Supplemental Materials: Lattice dynamics and magnetic exchange interactions in $GeCo_2O_4$, a spinel with S = 1/2 pyrochlore lattice

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I. Simulated infrared (IR) spectrum

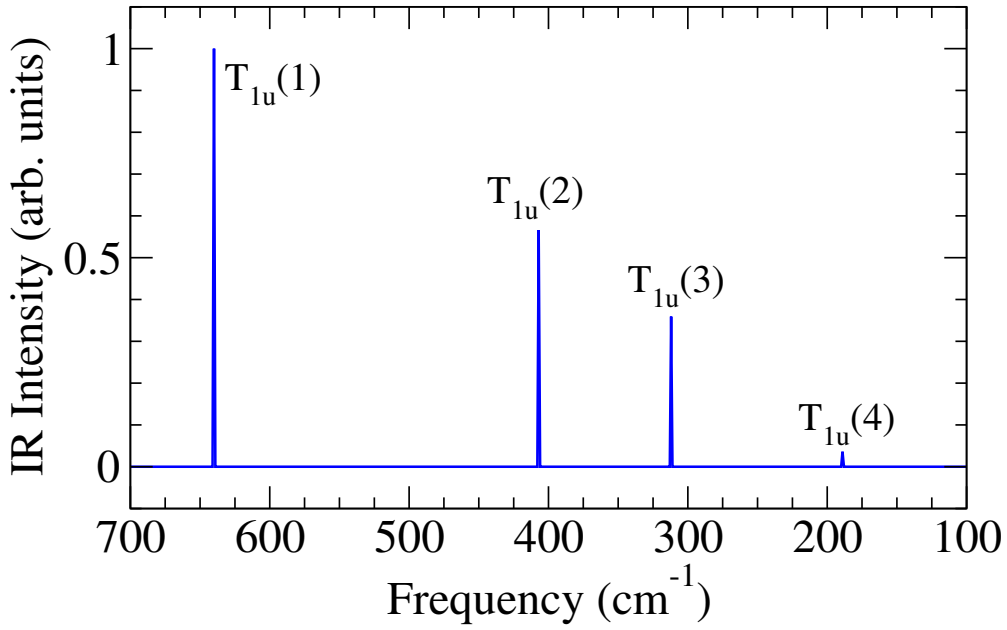

FIG. S1. PBEsol+U simulated IR spectrum. The IR intensities were calculated by projecting the Born-effective charge tensors, calculated within the primitive cell, into the space of IR-active phonon eigen displacements obtained from the statistically averaged force constants matrix for the high-temperature paramagnetic phase (see the main text).

II. Spin configurations employed for the spin Hamiltonian fitting

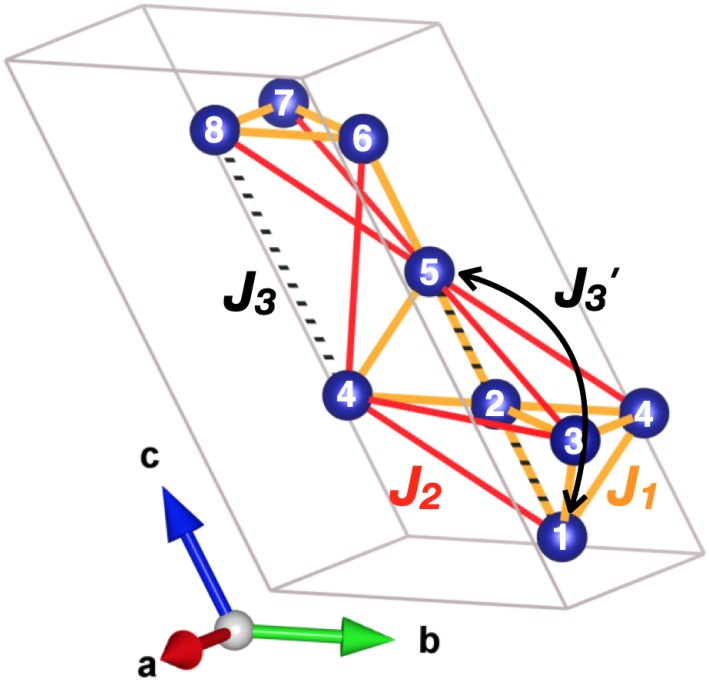

FIG. S2. Definition of all four magnetic-exchange interactions, first (J_1), second (J_2), and third (J_3 and $J_3^{'}$) nearest neighbors, considered in this work. Eight Co atoms are shown in blue color. Ge and O atoms are omitted for clarity. Note that $J_3^{'}$ passes through an intermediate Co atom (see the main text for details).

