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First principles EPR spectra of organic transition metal complexes DAVIDE CERESOLI, ELISE LI, NICOLA MARZARI, Massachusetts Institute of Technology — We present first principles, density functional theory calculations of the EPR spectrum of mononuclear and binuclear organic transition metal complexes, that constitute building block of more complicated catalysts for the water splitting reaction. We apply here the modern theory of orbital magnetization and we obtain the EPR g-tensor by computing the derivative of the orbital magnetization with respect to the electronic spin flip. This method allowed us to incorporate self-interaction corrections in the Hamiltonian, at the level of DFT+U. We found that the DFT+U method improves the agreement with respect to experiment, of the EPR g-tensor and hyperfine couplings parameters for high spin complexes. We also discuss the success and failures of DFT+U as an energy functional, and the importance of benchmarking improved- or post-DFT methods (such as hybrid functionals) against the EPR spectra of transition metals complexes.

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