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Elucidation Of The Wave Function Of The Ground Doublet In A Tb Complex Using INS In A Magnetic Field

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Lanthanoid Single Molecule Magnets (SMMs) are molecular materials that exhibit slow relaxation of the magnetisation of molecular origin, thus showing promise for a wide range of technological applications, such as spintronic devices, qubits for quantum computers and molecular memories. The origin of the energy barrier to the reorientation of the magnetic moment in a lanthanoid SMM lies in the weak splitting of the atomic lowest-lying spin-orbit multiplet of the Ln^{III} ion, induced by the electron density of the ligands, into a set of crystal field states. Since the crystal field splitting is significantly weaker than other energy contributions in lanthanoid complexes, the details of the ligand-Ln interaction are crucial for the development of SMMs which can operate at high temperatures.

For this reason, it is crucial to achieve a detailed understanding on how the crystal field influences the relaxation of Ln^{III} SMMs, and to this end computational and theoretical methods have been very useful to rationalise experimental results, and are routinely employed in the molecular magnetism literature. On the other hand, comparison of simulations with different experimental measurements can showcase the shortcomings of the theoretical or computational approach, providing a rationale on the improvements to introduce in order to improve their ability to successfully describe the properties of the system.

Previously, we have performed Inelastic Neutron Scattering (INS) measurements on the complex $[Tb(bpy)_2(Cl_4Cat)(Cl_4CatH)(MeOH)]$ using the PELICAN instrument, where we observed a shoulder to the elastic line which, after fitting, was established to arise from a single transition. While theoretical calculations of the low-lying energy spectrum of the complex rationalized the observed transition as arising from the quantum tunnelling of the ground Ising doublet, the predicted tunnelling gap is significantly smaller than what observed experimentally, highlighting the inability of such calculations to correctly predict the electronic properties of the ground state of the molecule.

In this study, therefore, we have measured the INS spectra for the aforementioned complex, under the same experimental conditions of the previous experiment, employing the newly commissioned magnet on PELICAN, where the evolution of the observed transition in an increasing magnetic field would provide useful information on (i) the properties of the ground state and (ii) the nature of the magnetic transition observed in the zero-field spectrum. The measurements show that, with increasing magnetic field, the INS transition (i) becomes less intense, thus confirming our theoretical description of the observed peak as arising from the quantum tunnelling of the ground state, and (ii) splits into multiple peaks, both as a result of the usage of a powder sample and the complex spin-orbit composition of the wave function of the ground Ising doublet.

Level of Expertise

Early Career <5 Years

Speakers Gender

Male

Do you wish to take part in the poster slam

Yes

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