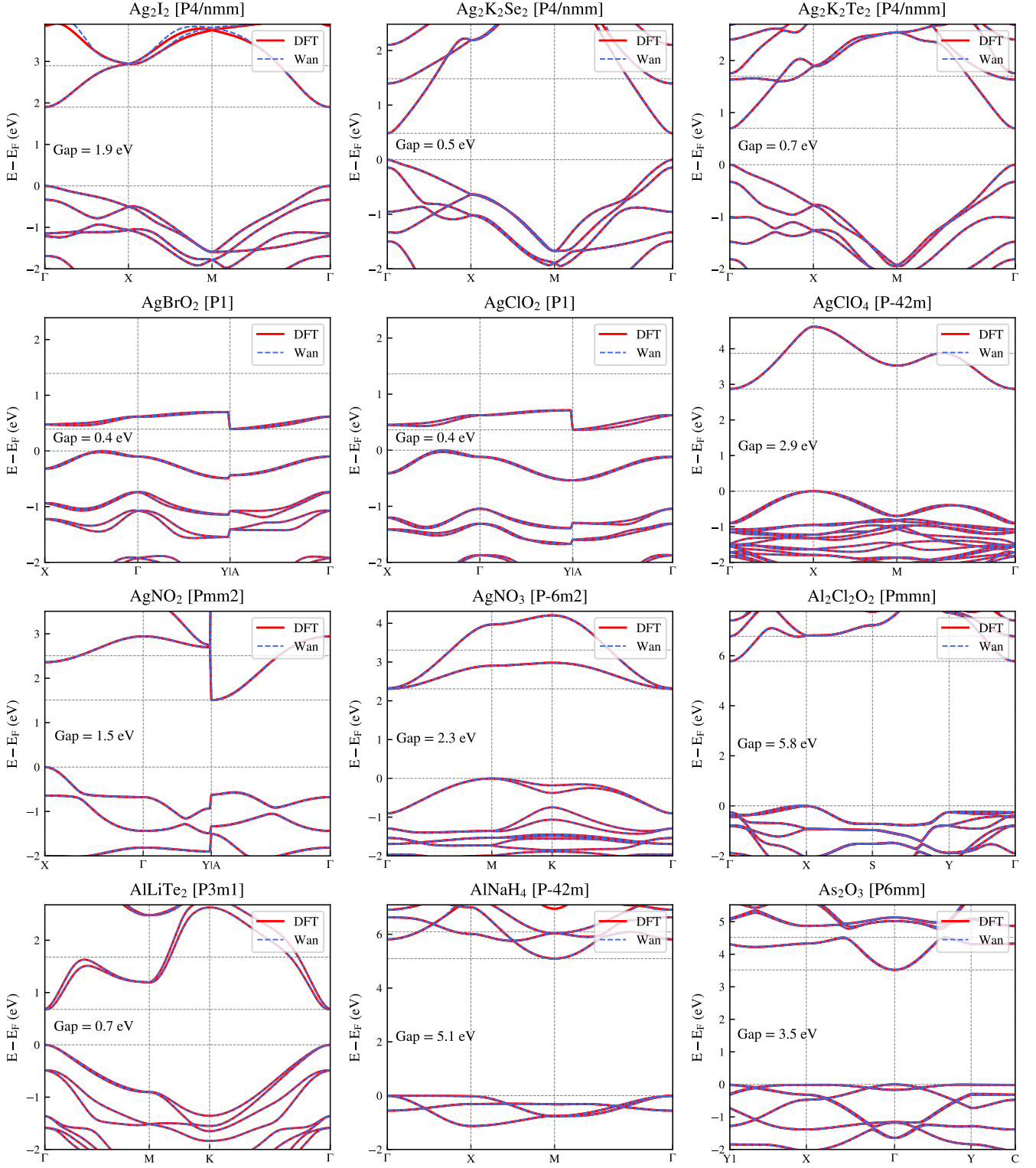


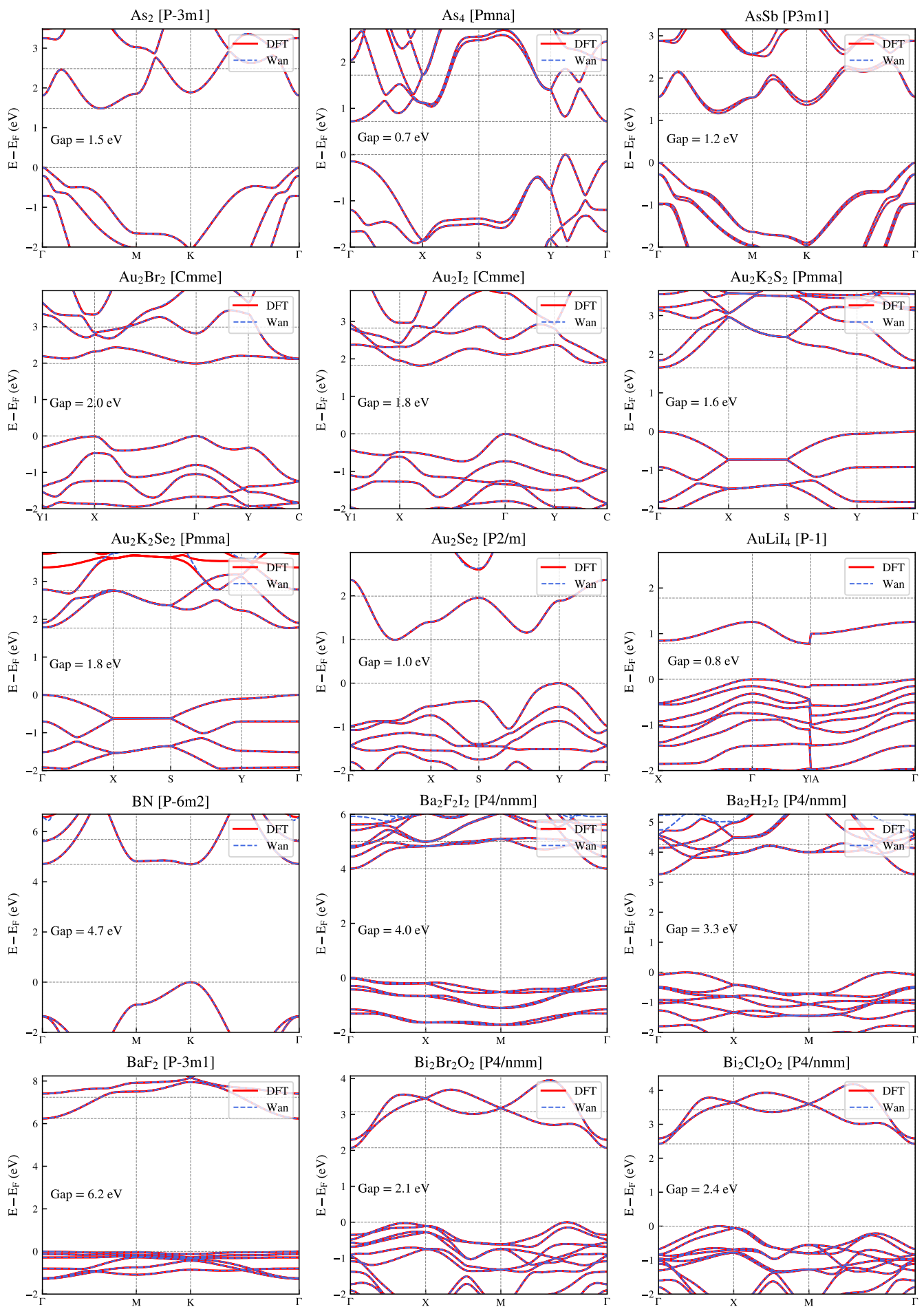


