



Contribution ID: 127

Type: **Poster Presentation**

Probing Domain Wall Dynamics in a Spin-State Ordered Manganese Spin Crossover Crystal

A novel domain wall architecture has recently been discovered in molecular crystals of the ferroelastic Mn^{3+} spin crossover compound $[\text{MnIII}(3,5\text{-diCl-sal}_2(323))]\text{BPh}_4$. [1] This complex exhibits a two-step thermal spin transition between the d^4 spin triplet and spin quintet states, with a spin-state ordered phase forming at low temperatures. Single-crystal X-ray diffraction identified three distinct symmetry-breaking phase transitions along the polar space group sequence: $\text{Cc} \leftrightarrow \text{Pc} \leftrightarrow \text{P1} \leftrightarrow \text{P1}(\frac{1}{2})$. Acoustic spectroscopy detected both pinned and mobile ferroelastic domain walls responding to mechanical stress during the low-temperature phase transitions.

To gain deeper insight into the distribution of spin states within the domains and to monitor domain wall dynamics at the higher temperature transition, muon spin relaxation (μ SR) measurements were performed. Evidence from the μ SR data signals the presence of multiple domain types—pinned, mobile, and domain-free regions, confirming the earlier acoustic data.

[1] V. B. Jakobsen et al. J. Am. Chem. Soc, 2022, 144, 195-211.

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Science Foundation of Ireland

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Session Classification: Poster session 2

Track Classification: Magnetism