

***Ab initio* Study of the Magnetic Anisotropy in Lanthanide-Based Single-Molecule Magnets**

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The two main factors for observing a slow magnetic relaxation at a molecular level are a high spin value and a large magnetic anisotropy. Because of that, in the last years there has been a growing interest in the use of lanthanide ions in the synthesis of Single Molecule Magnets[1] and Single Chain Magnets[2] since their usual high magnetic moments and large magnetic anisotropies. However, the treatment of the magnetism in this kind of compounds is not so straightforward than in transition-metal clusters because of the strong spin-orbit coupling and deeper theoretical studies are required.

In this contribution we will show several examples of molecular compounds containing lanthanide ions and showing slow magnetic relaxations in which the combination of *ab initio* calculations and Hamiltonian models has allowed to understand their experimental magnetic behaviour.

In the employed *ab initio* method, CASSCF/RASSI-SO[3], spin-free wavefunctions and energies are determined through the use of the Complete Active Space Self Consistent Field(CASSCF) method. Then, Spin–Orbit coupling is treated with a Restricted Active Space State Interaction computation(RASSI-SO). In the presented work, this quantum chemistry method has proved to be of great help in the theoretical study of lanthanide-based materials and, in particular, for predicting the direction of the easy single-ion anisotropy axes of the lanthanides ions and the nature of their ground and the low energy excited states. The results of the presented studies are very promising for the use of the CASSCF/RASSI-SO quantum chemistry method in order to shed light in the complex magnetic behaviour of the lanthanide-based magnetic molecular materials.

[1] N. Ishikawa *et al.*, *J. Am. Chem. Soc.*, **125**, 8694 (2003)

[2] K. Bernot *et al.*, *J. Am. Chem. Soc.*, **128**, 7947 (2006)

[3] B.O. Roos, P. A. Malmqvist, *Phys. Chem. Chem. Phys.*, **6**, 2919 (2004)