TABLE S1. DFT+U (PBEsol) calculated total energy for different spin configurations on the eight Co sites in $1 \times 1 \times 2$ supercell of $GeCo_2O_4$, as defined in Figure S2. The magnetic-exchange coupling parameters were estimated by fitting Eq. 1 in the main text using the data presented in this table.

Spin configuration index, n	$S_1\, S_2\, S_3\, S_4\, S_5\, S_6\, S_7\, S_8$	DFT+U (PBEsol) calculated total energy (eV)	Energy difference, $\Delta E = E_n - E_1$ (eV)
1	$+\,+\,+\,+\,-\,-\,-\,-$	-188.99773810	0.000000
2	$+\,+\,+\,+\,+\,-\,-\,-$	-188.98495812	0.012780
3	$-\,+\,+\,+\,-\,-\,-\,-$	-188.98494070	0.012797
4	$+\,+\,+\,+\,+\,+\,+\,+$	-188.96541050	0.032328
5	$-\,-\,+\,+\,-\,-\,-\,-$	-188.94245619	0.055282
6	$+\,+\,+\,+\,+\,+\,-\,-$	-188.94245101	0.055287
7	$+\,+\,+\,+\,+\,-\,-\,+$	-188.93296451	0.064774
8	$+\,+\,+\,+\,+\,+\,+\,-$	-188.92681620	0.070922
9	$-\,-\,-\,+\,-\,-\,-\,-$	-188.92680050	0.070938
10	$+\,+\,+\,+\,+\,+\,-\,+$	-188.92460530	0.073133
11	$+\,+\,+\,+\,-\,-\,-\,+$	-188.91849611	0.079242
12	$+\,+\,+\,-\,-\,-\,-\,+$	-188.89865016	0.099088
13	$+\,-\,-\,-\,-\,-\,-\,+$	-188.89411040	0.103628
14	$+\,+\,-\,-\,-\,-\,-\,+$	-188.88232052	0.115418
15	$+\,-\,-\,-\,+\,-\,-\,-$	-188.82054962	0.177188
16	$+\,-\,+\,-\,+\,-\,+\,-$	-188.79163870	0.206099
17	$+\,+\,-\,-\,+\,+\,-\,-$	-188.78892830	0.208810

III. Density of states calculated for different spin configurations (PBEsol+U)

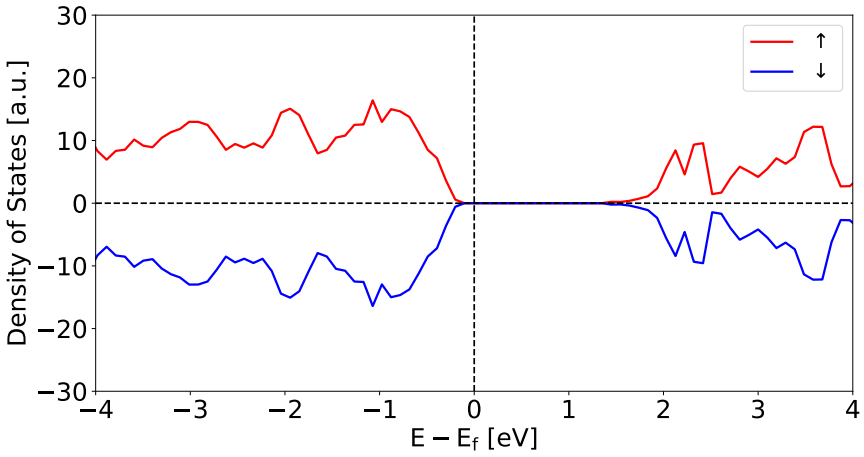

FIG. S3. Density of states calculated for the spin configuration $++++----$ listed in Table S1.

