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Ab-initio simulation of Heme using GGA+U: a step toward accurate spin-state energetics DAMIAN SCHERLIS, MATTEO COCOCCIONI, Materials Science and Engineering, MIT, P. H.-L. SIT, Physics, MIT, NICOLA MARZARI, Materials Science and Engineering, MIT — Enzymatic sites containing transition metals are among the most relevant biophysical systems currently studied using first-principles quantum mechanical approaches. In this context, however, the application of ab-initio methods is often severely limited as a consequence of the inability of conventional electronic structure methods - as Hartree-Fock or DFT - to provide a qualitatively correct description of the spin-state energetics of the metal center. In this work we apply the DFT based GGA+U approach to compute the low-lying states of ligated and unligated iron heme complexes. We show that this technique, in which the Hubbard- like correction is obtained in a fully ab-initio fashion using linear-response theory, is extremely useful for the treatment of organometallic compounds, in particular the heme system, where LDA, GGA, and common hybrid DFT functionals predict incorrect spin energetics. Calculations of ligand exchange thermodynamics, spin transitions, and other properties, point to GGA+U as an appealing tool to overcome the limitations entailed by the use of DFT in the description of bioinorganic complexes. Moreover, its straightforward implementation in a plane wave basis set code allows us to address systems of hundreds of atoms on commodity workstations.

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