Computational Coordination Chemistry:
Harnessing Quantum Chemistry to Build Better Force Fields

Professor Robert J. Deeth, Universities of Bath and Warwick, UK

Abstract

Coordination complexes of transition metal ions are frequently paramagnetic with complicated electronic structures. These appear to demand complex, multi-reference quantum mechanical (QM) methods but such calculations can be extremely expensive. In contrast, ligand field theory (LFT) provides a far simpler, albeit parametric, approach to the multi-determinant states of predominantly d-orbital parentage. However, LFT alone is an insufficient basis for molecular modelling since it only generates an electronic energy without any nuclear contributions. The latter require some additional treatment, the simplest of which is classical molecular mechanics (MM). The ligand field/molecular mechanics (LFMM) combination efficiently captures the important structural and energetic consequences of d-electron effects such as Jahn-Teller distortions and spin-state changes. The challenge now is the recognition that a MM force field (FF) is only as good as its parameters. In this talk, using the spin states of d^6 Fe(II) complexes as examplars, the basis of the LFMM approach will be outlined and we will explore how to harness DFT and/or wavefunction methods to generate selective data upon which an accurate LFMM FF may be trained and then deployed to generate quantum-level accuracy at a tiny fraction of the cost of any QM-based method.