

## Supplementary materials

### Ab initio investigation of electronic and lattice properties of $\text{Fe}_4(\text{P}_2\text{O}_7)_3$

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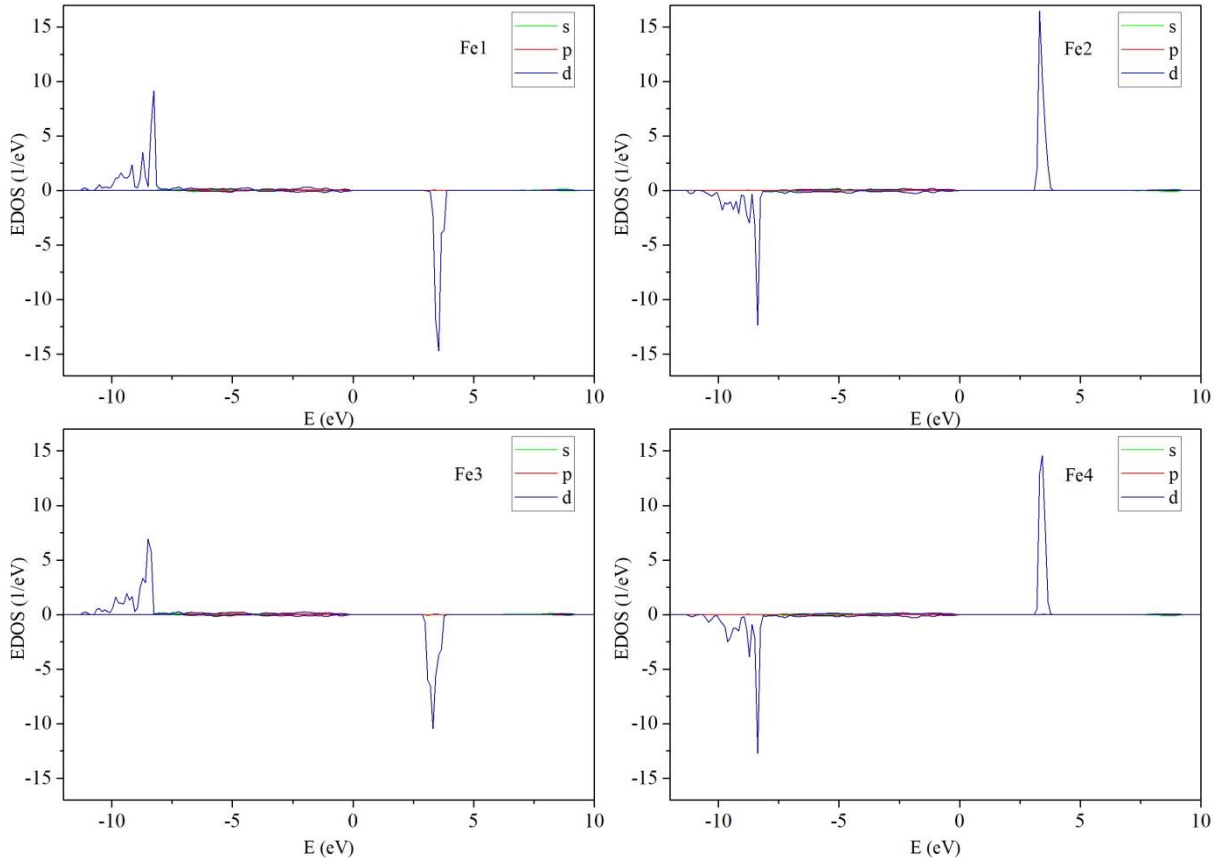
In this Supplementary Materials, we present the Wyckoff positions, magnetic moments and electronic density of states obtained for iron atoms at four different crystallographic sites in the iron pyrophosphate crystal. Additionally, we provide phonon frequencies and phonon density of states (PDOS) calculated for the primitive cell containing 124 atoms.

Table [S1] shows the magnetic moments for different antiferromagnetic states on four independent iron atoms compared with the experimental value.

S1. Magnetic moments in iron pyrophosphate compared with the experimental value.

	AFM1	AFM2	AFM3	AFM1 D2	Experimental[1]
$m_{\text{Fe1}} (\mu_B)$	4.602	4.602	4.600	4.602	4.55(5)
$m_{\text{Fe2}} (\mu_B)$	4.593	4.599	4.598	4.593	
$m_{\text{Fe3}} (\mu_B)$	4.571	4.577	4.574	4.572	
$m_{\text{Fe4}} (\mu_B)$	4.596	4.601	4.589	4.596	

