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MUSR evidence of a marked exchange-interaction effect on the local spin dynamics of Tb-based molecular nanomagnets

Single Molecule Magnets (SMMs), depending on the molecular ligand, experience a different interaction (exchange and spin orbit/phonon coupling, crystal-field effects, etc.). Their main drawback is the presence of multiple relaxation pathways accelerating magnetization loss and limiting the blocking temperature. We report our investigation on local spin dynamics of two isostructural terbium(III)-based complexes: [Tb(DTBSQ)(HBPz3)2] (Tb-SQ) and [Tb(trp)(HBPz3)2] (Tb-trp) through ac susceptometry and muon-spin rotation (MUSR) as a function of temperature and magnetic field. The paramagnetic semiquinonato (SQ) ligand gives an effective lanthanide-radical exchange interaction, while the tropolonato (trp) ligand affords a very similar ligand field, but is diamagnetic. The study highlights that: (i) the combined effect of the ligand field on terbium(III) and of the terbium(III)-radical exchange interaction produces a magnetic anisotropy barrier to the relaxation; (ii) the suppression of the exchange interaction channel for relaxation and of the magnetic anisotropy barrier in Tb-trp generates unusual spin dynamics for SMMs.

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