

Elucidation of ligand fields, metal-metal interactions and phonon spectra for a new family of highly symmetric lanthanide single molecule magnets by use of INS.

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Description: Single-molecule magnets (SMMs), which are molecular paramagnets exhibiting a magnet-like behaviour in the absence of long-range magnetic order, have attracted considerable attention in field of chemistry and physics in recent years. Their nanoscopic size and the perfectly monodisperse nature points too future applications in spintronic devices^[1] and quantum computing.^[2] The simplest way of realizing an SMM is by placing a single lanthanide ion in an appropriate crystal field. Due to the subordination of crystal field effects to the spin-orbit coupling, the effect of the crystal field is to split the spin-orbit coupled ground J multiplet into its m_J substates. This crystal field splitting generates an intrinsic “barrier” for reversing the magnetic moment of the molecule, pictorially equal to the anisotropy barrier responsible for the superparamagnetic behaviour of magnetic nanoparticles. While this picture is pleasingly simple, it has been shown to be severely oversimplified by us^[3] among others. The simplification lies in the over-barrier process assumed operative for the relaxation of the magnetization to the surrounding lattice. While this process, the so-called Orbach process, is relevant in some cases, the Raman process, in which energy is transferred from the spin system to the lattice by the inelastic scattering of a lattice phonon by the spin system, has been found equally important.^[3] Additional processes exist, and the spin relaxation of SMMs seem to rely on a delicate balance between the spin system and the surrounding lattice phonons, in accordance with old spin-lattice relaxation theory.^[4,5]

The project is based on a new type of lanthanide SMMs where the lanthanide ion is sandwiched between two metallo-ligands each providing squares of oxygen donor atoms for the lanthanide ion. These systems can be obtained in a perfectly tetragonal environment and offer ample possibilities for chemical tuning. An interesting added feature of these systems is the possibility of using metallophilic interactions between the surface exposed noble-metals to tailor more complicated multimetallic structures of possible interest in quantum information processing schemes.

In the Ph.D. project, select members of this series will be studied by INS spectroscopy, aiming at mapping out the ligand field, which is a manageable task due to the high symmetry of the systems. Additionally, determination of the phonon spectra and their importance for the relaxation processes of these systems is a central goal of the project.

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