Supporting Information

CHARMM-format topology file—please see file "FeFe_hydrogenase_clusters.top" available on the ACS Website.

CHARMM-format parameter file—please see file "FeFe_hydrogenase_clusters.par" available on the ACS Website.

Example VMD/PSFGen build script—please see file "1FEH_build.tcl" available on the ACS Website.

Table S1. DFT-optimized [4Fe4S]Cys4 circumsphere radii

	Fe	Sulfide	Protein ligands
Oxidized Cys_4^{2+}	1.619	2.145	3.942
Reduced Cys_4^{+}	1.736	2.331	4.175
Oxidized Cys_3His^{2+}	1.601	2.099	3.766
Reduced Cys_3His^{+}	1.801	2.280	4.088

Table S2. Average values of geometric parameters evaluated over [4Fe4S] clusters contained in the Protein Data Bank.

Fe-sulfide bond length	2.31 ± 0.09
Fe-external ligand bond length	2.28 ± 0.08
Fe-sulfide circumcenter origin distance	0.088 ± 0.068
Fe-external ligand circumcenter origin distance	0.080 ± 0.073
Fe circumcenter radius	1.67 ± 0.04
Sulfide circumcenter radius	2.25 ± 0.09
External ligand circumcenter radius	3.93 ± 0.07

Figure S1. Distribution of circumcenter theta values of [4Fe4S] clusters found in the Protein Data Bank.

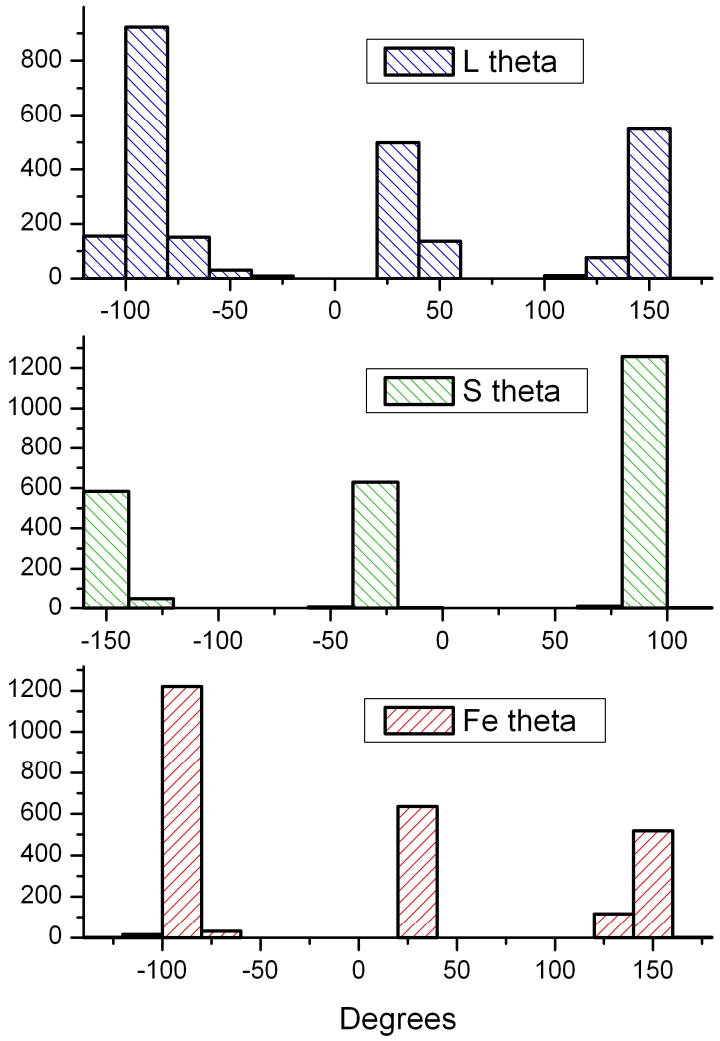
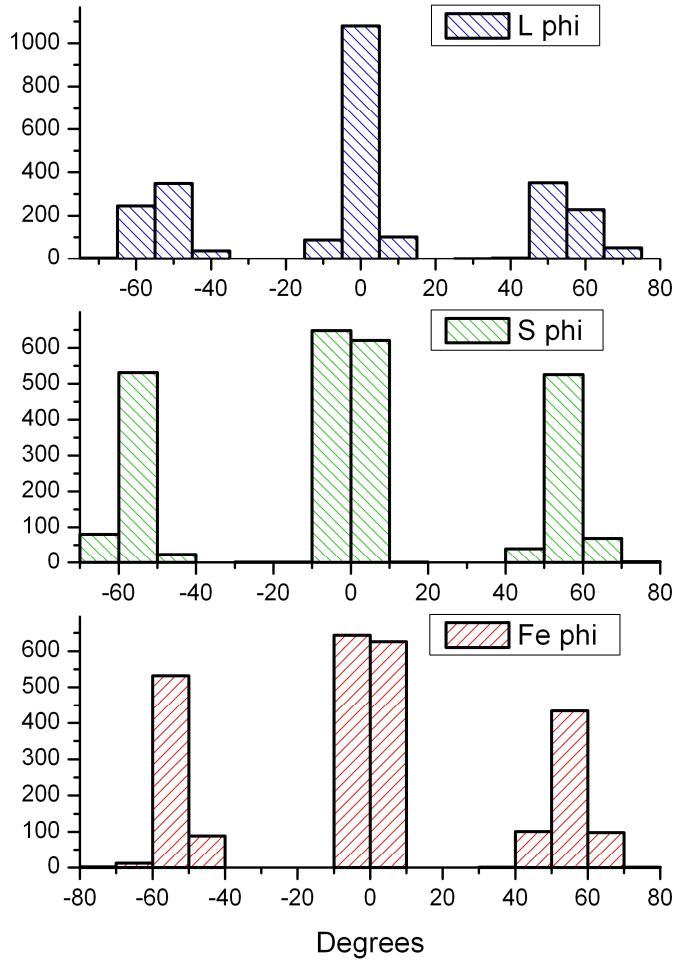


Figure S2. Distribution of circumcenter phi values of [4Fe4S] clusters found in the Protein Data Bank.



Trajectory (32-bit DCD) and structure files for normal mode animations—please see zipped archives “oxidized_modes.zip” and “reduced_modes.zip” available on the ACS Website. To unpack them, create a directory, copy the archives there, and use “unzip” (Linux/UNIX) or a Windows utility to decompress and copy the files into that directory. To view, load the PSF file into VMD, add any of the corresponding DCD files, and start the animation.