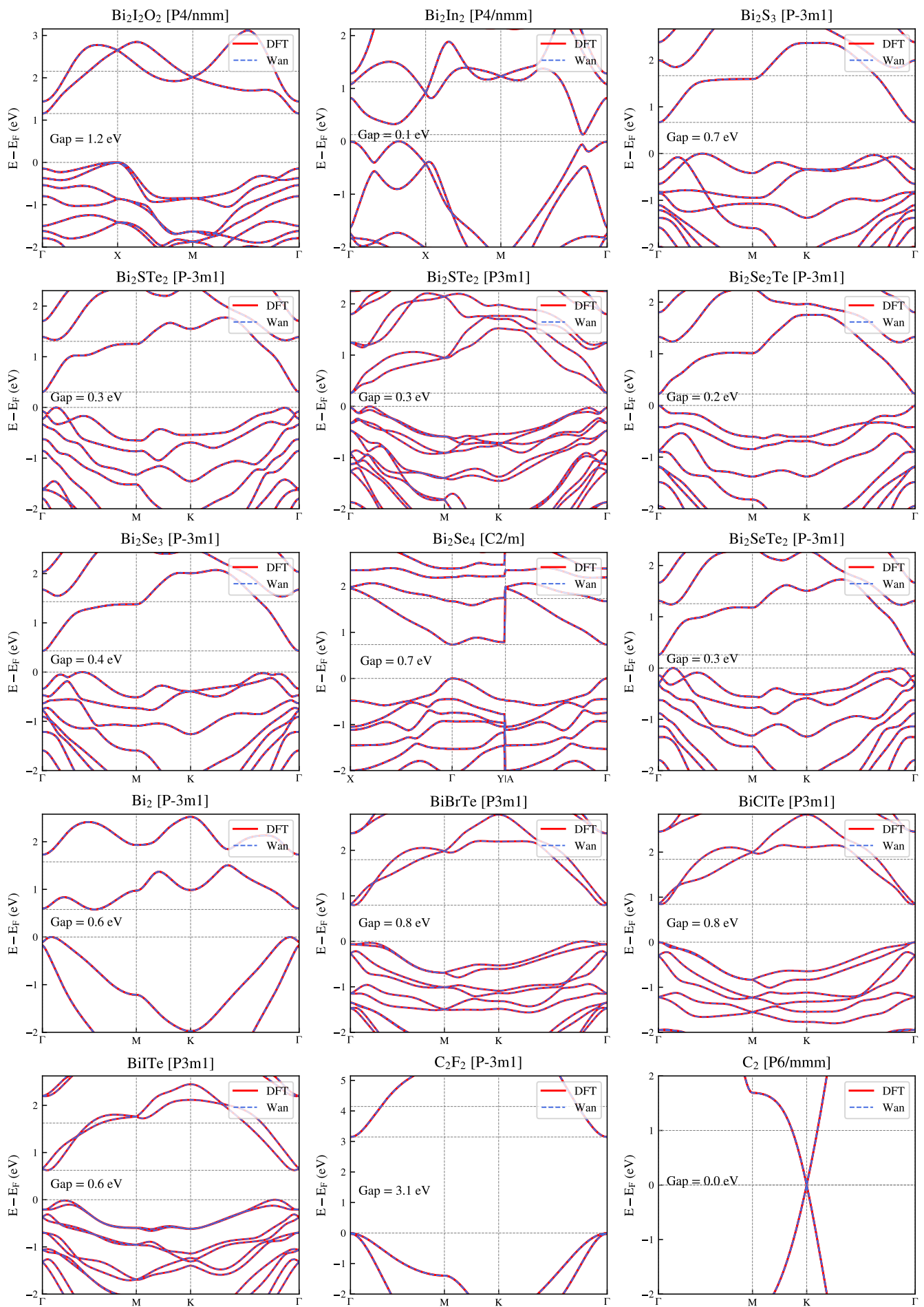


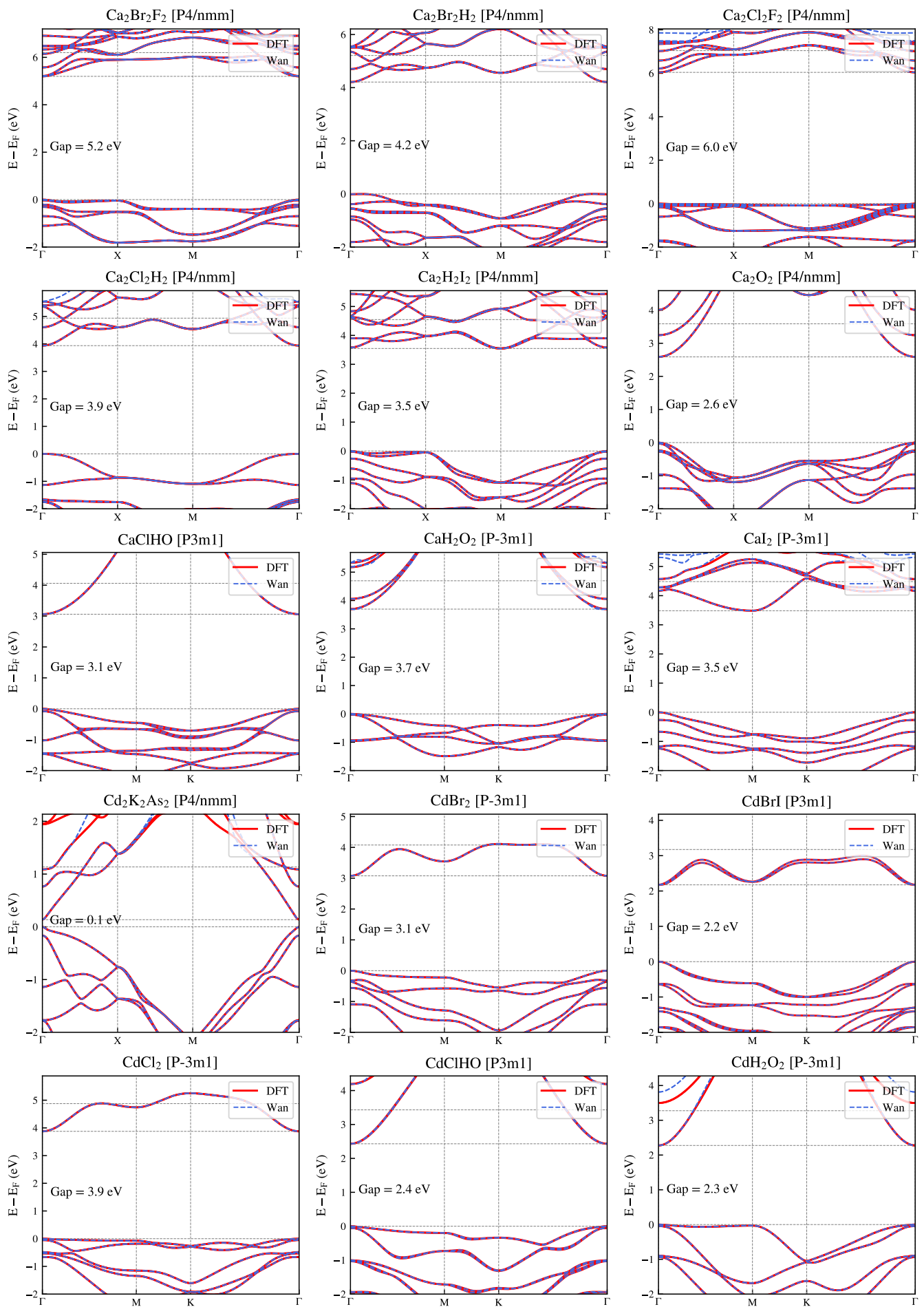


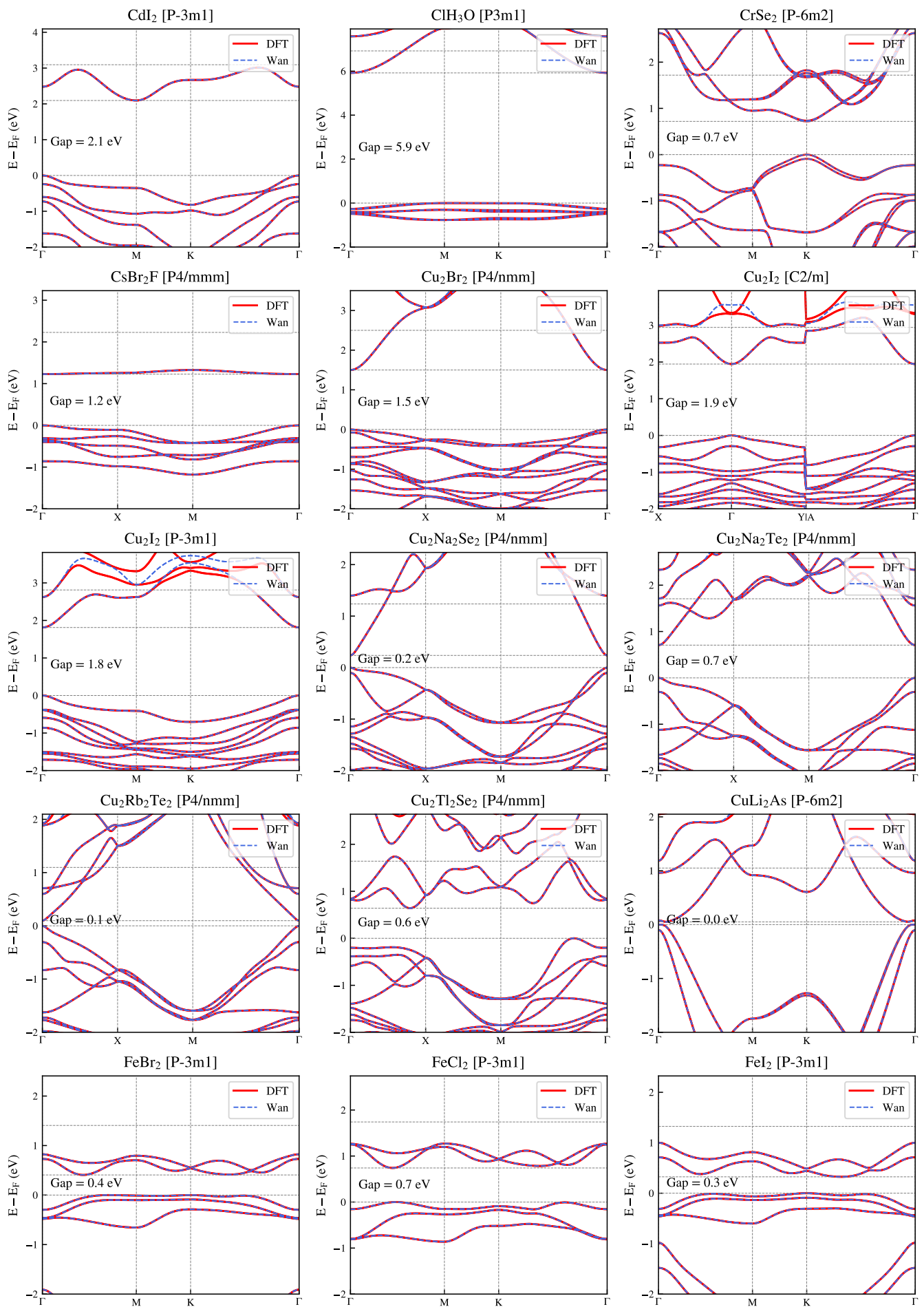
Supplementary Sec. 5. Fully relativistic electronic structures of 216 monolayer insulators

