

Magnetoelectric coupling at a transition between two spin statesS. Chikara^{1,2}, J. Gu³, X.-G. Zhang³, H.-P. Cheng³, N. Smythe⁴, J. Singleton¹, B, Scott⁴, E. Krenkel⁵, J. Eckert⁵, and V. S. Zapf¹1. NHMFL-PFF2. NHMFL-DC3. University of Florida4. Los Alamos National Lab5. Harvey Mudd College

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Traditionally, magnetoelectric coupling and multiferroic behavior are studied in inorganic oxide materials in which ferromagnetic order or antiferromagnetic order couple to electric polarization. Materials with magnetoelectric coupling have potential application in low-power magnetic sensing, new computational devices, and high-frequency electronics.

MagLab users studied a spin state transition, a different type of magnetic order common in molecular materials that offers greater coupling to the lattice and therefore enhanced potential for magnetoelectric coupling. At the low-spin to high-spin transition in a Mn^{3+} -based molecular material denoted "Mn(taa)", the spin state changes from S = 1 to S = 2 at a first-order phase transition. The high spin state is Jahn-Teller active, which allows three choices for distorting the molecules in the crystalline lattice, thereby creating electric dipoles. These electric dipoles undergo different orderings controlled by magnetic field and temperature. The upper figure shows the DC-field phase diagram of Mn(taa) with three different ferro- or para-electric phases depending on magnetic field and temperature.

This work demonstrates that multi-ferroic-like magnetoelectric behavior is possible using a spin state transition with a resulting magnetic-fieldinduced electric polarization that is within 10% of the record of any magnetoelectric material. This discovery vastly expands the possible materials and types of magnetic orders at which magnetoelectric coupling can be studied. In molecular materials, such spin state transitions are common and widely studied, since the soft lattice accommodates the change in orbital occupation that accompanies the spin state transition.

Facilities used: 45T Hybrid Magnet; 65T Pulsed Magnets. **Citation**: S Chikara, J Gu, X-G Zhang, H-P Cheng, N Smythe, J Singleton, B Scott, E Krenkel, J Eckert, and VS Zapf. "Magnetoelectric behavior via a spin state transition," *Nature Comm.* **10**, 4043 (2019) **doi.org/10.1038/s41467-019-11967-3**

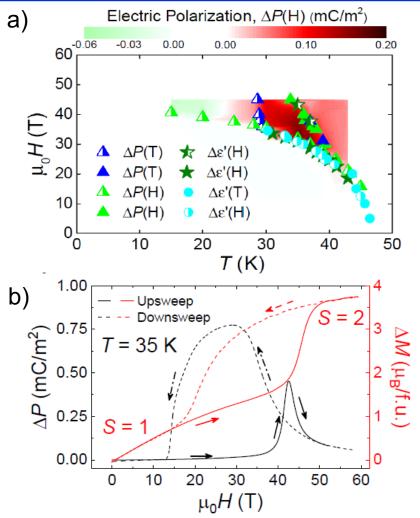


Figure: a) Phase diagram in DC fields of Mn(taa) in which the color scale is the electric polarization. b) Electric polarization ΔP and magnetization ΔM change in pulsed fields showing the spin crossover and region of electric polarization.