Supplementary Materials for:
Orthogonal magnetic structures of Fe_4O_5: representation analysis and DFT calculations

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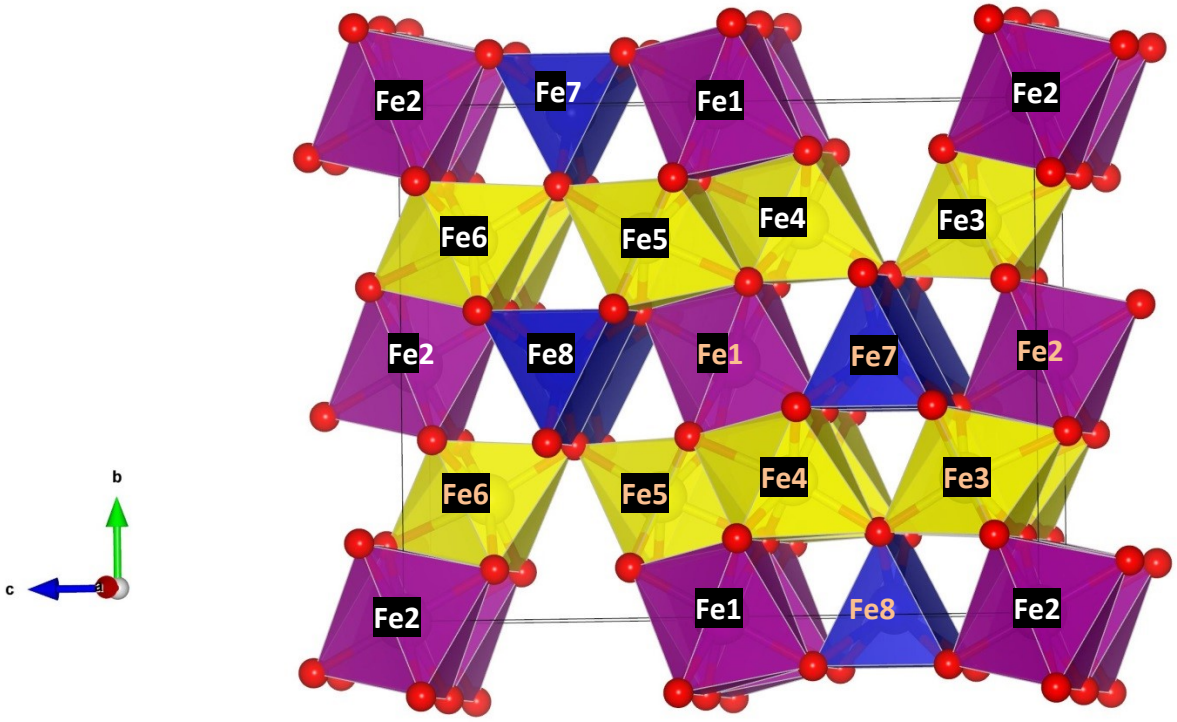


Figure S1. The numbering of the magnetically independent atoms Fe*i* (*i*=1-8) for the magnetic propagation vector k = (0, 0, 0). For k = (1/2, 0, 0) the magnetic moment of *j*'-th atom is transformed according to the expression $m(R_j + a) = -\Psi(R_j)$, where $\Psi(R_j)$ is a basis vector of *i*-th Fe atom and a is a lattice constant, $R_j' = R_j + a$.

