

# Abstract

The concept of magnetism has been known to humankind since several millennia, whereas the idea of a molecule working as a magnet is a rather recent discovery. Molecular magnets, also known as single molecule magnets (SMM), have great potential in data storage and quantum computing (**Chapter 1**). A typical SMM hosts a magnetic core or a spin centre surrounded by organic ligands that shield the magnetic core from intermolecular interactions. An essential quality of an SMM is its ability to show slow relaxation of magnetization controlled by a large magnetic anisotropy barrier. Electronic structure calculations with accurate treatment of multi-configurational interactions and spin-orbit coupling have been employed in this work to investigate the magnetic properties of a series of transitional metal based complexes (**Chapter 2**). In the search of efficient SMMs, the metal ions are considered pivotal in achieving large magnetic anisotropy barriers. To this end, the potential of bi-coordinated Co(II) complexes has been investigated. The strong spin-orbit coupling constants of Co(II) ions together with low coordination number keep the orbital angular momentum of the metal centre unquenched, which serves as a source of magnetic anisotropy. The electronic structure calculations reveal a Co(II) complex with effective anisotropy barrier of  $974\text{ cm}^{-1}$ , which is the largest anisotropy barrier shown by any transition metal complex reported so far. Additionally, the role of ligand field as well as steric crowding on governing the magnetic anisotropy has been investigated. A magneto-structural investigation reveals insightful observations on the relationship between conformational arrangement and magnetic anisotropy (**Chapter 3**). Mn(II) complexes are typically considered unsuitable as SMM candidates due to their vanishing orbital-angular momentum. However, in this thesis, it is demonstrated that in Mn(II) complexes with heavy ligand atoms, e.g., Br and I, several interesting inter-electronic state mixing occurs via the spin-orbit operator that results in large magnetic anisotropy (**Chapter 4**). In addition to the high-spin complexes described above, magnetic anisotropy in a pair of trigonal-bi-pyramidal (TBP) Fe(III) complexes has been investigated and the role of axial ligands in governing the stability and magnetic anisotropy of these TBP complexes has been established. A simple conformational change that enhances the symmetry in one of these studied complexes has been shown to influence the magnetic anisotropy to a large extent (**Chapter 5**). These intermediate-spin complexes are found to exhibit a high-spin state in close vicinity of the ground state. Hence, the probability of spin crossover between these two states has been investigated. While the possibility of a thermal crossover mechanism is ruled out due to a large underlying ground state barrier, an alternate low-temperature light-induced pathway for spin crossover has been illustrated. By following the minimum energy pathway along different electronic states, two possible excited state pathways have been outlined through which the intermediate-spin state can reach the high-spin state (**Chapter 6**).

**Keywords:** Single Molecule Magnets; Magnetic Anisotropy; Intermediate Spin; Spin Crossover; CASSCF Calculations.