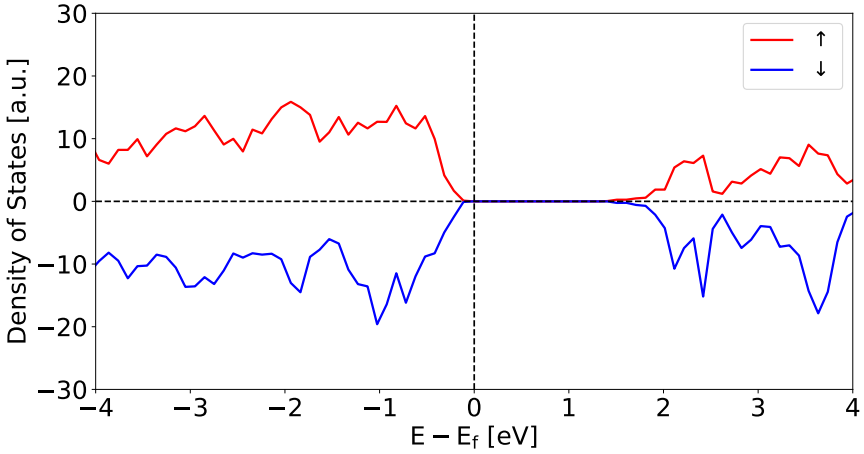

FIG. S4. Density of states calculated for the spin configuration $+++++---$ listed in Table S1.

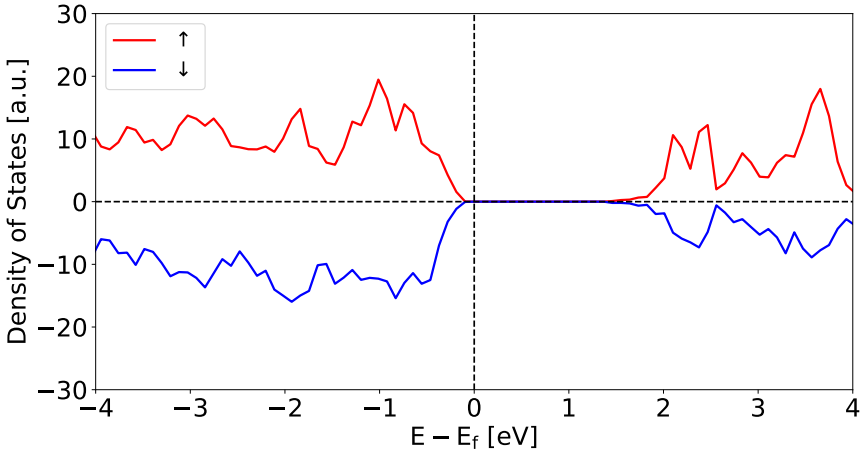

FIG. S5. Density of states calculated for the spin configuration $-++-----$ listed in Table S1.

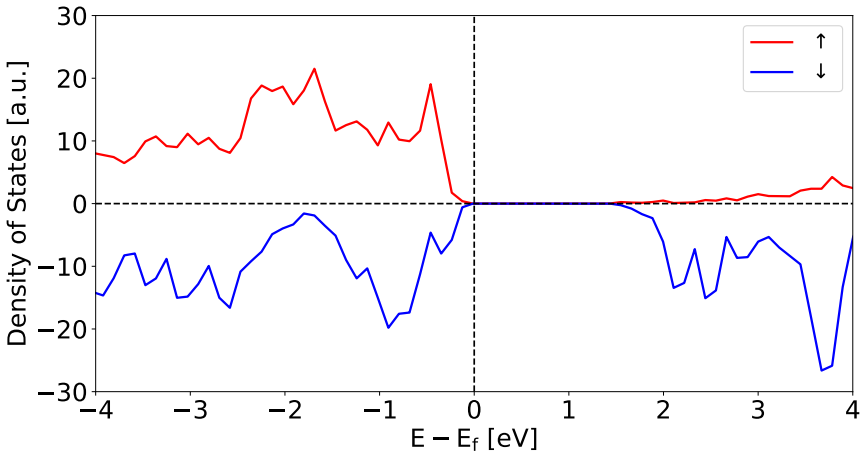

FIG. S6. Density of states calculated for the spin configuration $+++++++++$ listed in Table S1.