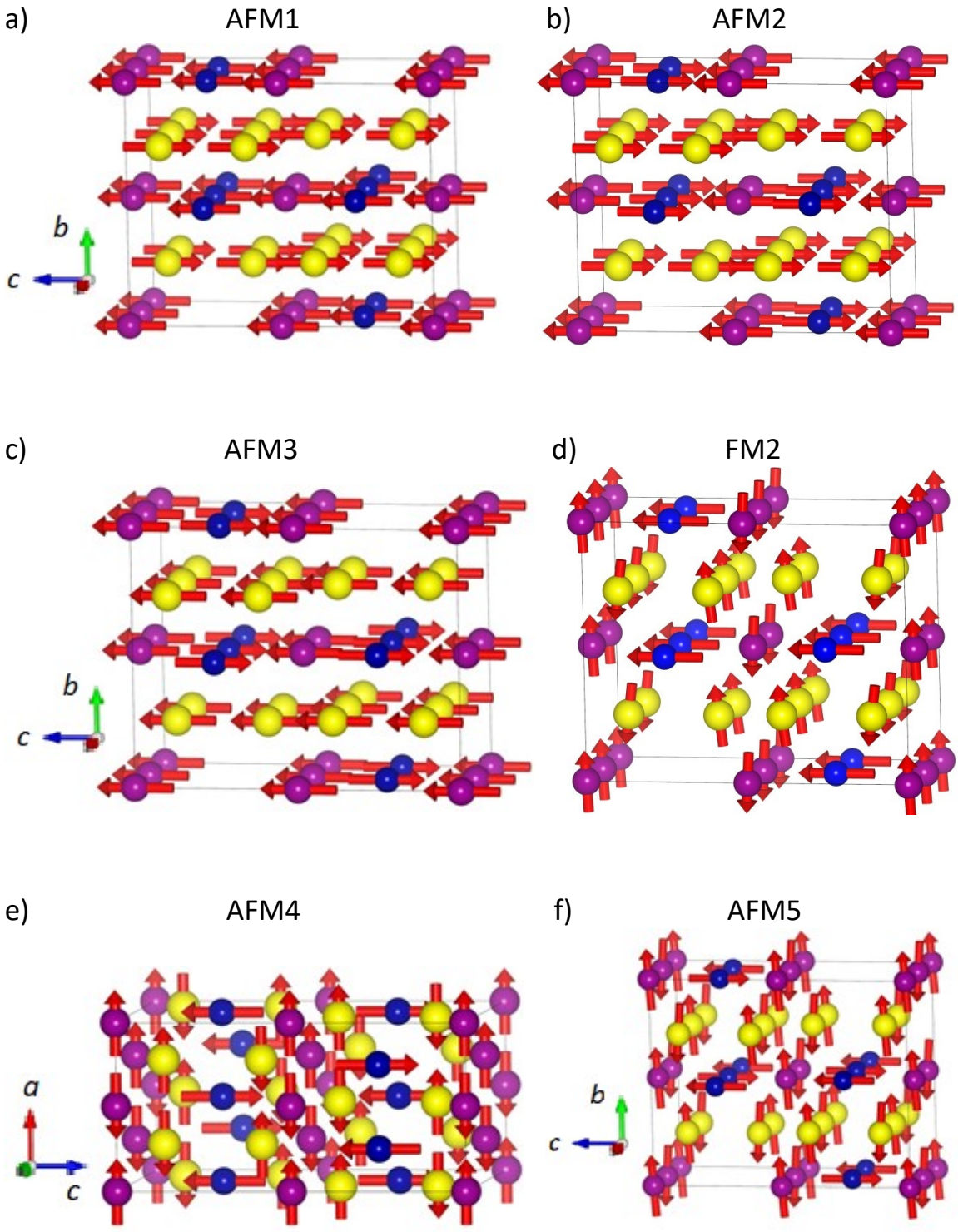
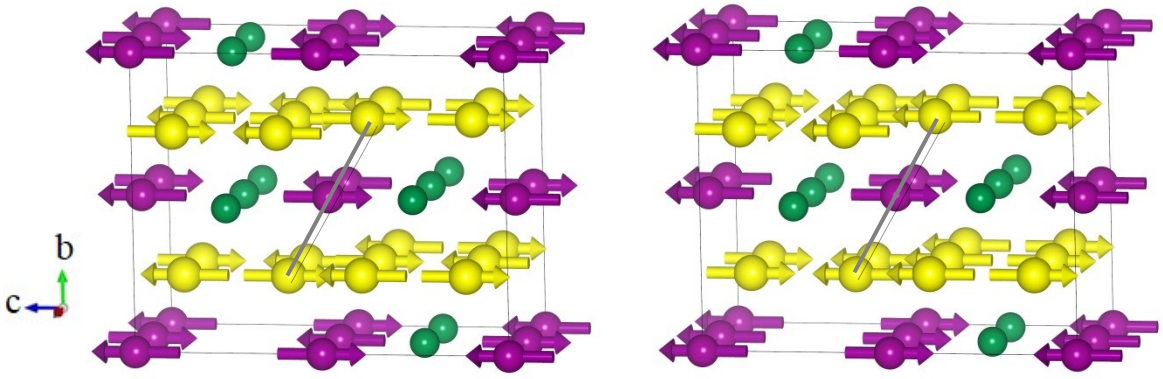


Figure S2. The AFM1-AFM3 (a-c), FM2 (d) and AFM4-5 (e-f) magnetic structures. All magnetic structures are shown within doubled unit cell along the *a* axis. The arrows show the directions of the Fe spins at the Fe1 (purple), Fe2 (yellow), and Fe3 (blue) sites.

