Electronic Supplementary Material (ESI) for Dalton Transactions.

Supplementary information

Tailoring spin waves in 2D transition metal phosphorus trichalcogenides via atomic-layer substitution

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Table 1. Relative strength of $NiPS_3$ isotropic exchange and DMI

	U = 4	**U = 5**
$\lvert D_1/J_1 \rvert$	0.0028	0.0014
$\lvert D_1'/J_1' \rvert$	0.0013	0.0006
$\lvert D_2/J_2 \rvert$	0.1276	0.0763
$\lvert D_2'/J_2' \rvert$	0.0593	0.0682
$\lvert D_3/J_3 \rvert$	0.0002	0.0001
$\lvert D_3'/J_3' \rvert$	0.0002	0.0002

Table 2. Relative strength of $NiPSe_3$ isotropic exchange and DMI

	U = 4	**U = 5**
$\lvert D_1/J_1 \rvert$	0.2156	0.1421
$\lvert D_1'/J_1' \rvert$	0.8277	0.6292
$\lvert D_2/J_2 \rvert$	0.6311	7.6345
$\lvert D_2'/J_2' \rvert$	0.3431	0.7972
$\lvert D_3/J_3 \rvert$	0.0522	0.0528
$\lvert D_3'/J_3' \rvert$	0.0231	0.0265

Table 3. Relative strength of $MnPS_3$ isotropic exchange and DMI

	U = 2	**U = 4**
$\lvert D_1/J_1 \rvert$	0.0005	0.0008
$\lvert D_1'/J_1' \rvert$	0.0008	0.0036
$\lvert D_2/J_2 \rvert$	0.0486	0.1444
$\lvert D_2'/J_2' \rvert$	0.0474	0.136
$\lvert D_3/J_3 \rvert$	0.0006	0.0023
$\lvert D_3'/J_3' \rvert$	0.001	0.0009

Table 4. Relative strength of $MnPSe_3$ isotropic exchange and DMI

	U = 2	**U = 4**
$\lvert D_1/J_1 \rvert$	0.0258	0.0267
$\lvert D_1'/J_1' \rvert$	0.0244	0.0268
$\lvert D_2/J_2 \rvert$	0.13	0.2199
$\lvert D_2'/J_2' \rvert$	0.1273	0.2196
$\lvert D_3/J_3 \rvert$	0.0341	0.0291
$\lvert D_3'/J_3' \rvert$	0.0365	0.036