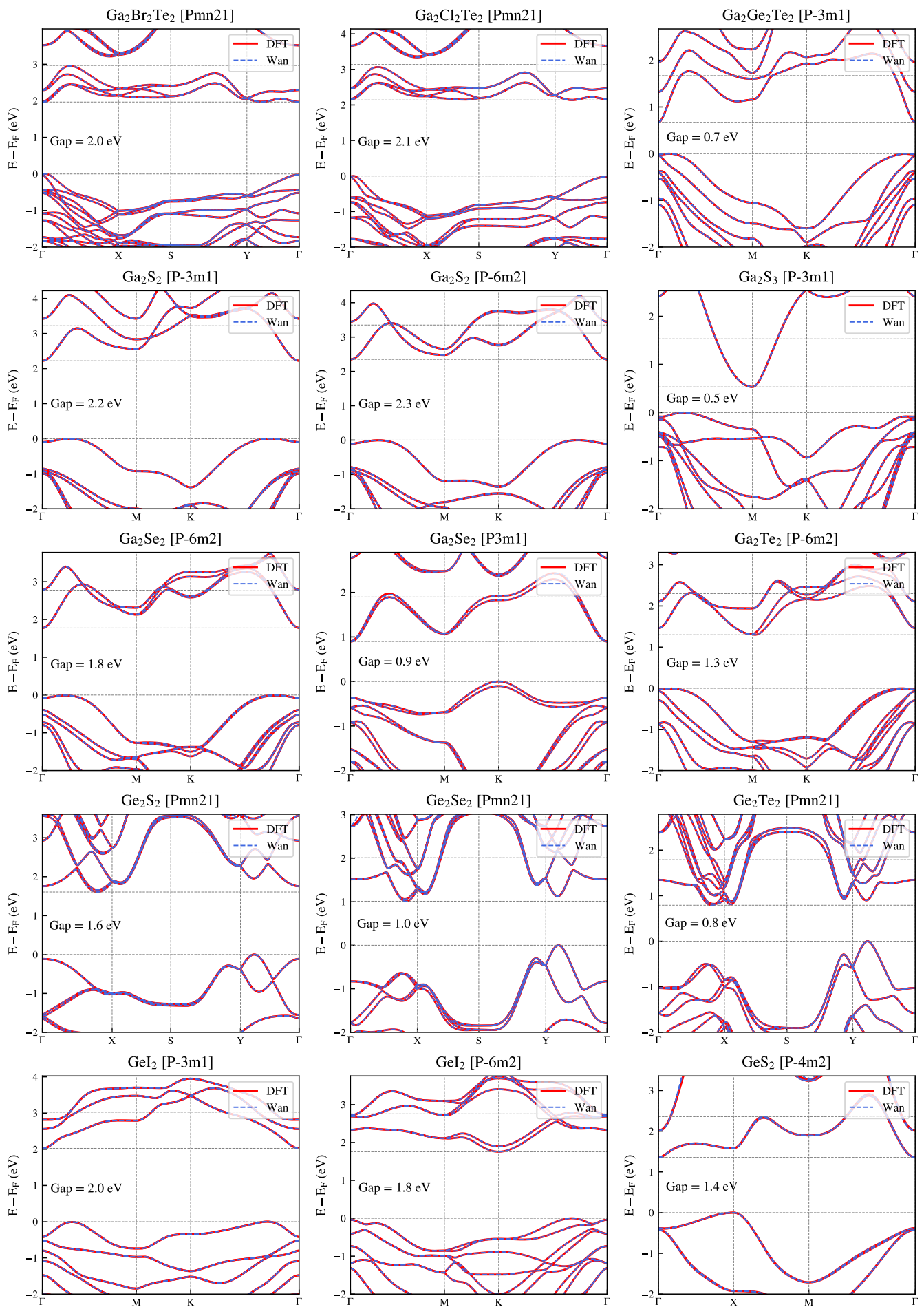


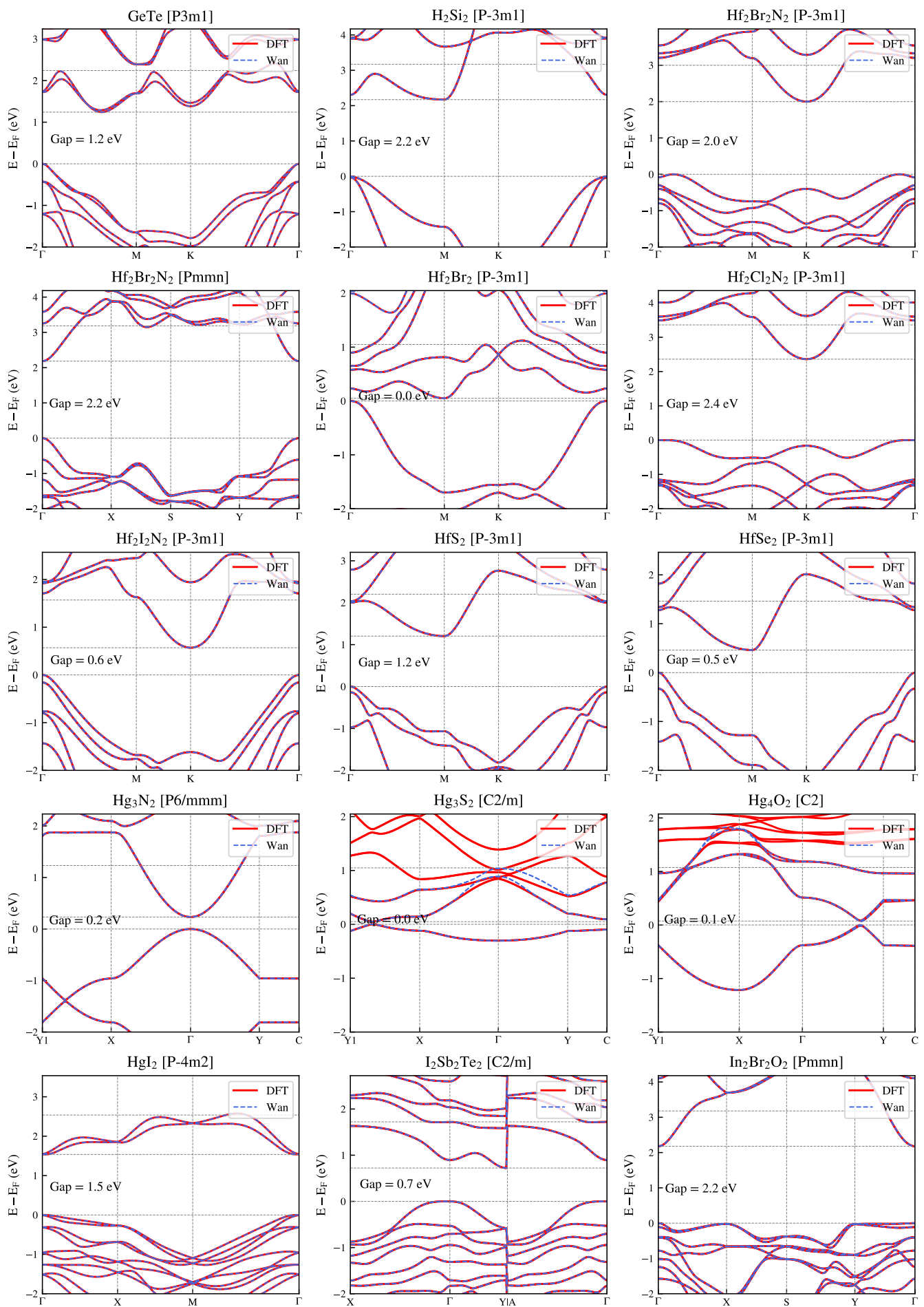


