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A GGA+U approach to realistic modeling of transition-metal complexes HEATHER KULIK, NICOLA MARZARI, DMSE, Massachusetts Institute of Technology — Despite the importance of transition metal centers in a variety of biological and inorganic chemical reactions, density functional theory calculations often fail quantitatively in describing both the stable intermediate electronic structures, splittings, and geometries as well as reaction barriers and geometries of transition states. We have shown[1] that augmenting the generalized-gradient approximation (GGA) with a Hubbard U which is obtained from a self-consistent linear response procedure can greatly improve the description of both spin state splittings in the iron dimer as well as reaction barriers in the addition-elimination reaction of hydrogen and methane with FeO^+. This fully ab-initio GGA+U approach provides excellent agreement with accurate, correlated-electron quantum chemistry calculations but at a fraction of the cost of these methods. We will further highlight how our method affords substantial improvement in the physical description of hybridization and bonding irrespective of system size. We thus fruitfully employ GGA+U in the study of large-scale complexes which contain hundreds of atoms such as the active site of halogenating enzymes and various porphyrin complexes.

[1] H. J. Kulik, M. Cococcioni, D. Scherlis and N. Marzari, PRL **97** 103001, (2006).

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