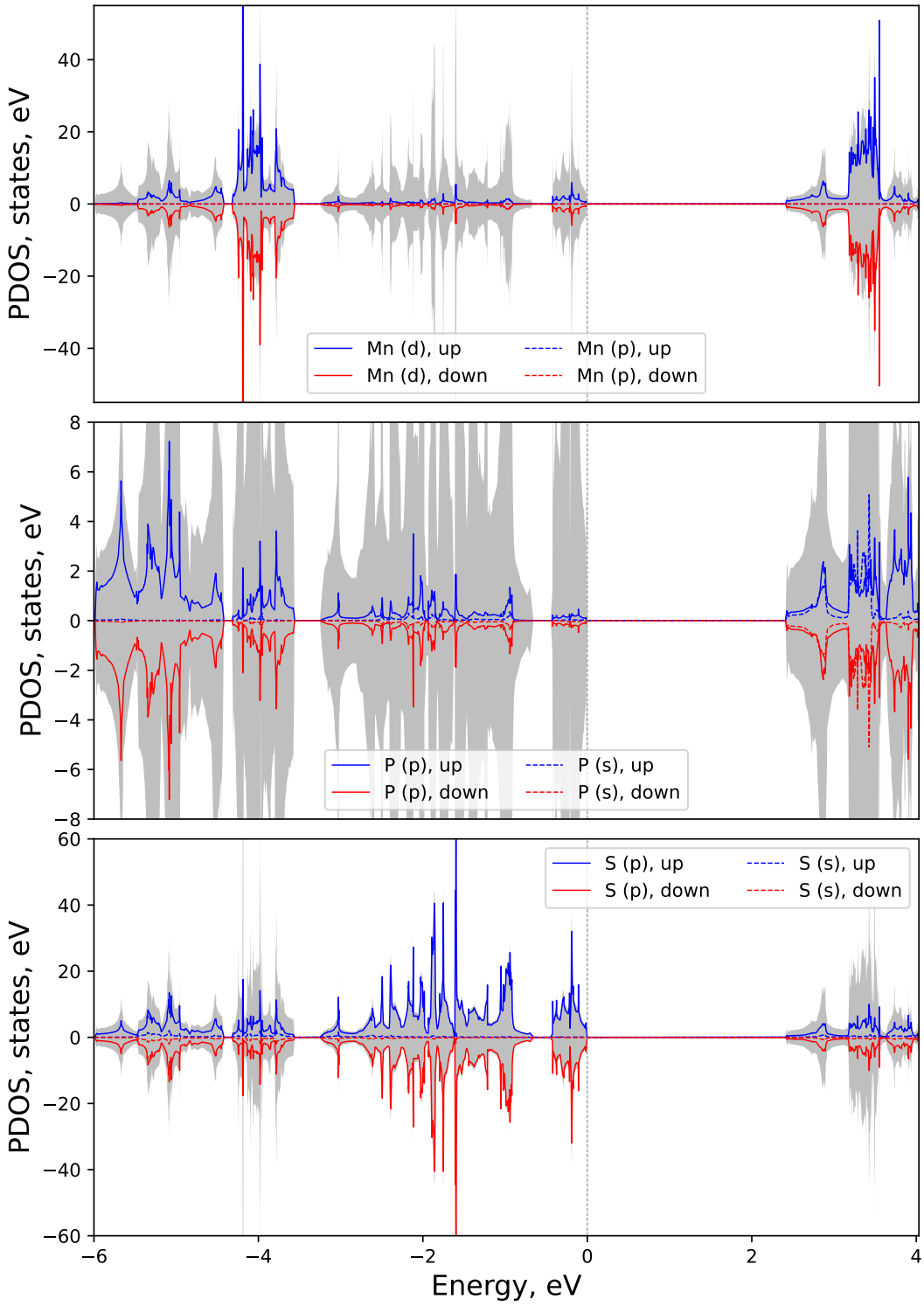


Figure S1 Projected density of states (orbital resolved) for $MnPS_3$. From top to down: Mn, P and S. Spin up (blue); Spin down (red); Grey color corresponds to the total density of states. The valence band edge has been placed at E_F = 0 eV.