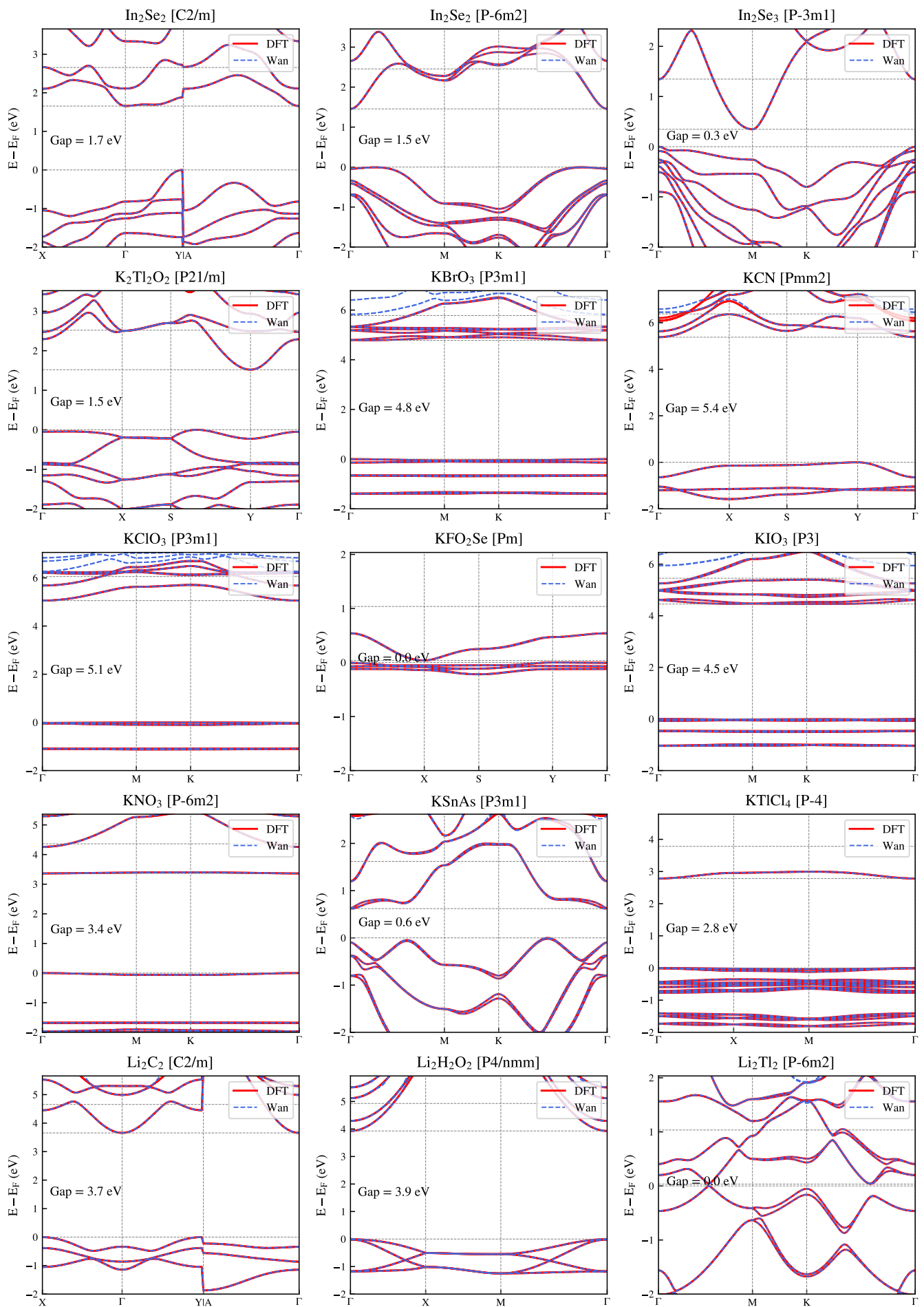


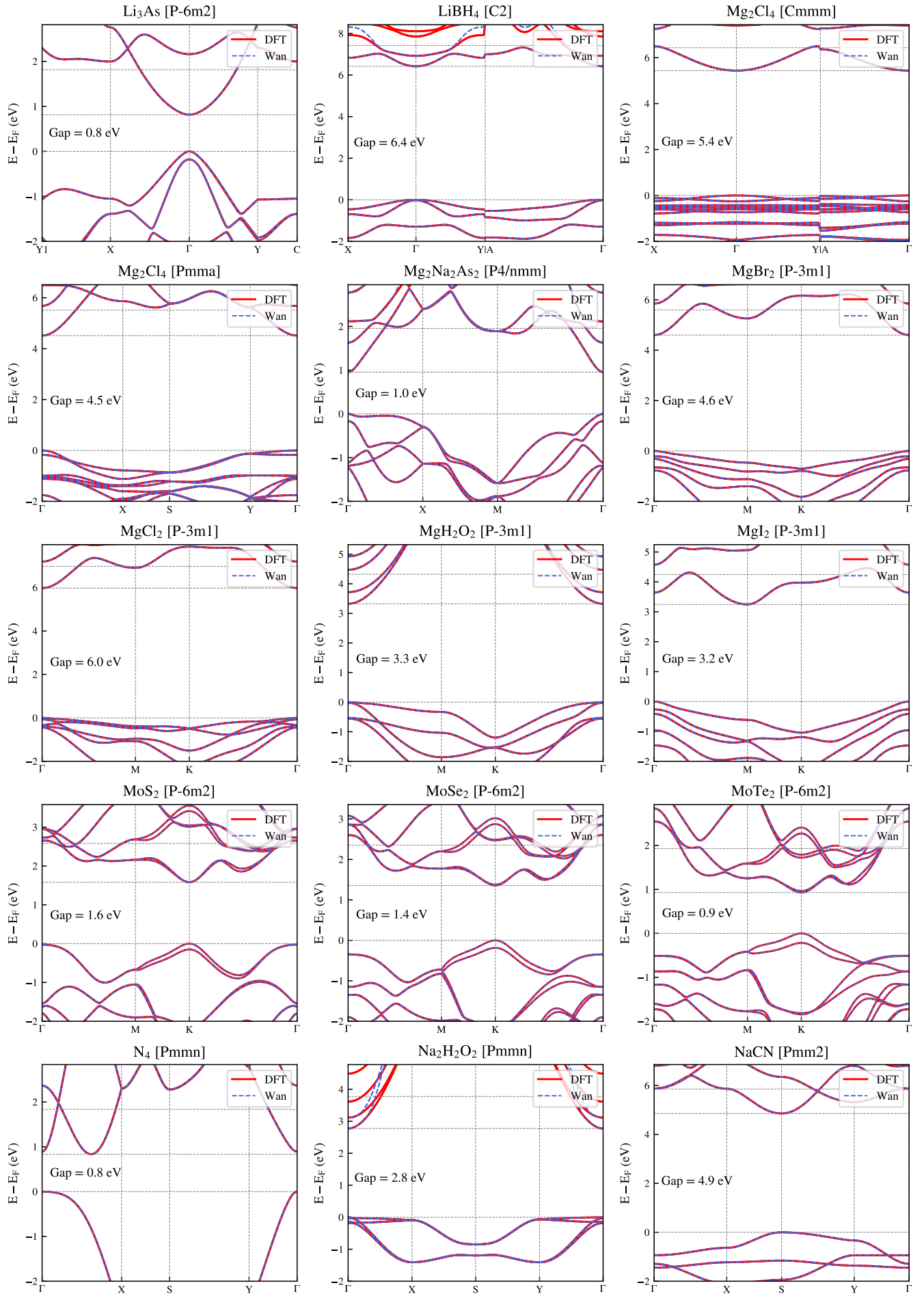


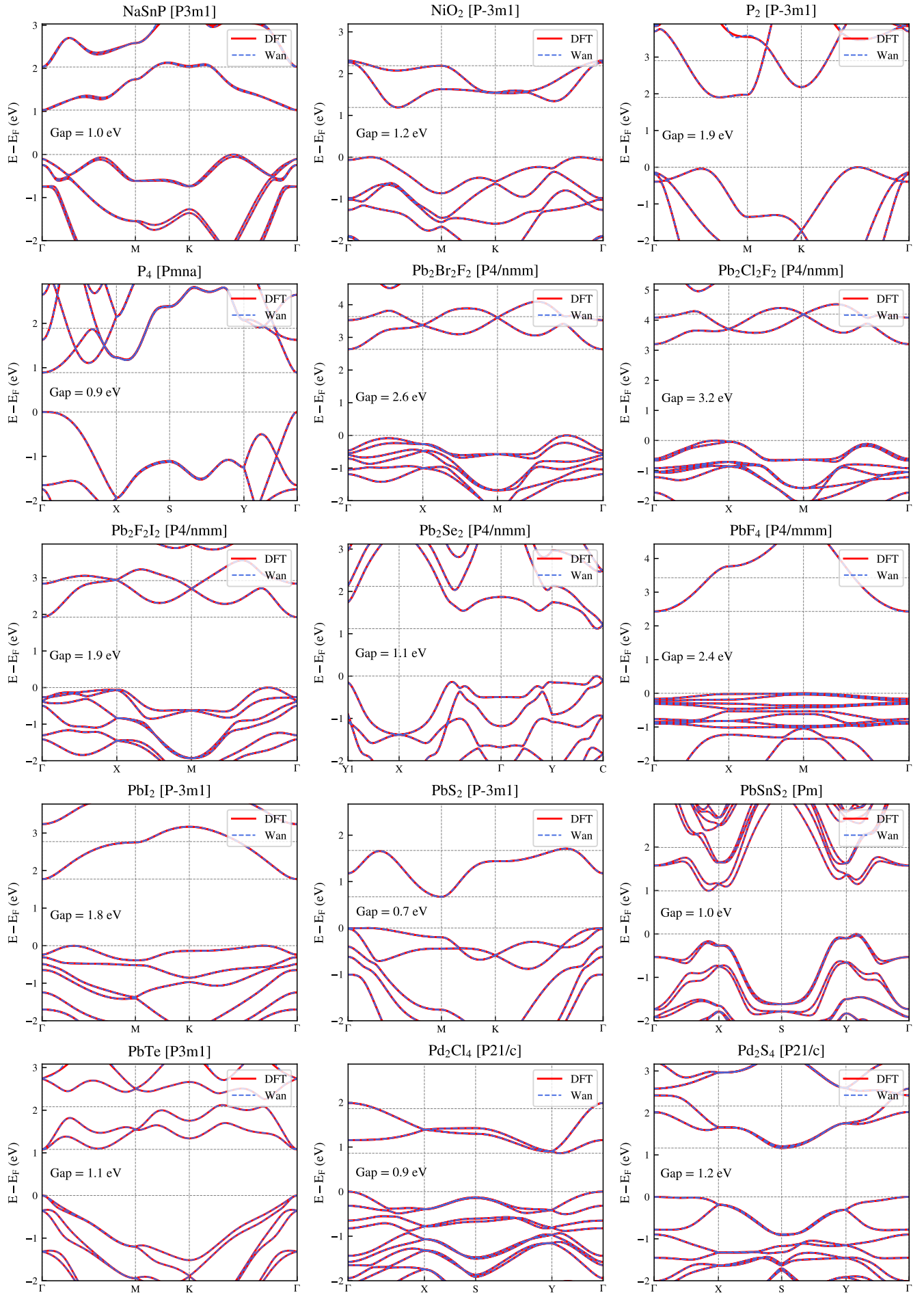