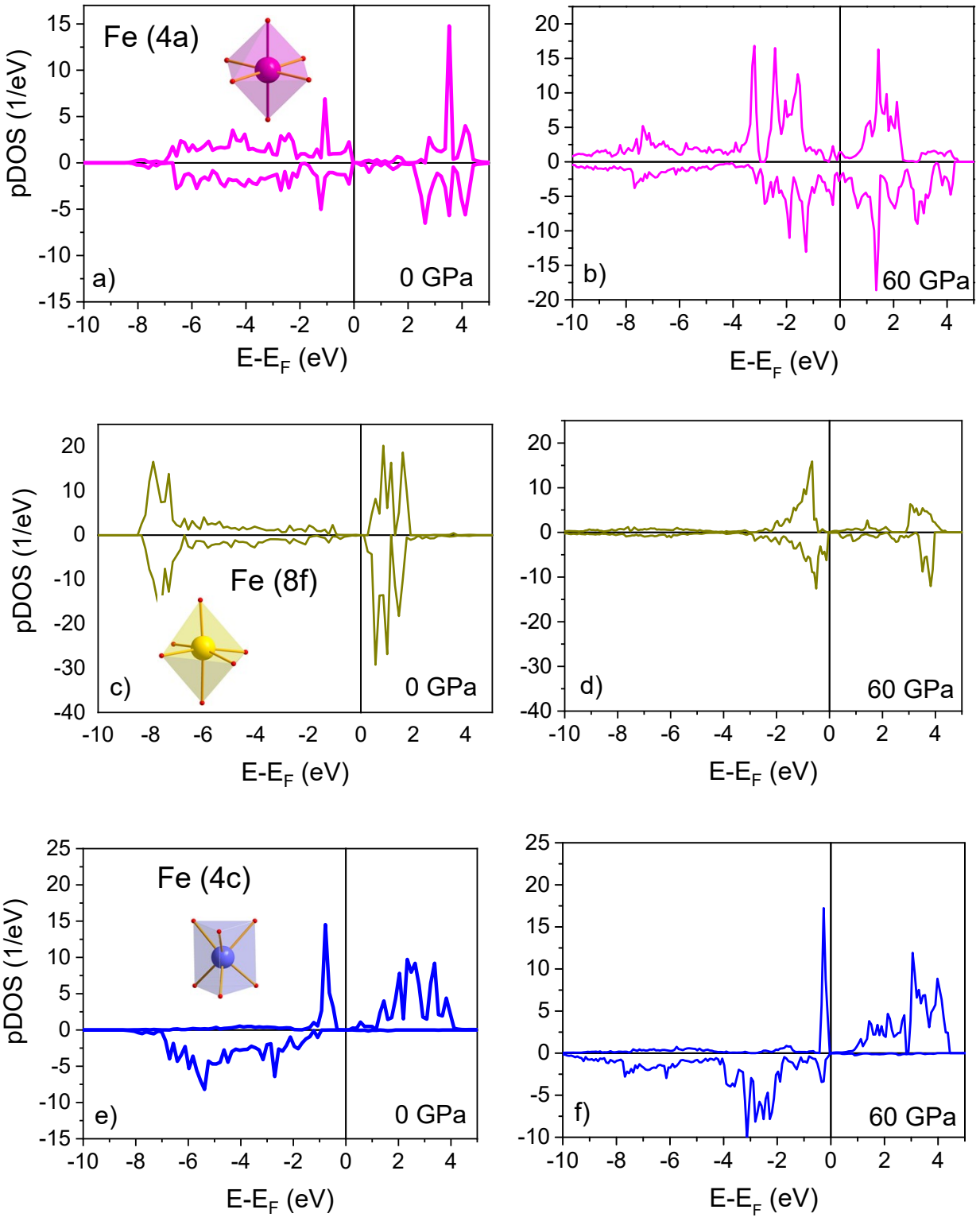
Table S1. The spin configurations, Fe magnetic moment components, and total energies for the AFM6 and FM3 phases in $CaFe_3O_5$. The arrows show the spin alignment within Fe2-Fe1-Fe2 triad, the ↑ stands for spin up and ↓ for spin down.