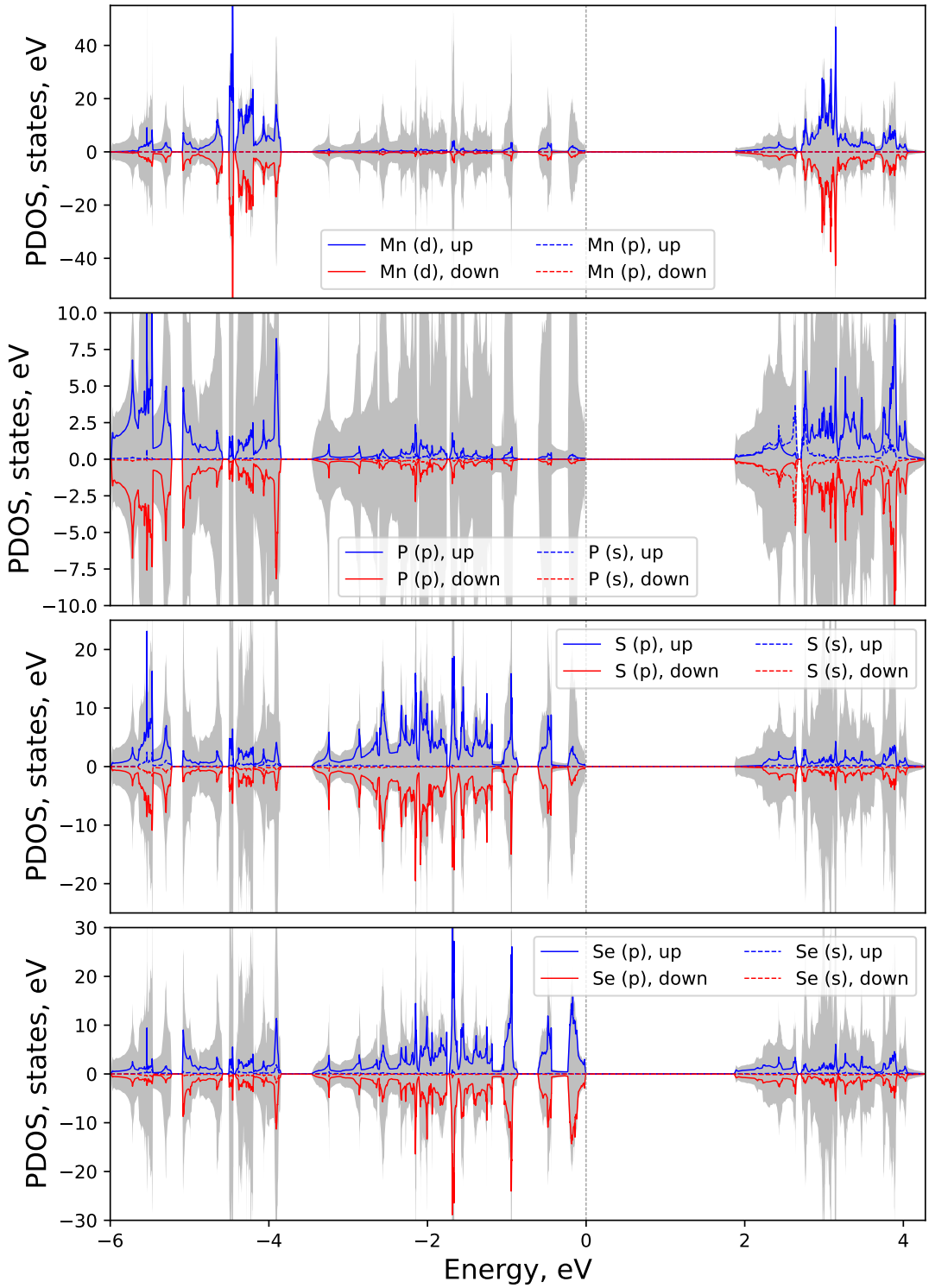


Figure S2 Projected density of states (orbital resolved) for $MnPS_{1.5}Se_{1.5.}$ From top to down: Mn, P, S and Se. Spin up (blue); Spin down (red); Grey color corresponds to the total density of states. The valence band edge has been placed at E_F = 0 eV.