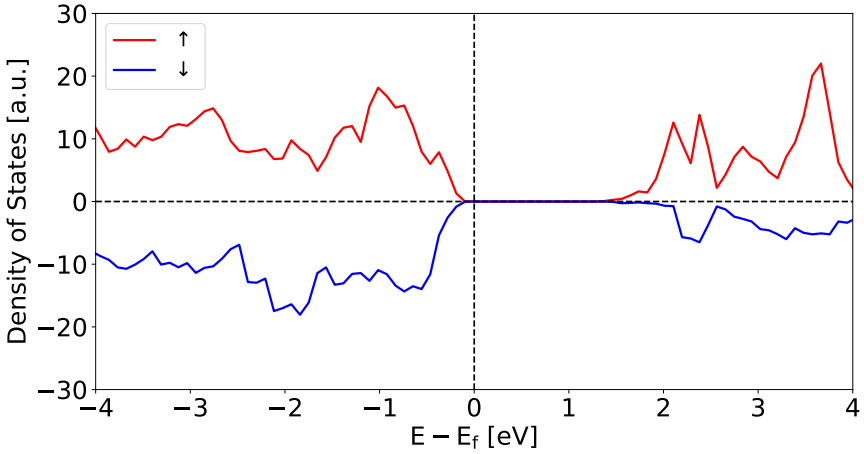

FIG. S7. Density of states calculated for the spin configuration $--++----$ listed in Table S1.

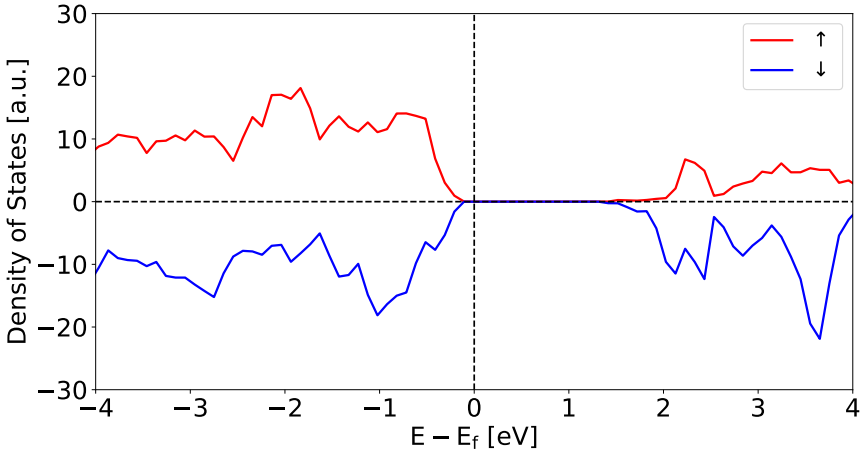

FIG. S8. Density of states calculated for the spin configuration $+++++++--$ listed in Table S1.

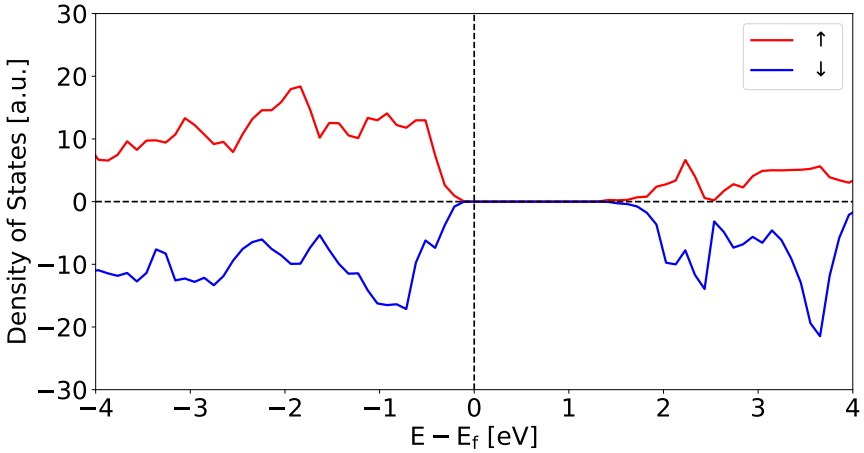

FIG. S9. Density of states calculated for the spin configuration $++++++--+$ listed in Table S1.

