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Detection of Spin State Ordering Phenomena in Mn(III) Spin Crossover Solids by Muon Spin Relaxation

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Spin crossover (SCO) compounds are metal-ligand complexes which can easily switch their internal arrangement of electrons in d-orbitals from paired (low-spin) to unpaired (high-spin), by a minor variation in temperature, pressure, magnetic or electric field, or via irradiation. SCO is observed for some metal-organics containing Fe(II), Fe(III), Mn(III) or Co(II) ions, and the switching is associated with bond length changes of up to 0.2 Å in each metal-donor distance due to depopulation/population of anti-bonding orbitals during the electron pairing/unpairing process. SCO with Jahn-Teller ions such as Mn(III) or Co(II) is of particular interest as the Jahn-Teller distortion can be easily injected into or removed from a lattice by any of the perturbations described above.

To date μSR has been used to follow thermal SCO in only a handlful of Fe(II) compounds. We report now our recent results on using μSR to follow SCO in a Mn(III) complex for the first time. We show that multiple structural symmetry breaking transitions along the polar space group sequence: Cc ↔ Pc ↔ P1 ↔ P1(½)[1]can be successfully detected by muon spin relaxation measurements and that the technique also confirms the emergence of domain walls at different temperatures.

[1] V. B. Jakobsen, E. Trzop, E. Dobbelaar, L. C. Gavin, S. Chikara, X. Ding, K. Esien, H. Müller-Bunz, S. Felton, E. Collet, M. A. Carpenter, V. S. Zapf, G. G. Morgan, J. Am. Chem. Soc, 2022, 144, 195-211.

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