Magnetic (charge) ordering	Magnetic propagation vector, *k*	Spin config.	ΔE (meV/Fe atom)	Magn. moment (μ_B)	
				Fe1(4a)	Fe2(8f)
AFM6	(1/2, 0, 0)	↑↓↑	0	(0.00, 0.00, ±3.67)	(0.00, 0.00, ±4.10)
FM3	(0, 0, 0)	↑↑↑	6.5	(0.00, 0.00, ±3.60)	0.00, 0.00, ±4.15)



(a) (b)

Figure S3. The orthogonal antiferromagnetic structures of $CaFe_3O_5$ for the AFM6 phase with propagation vector k = (1/2, 0, 0) (a) and FM3 phase with k = (0, 0 ,0) (b). The magnetic structure of FM3 phase is shown within doubled cell along the *a*-axis. The arrows show the directions of the Fe spins at the 4a (purple) and 8f (yellow) sites. The nonmagnetic calcium atoms at 4c sites are highlighted in green The dashed lines indicate the ferromagnetic (a) and antiferromagnetic (b) orderings within triad Fe2-Fe1-Fe2.


Fe (4a)
a)
0 GPa
b)
60 GPa
Fe (8f)
c)
0 GPa
d)
60 GPa
Fe (4c)
e)
0 GPa
f)
60 GPa
pDOS (1/eV)
E-E$_F$ (eV)

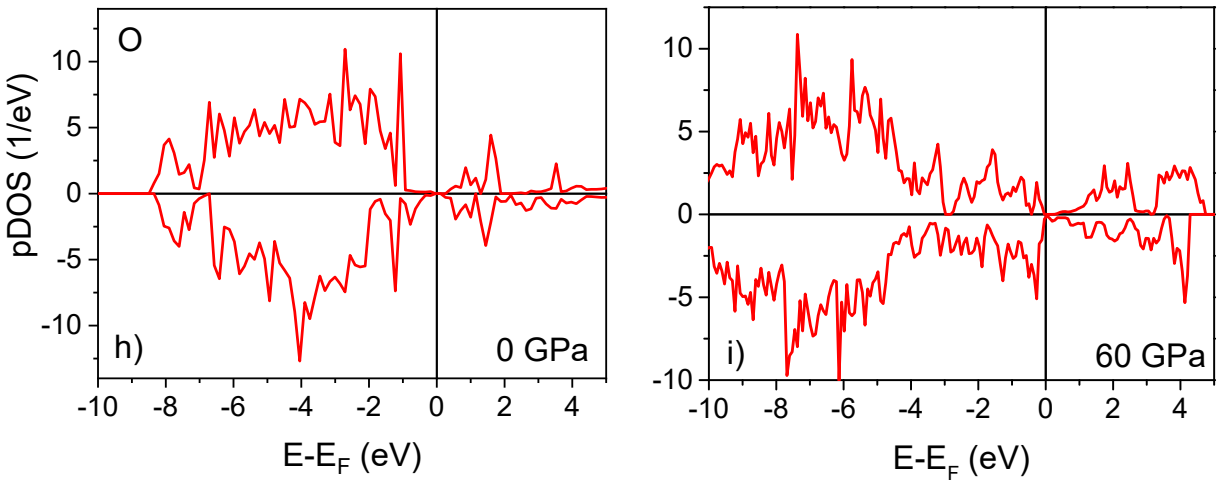


Figure S4. The iron projected density of *d*-electronic states (DOS) (a, b) Fe1, (c, d) Fe2, (e, g) Fe3; (h, i) oxygen projected density of *p*-electronic states of Fe_4O_5 at ambient pressure and P_{MIT}=60 GPa. The straight lines correspond to Fermi energy. Negative values of DOS show spin-down states.