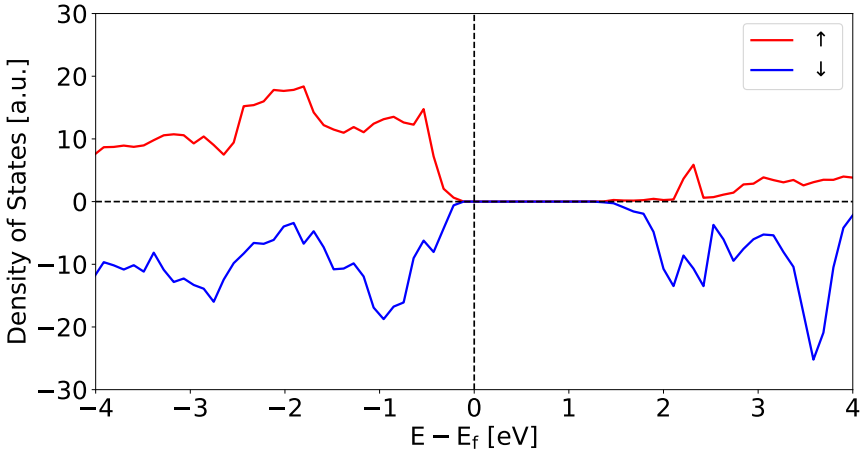

FIG. S10. Density of states calculated for the spin configuration $++++++++-$ listed in Table S1.

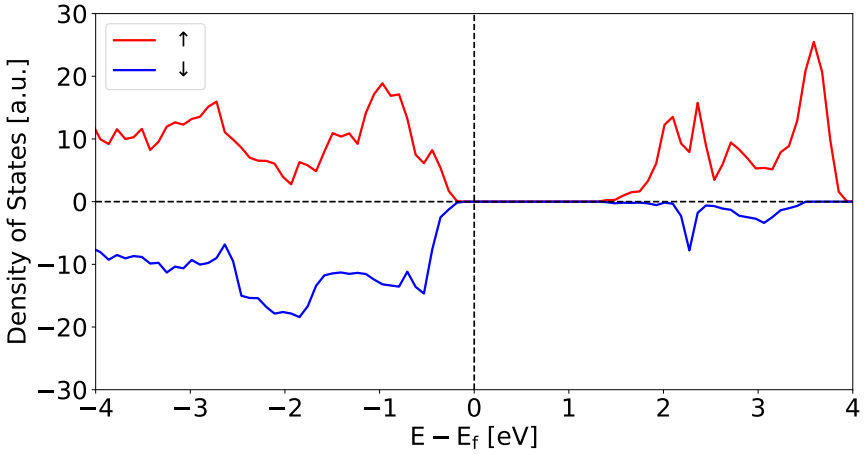

FIG. S11. Density of states calculated for the spin configuration $---+----$ listed in Table S1.

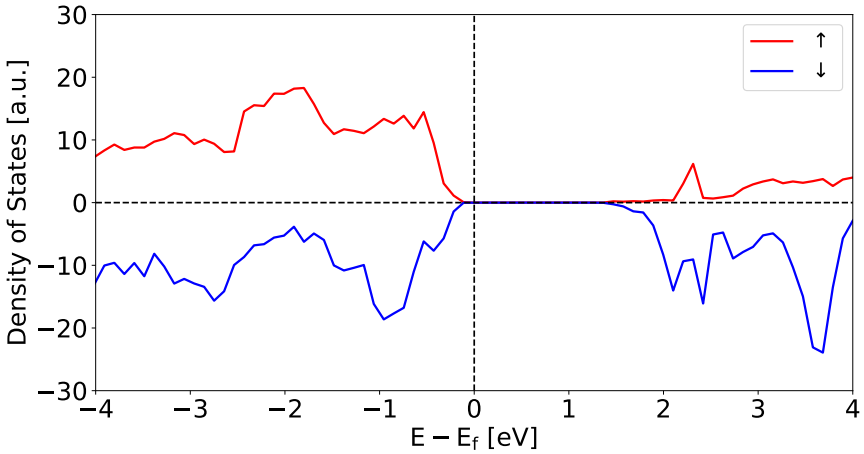

FIG. S12. Density of states calculated for the spin configuration $++++++-+$ listed in Table S1.