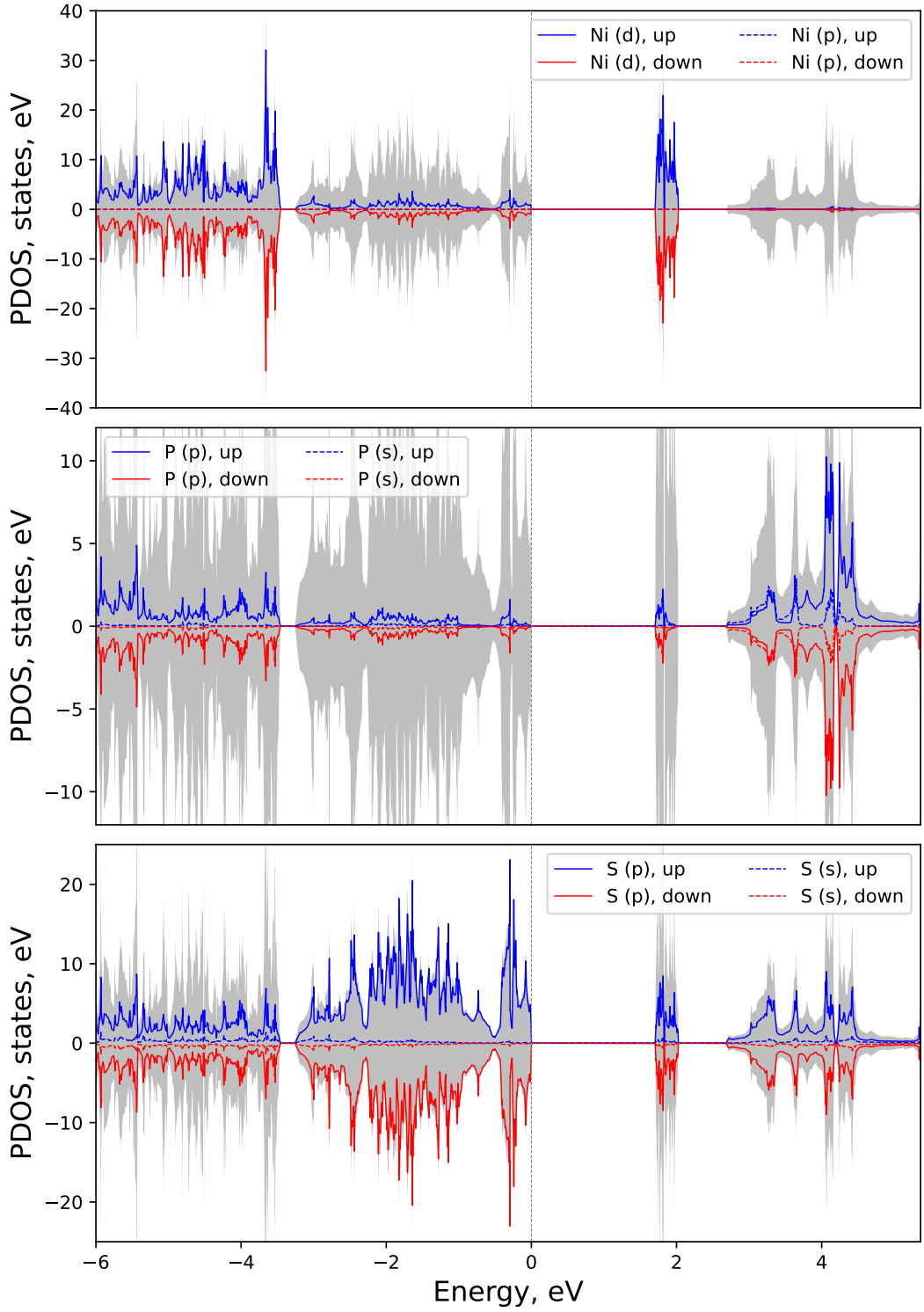


Figure S3 Projected density of states (orbital resolved) for $NiPS_3$. From top to down: Ni, P and S. Spin up (blue); Spin down (red); Grey color corresponds to the total density of states. The valence band edge has been placed at E_F = 0 eV.

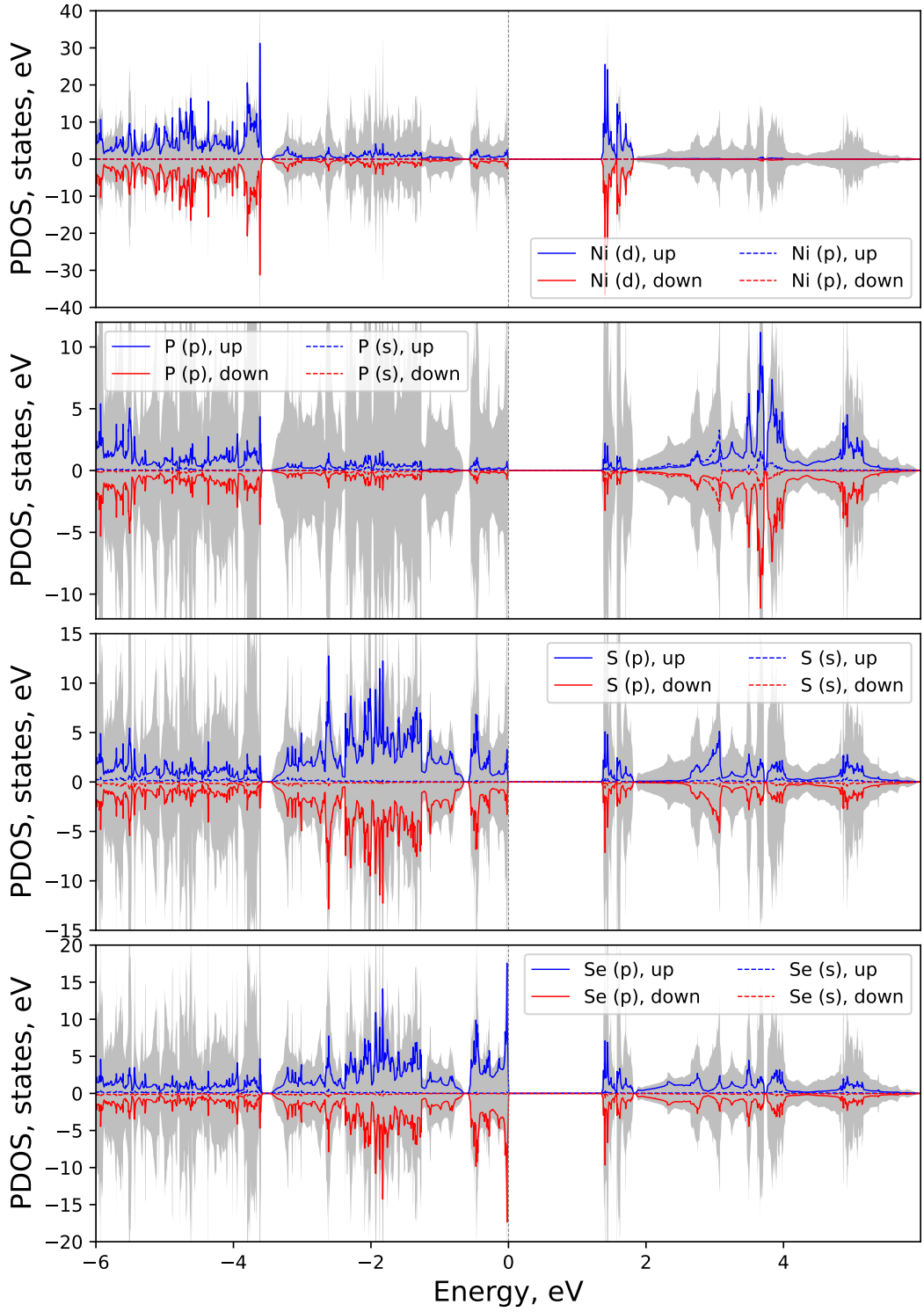
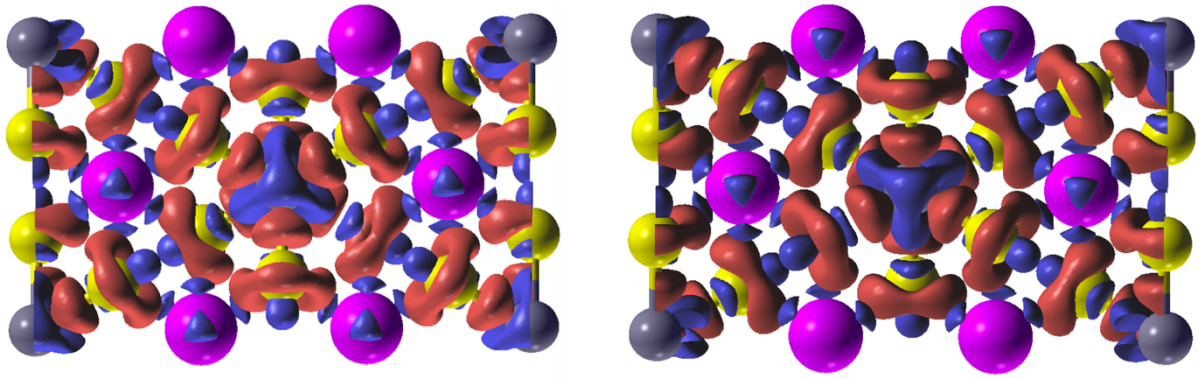


