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Muon-spin relaxation investigation of magnetic bistability in a molecule-based material

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We present the results of a muon-spin relaxation (μ^+SR) investigation of the crystalline organic radical compound 4-(2-benzimidazolyl)-1,2,3,5-dithiadiazolyl (HbimDTDA), in which we demonstrate the hysteretic magnetic switching of the system that takes place at $T = (249 \pm 13)$ K caused by a structural phase transition. Muon-site analysis using electronic structure calculations suggests a range of candidate muon stopping sites. The sites are
numerous and similar in energy, but, significantly, differ between the two structural phases of the material. Despite the difference in the sites, the muon remains a faithful probe of the transition, revealing a dynamically-fluctuating magnetically disordered state in the low-temperature structural phase, which was previously believed to be diamagnetic. This is evidenced by relaxation following the Redfield formula in longitudinal field (LF) measurements, which is observed
only in the low temperature phase. In contrast, in the high temperature phase the relaxation is caused by static nuclear moments, with rapid electronic dynamics being motionally narrowed from the muon spectra.

Primary authors: HERNANDEZ-MELIAN, Alberto (Durham University); HUDDART, Benjamin (Durham University); PRATT, Francis (STFC); BLUNDELL, Stephen (University of Oxford); Dr MILLS, Michelle (University of Guelph); LANCASTER, Tom (Durham University); Dr PREUSS, Kathryn (University of Guelph)

Presenter: HERNANDEZ-MELIAN, Alberto (Durham University)

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