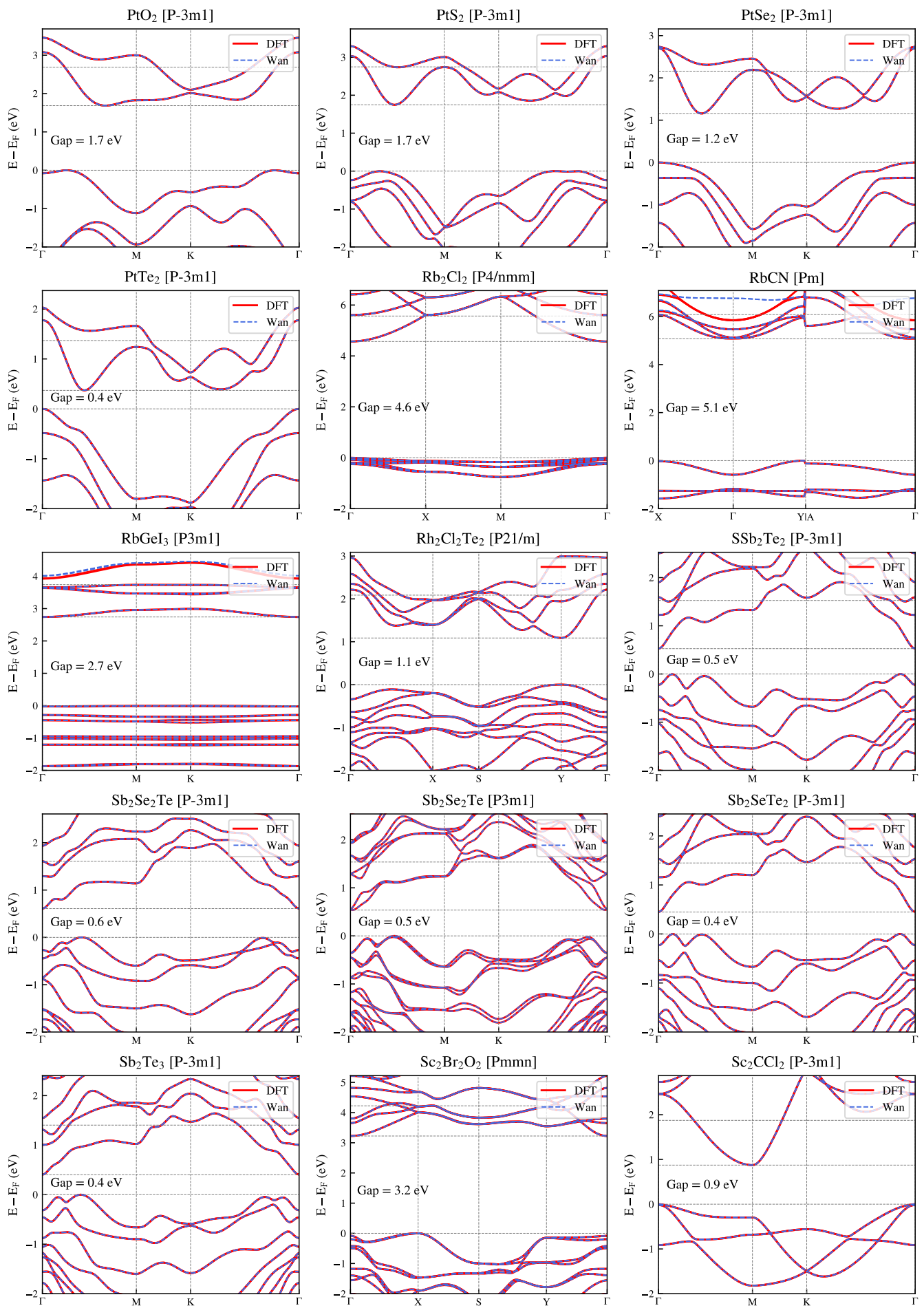


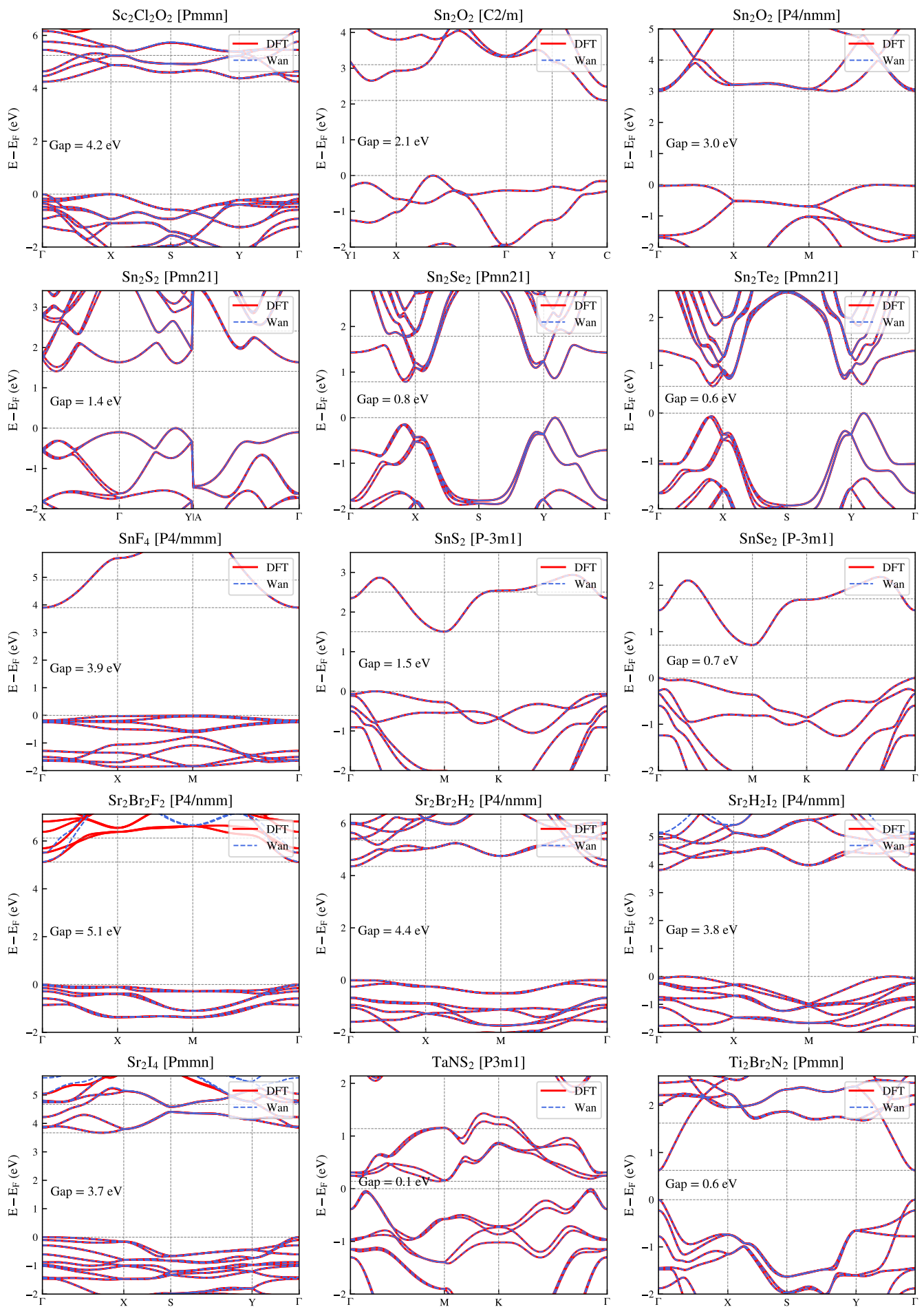