Figure S4 Projected density of states (orbital resolved) for $NiPS_{1.5}Se_{1.5}$. From top to down: Ni, P, S and Se. Spin up (blue); Spin down (red); Grey color corresponds to the total density of states. The valence band edge has been placed at E_F = 0 eV.

(a) (b)



(c) (d)

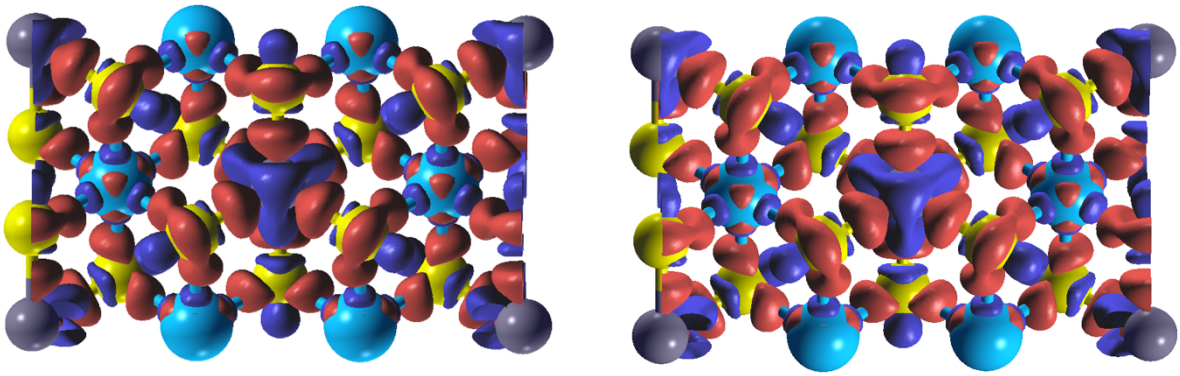


Figure S5 Valence electronic density differential plot with an isosurface value of 0.007 for the (a) top and (b) bottom view of $MnPS_3$; (c) top and (d) bottom view of $NiPS_3$. Color code: Mn (pink), Ni (clear blue), P (gray) and S (yellow). Blue (red) depicts electron accumulation (depletion) regions.