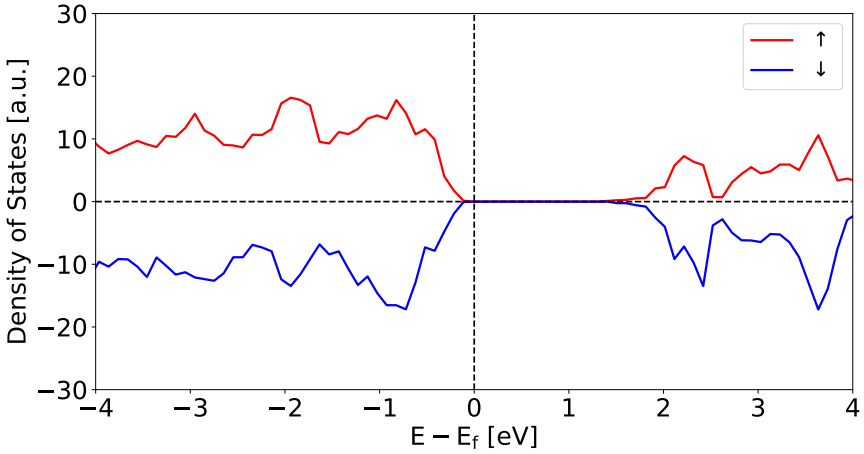

FIG. S13. Density of states calculated for the spin configuration $++++----+$ listed in Table S1.

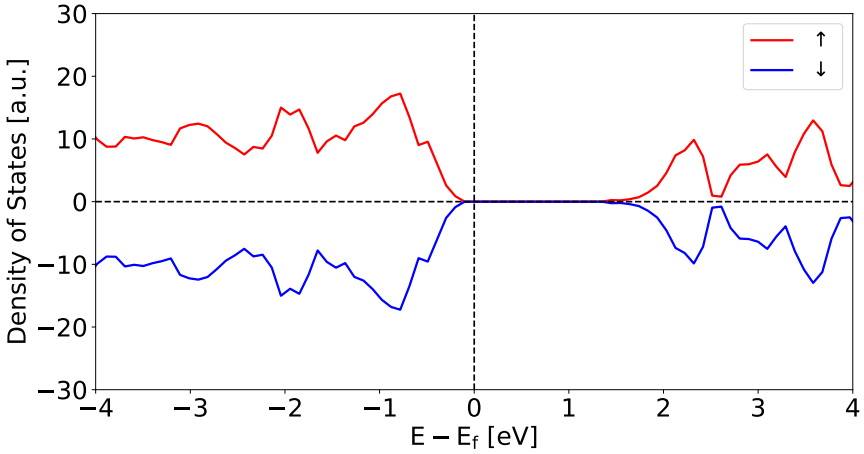

FIG. S14. Density of states calculated for the spin configuration $++++----+$ listed in Table S1.

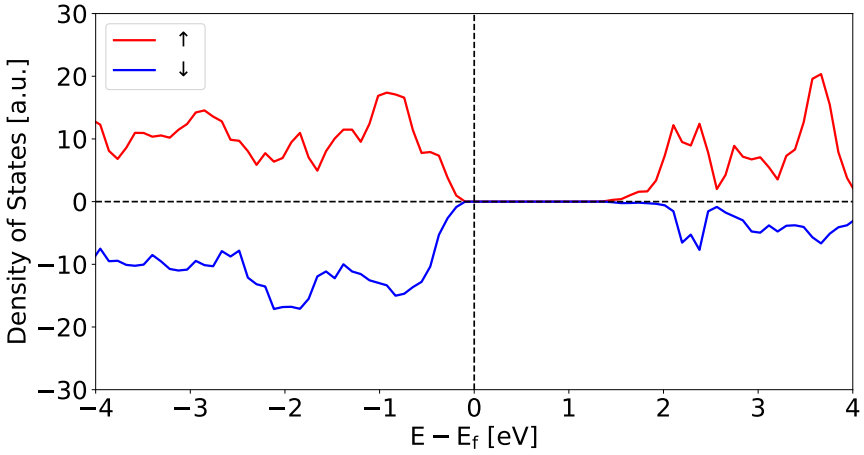

FIG. S15. Density of states calculated for the spin configuration $+------+$ listed in Table S1.