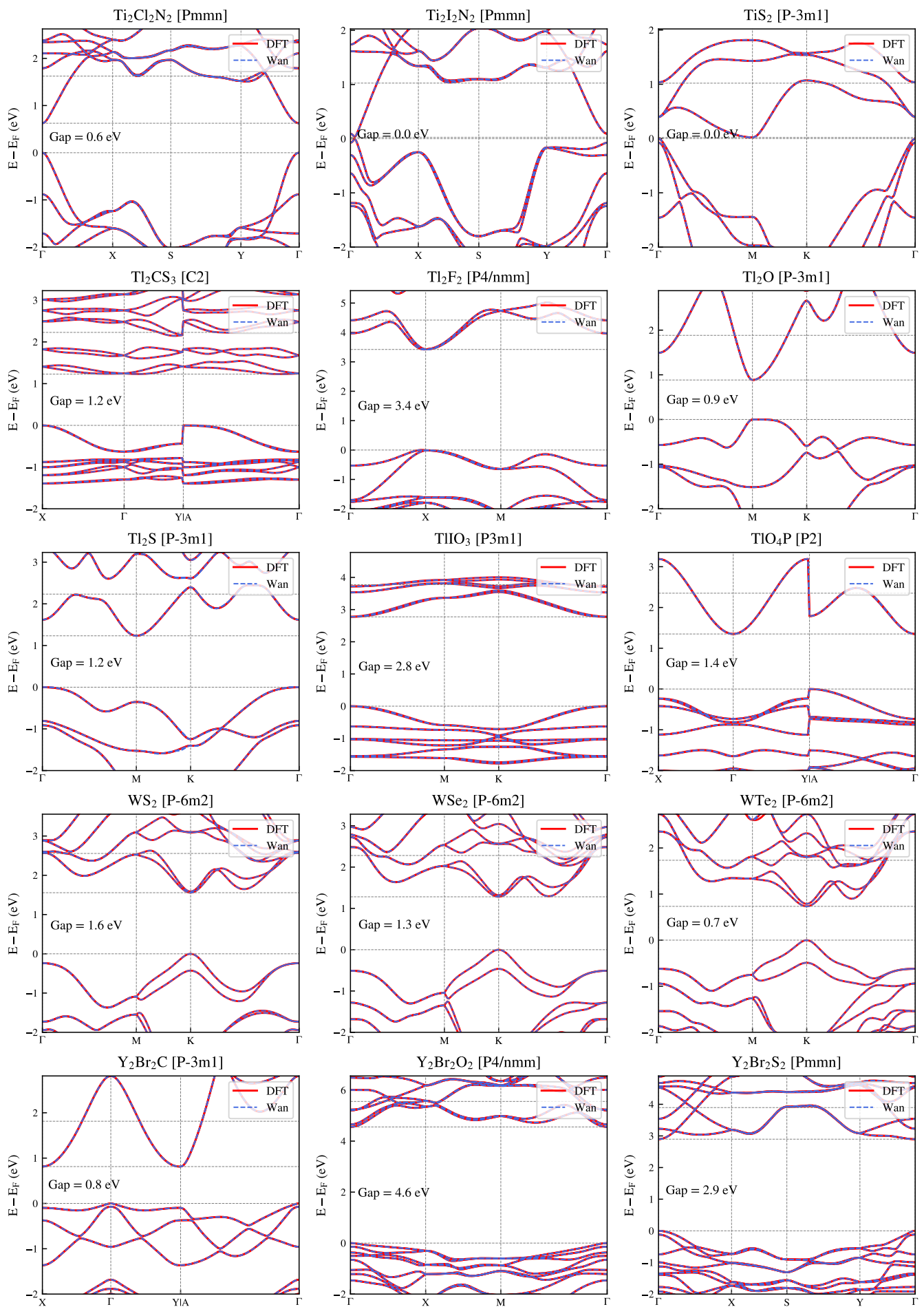


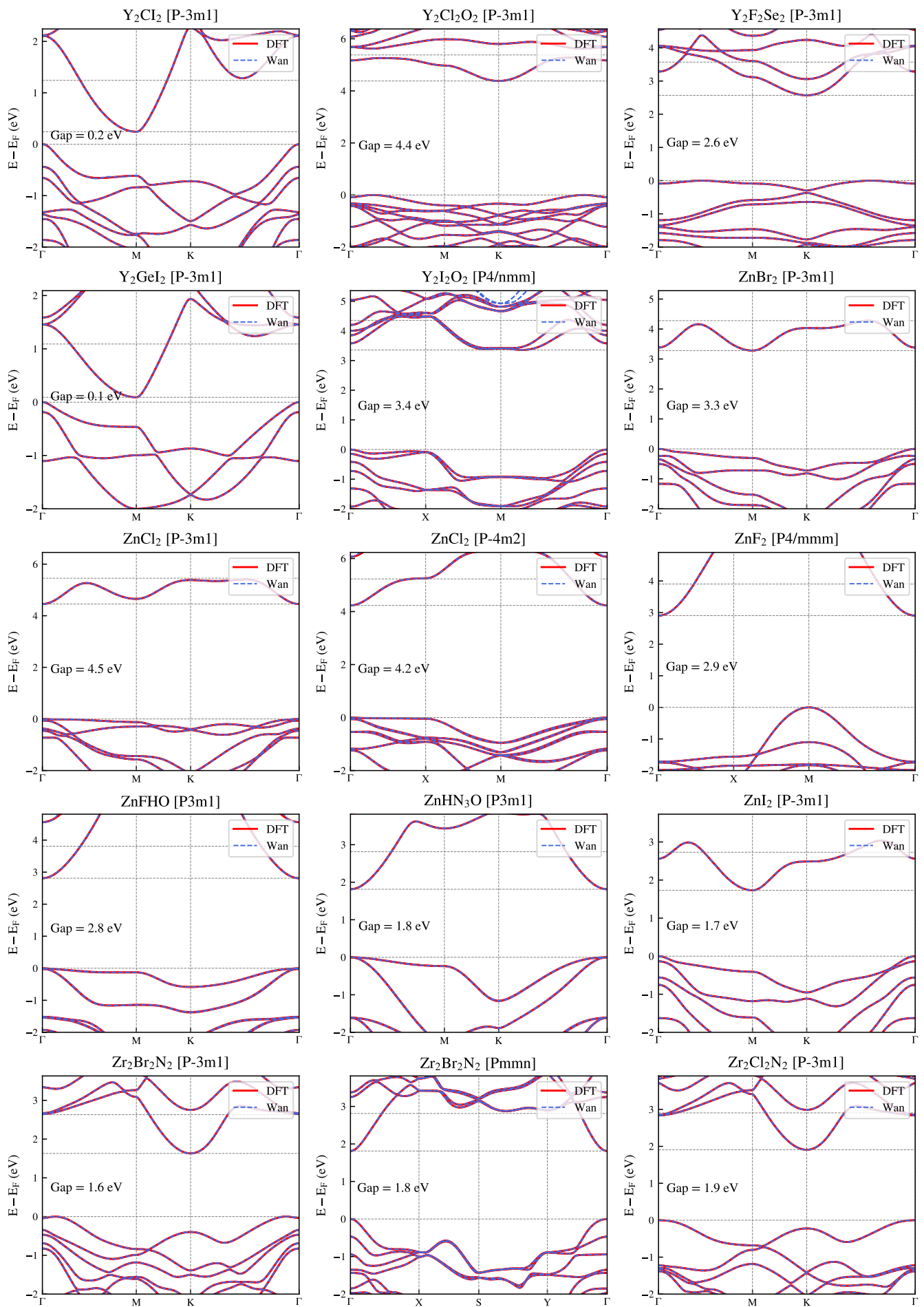


