Change in Electric Polarization of a Double Step Spin-State Transition Mn^{3+} Complex

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Introduction

The investigation and creation of multifunctional materials which exhibit a change in magnetic order and changes in electric polarization continue to fascinate. We have previously established that coupling of the magnetic field to the change in electric polarization is possible in transition metal compounds which show a change in spin state, e.g. the S=1 → S=2 spin state change in Mn^{3+}. This was achieved by measuring the Mn(taa) complex in high magnetic fields[1] where it was shown that the complex becomes ferroelectric via lattice strains that originate with a magnetic spin state transition and therefore the ferroelectricity does not require any long range magnetic order. We have subsequently measured other Mn-containing metal organic complexes that show coupling of the magnetism and electric polarization. Here we present a Mn^{3+} compound which undergoes a two-step thermal spin-state transition (SST) where the magnetism and change in electric polarization are strongly coupled to each other.

Experimental

We performed pulsed field magnetization measurements to track the magnetic field induced spin state transition and calibrated the pulsed field magnetization data on a 13 T PPMS (Quantum Design). The change in electric polarization in the polar Mn^{3+} compound was measured with pulsed magnetic fields up to 65 T.

Results and Discussion

We have synthesized a new molecular Mn^{3+} compound which shows a two-step thermal SST. We have observed a robust change in electric polarization in a single crystal of the polar Mn^{3+} compound by applying pulsed magnetic fields up to 65 T in the lower of the two thermal hysteresis windows, **Fig. 1a**. In the upper hysteretic temperature window, the polarization magnitude decreases but surprisingly the sign of the polarization is switched (**Fig. 1a**). Moreover, we observed that the change in electric polarization becomes remnant, at 90 K in the lower hysteresis window suggesting that the structural phase transition associated with the spin state transition persists after the magnetic field is removed. By not resetting the system by cooling down to 40 K between pulses, we have observed a memory effect at 90 K where no change in electric polarization is observed when the second pulse is fired after 45 min (**Fig. 1b**).

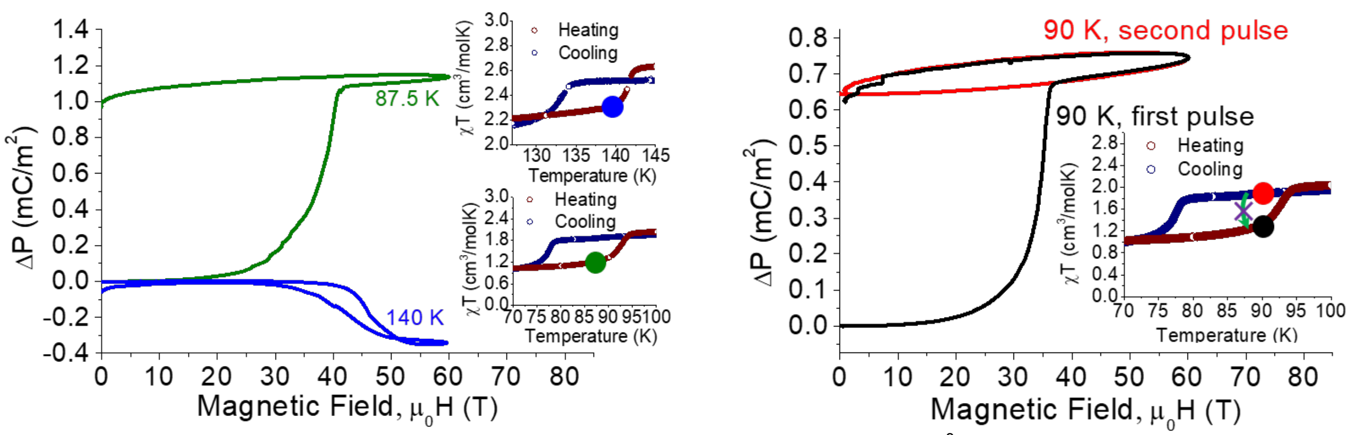

Fig. 1a: Change in electric polarization in the lower hysteresis window of the SST Mn^{3+} compound at 87.5 K (green) reaching the maximum value at 1.15 mC/m^2 at 40 T. The change in the electric polarization at 140 K (blue) becomes negative relative to the sign of the change in electric polarization in the first hysteresis window; **Fig. 1b**: At 90 K, the change in electric polarization becomes remnant and after 45 min another 60 T pulse was fired revealing no change in electric polarization.

Acknowledgements

The National High Magnetic Field Laboratory is supported by the National Science Foundation through NSF/DMR-1157490/1644779 and the State of Florida. Funded research trip to LANL by Science Foundation Ireland 12/IP/1703 (G.G.M.) and Studielegat Augustinus Fonden no.18-0338 (V.J.) is also gratefully acknowledged.

References

[1] Shalinee Chikara, Shizeng Lin, Nathan Smythe, John Singleton, Brian Scott, Elizabeth Krenkel, Jim Eckert, Cristian D. Batista, and Vivien S. Zapf, "Multiferroic behavior via a spin state transition," submitted