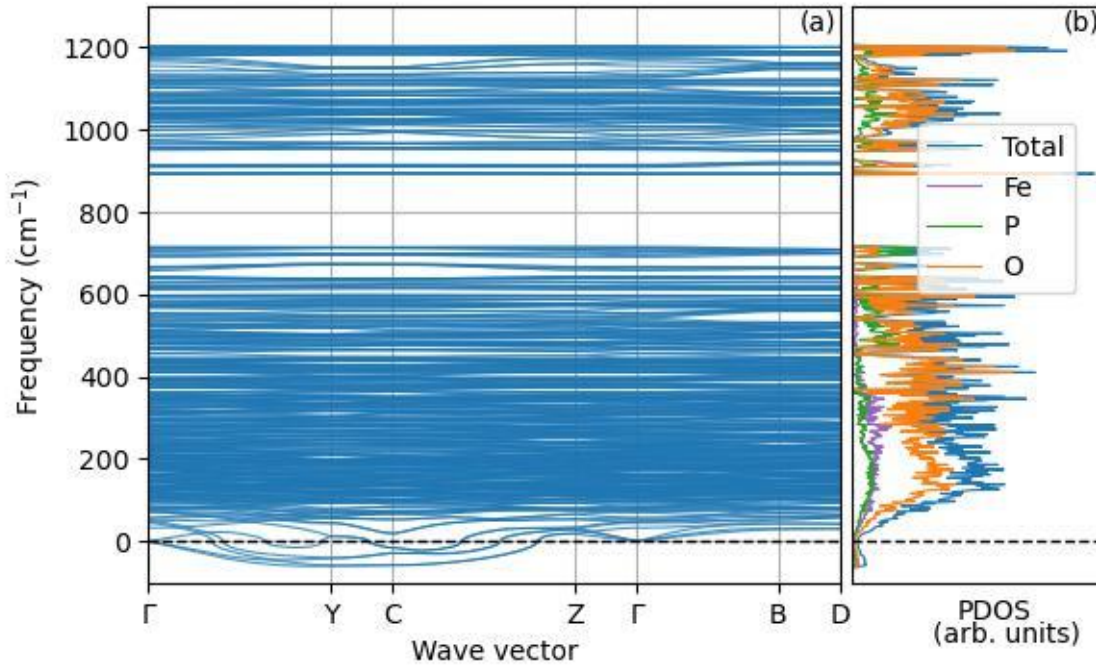
Figure [S2] illustrates the electronic density of states for four independent types of iron atoms.



S3. Wyckoff positions of independent atoms in the iron pyrophosphate structure.

Atom	x	y	z
Fe1	0.29376	0.24577	-0.28033
Fe2	-0.31382	0.24090	-0.29126
Fe3	0.22440	0.49273	0.26230
Fe4	-0.18834	-0.49179	0.25673
P1	0.21952	0.30029	0.44515
P2	0.28449	0.43969	-0.47109
P3	-0.28256	0.45413	-0.03202
P4	0.07063	-0.35718	0.34467
P5	-0.22764	0.31381	-0.00732
P6	-0.40189	0.38833	0.32974
O1	0.00531	-0.41941	0.26724
O2	-0.37723	0.28293	-0.15215
O3	0.14526	0.47556	0.38983
O4	0.25170	0.37000	0.48676
O5	-0.12454	0.47134	0.12553
O6	0.35786	0.26474	-0.41565
O7	-0.28951	0.43054	0.28819
O8	-0.47778	0.17290	-0.25323
O9	0.46517	-0.45253	0.38868
O10	0.39630	0.41813	0.31300
O11	-0.32844	0.19193	-0.43056
O12	-0.18667	-0.44967	0.39523
O13	-0.21735	0.31813	-0.32310
O14	0.14124	0.32277	-0.30405
O15	0.01834	0.20492	-0.42340
O16	0.21907	-0.37111	-0.49082
O17	-0.46893	-0.45814	0.09609
O18	0.32084	0.21084	-0.12819
O19	-0.22527	0.38496	-0.04112
O20	-0.02862	0.28675	0.36397
O21	-0.21982	0.49106	-0.10922

Figure [S4]: a) Phonon dispersion relations and b) phonon density of states (PDOS) calculated for primitive cell (1x1x1), including anharmonic effects, computed using the temperature-dependent effective potential (TDEP) approach [2,3] with force-constant matrices obtained via high-efficiency configuration space sampling (HECSS) [4]. The calculations were performed at T=100 K, and phonon energies and polarization vectors were determined through the exact diagonalization of the dynamical matrix.



## References

- [1] L. K. Elbouaanani, B. Malaman, R. Gérardin, and M. Ijjaali. Crystal structure refinement and magnetic properties of  $\text{Fe}_4(\text{P}_2\text{O}_7)_3$  studied by neutron diffraction and Mössbauer techniques. *Journal of Solid State Chemistry*, 163(2):412–420, 2002.
- [2] O. Hellman, I. A. Abrikosov, and S. I. Simak. Lattice dynamics of anharmonic solids from first principles. *Phys. Rev. B*, 84:180301, Nov 2011. doi: 10.1103/PhysRevB.84.180301.
- [3] O. Hellman, P. Steneteg, I. A. Abrikosov, and S. I. Simak. Temperature dependent effective potential method for accurate free energy calculations of solids. *Phys. Rev. B*, 87:104111, Mar 2013. doi:10.1103/PhysRevB.87.104111.
- [4] P. T. Jochym and J. Łażewski. High Efficiency Configuration Space Sampling – probing the distribution of available states. *SciPost Phys.*, 10:129, 2021. doi: 10.21468/SciPostPhys.10.6.129.