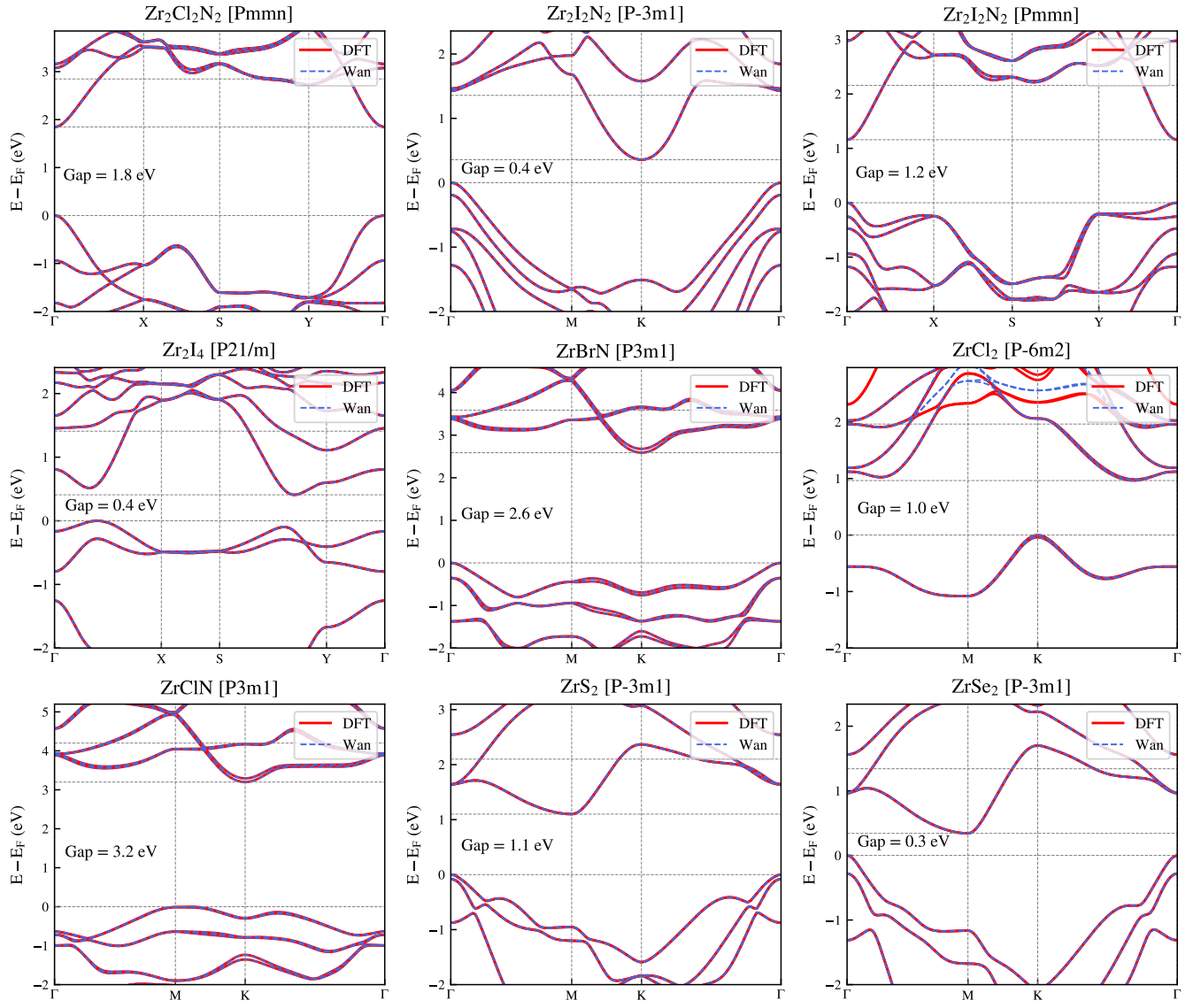












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