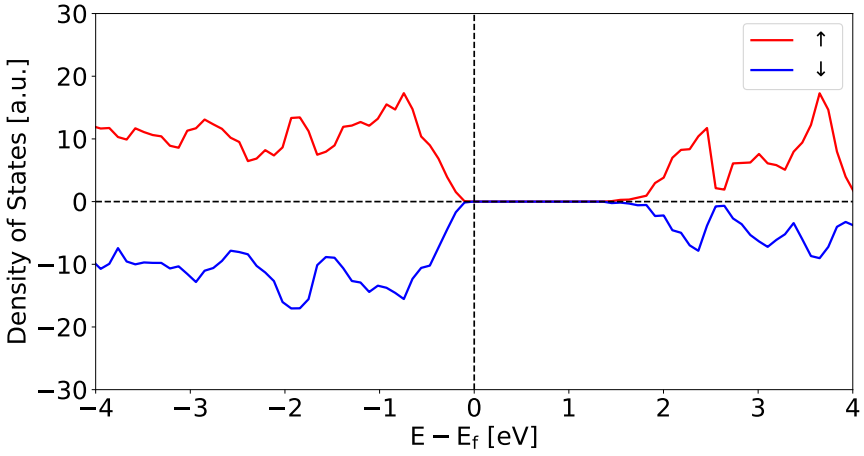

FIG. S16. Density of states calculated for the spin configuration $++-----+$ listed in Table S1.

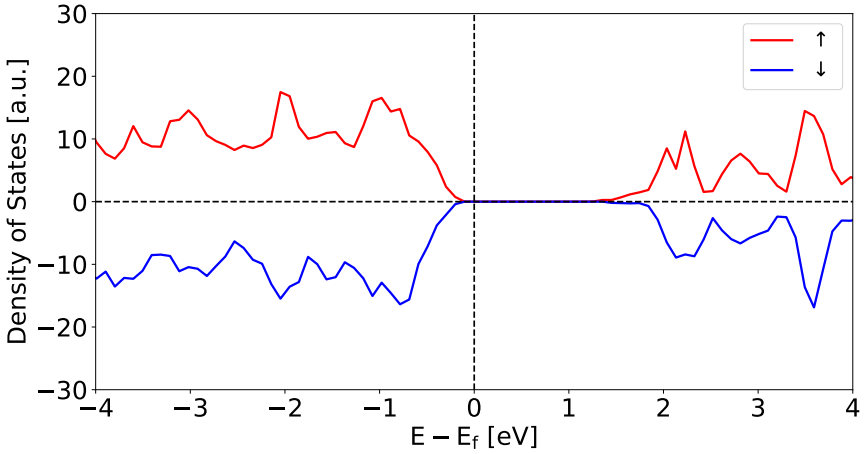

FIG. S17. Density of states calculated for the spin configuration $+-+-+-+-$ listed in Table S1.

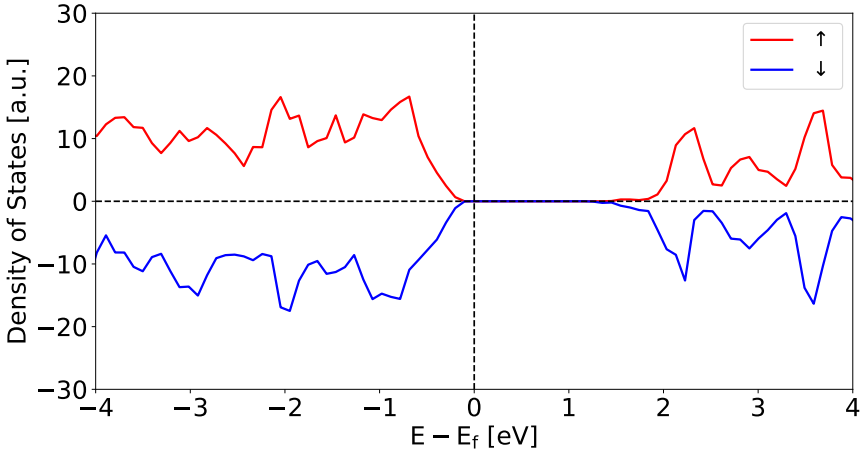

FIG. S18. Density of states calculated for the spin configuration $++--++--$ listed in Table S1.