Multiferroic behavior at a spin state transition

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To date, multiferroic research has focused on conventional magnetic order such as ferromagnetism and antiferromagnetism, which is created by magnetic exchange interactions when atomic orbitals overlap each other. However, spin state transitions could provide an attractive alternative route to multiferroic behavior due to the large involvement of the lattice. With spin state transitions in inorganic-organic hybrid materials showing lattice changes several orders of magnitude larger than inorganic materials, they should be able to create significant magnetic-ferroelectric coupling in the right materials. Here, we demonstrate that a spin state transition can indeed lead to multiferroic behavior. We show that in the molecular magnet Mn(taa) (H_3taa=tris(1-2-azoyl)-2-azabuten-4-yl)amine) a magnetic field-induced spin-state transition toggles the existence of electric polarization at a first-order hysteretic magnetoelectric, magnetoelastic phase transition. [1] The high spin state is Jahn-Teller active, with the Jahn-Teller distortion creating an electric dipole. In high magnetic fields these dipoles become static instead of dynamic and freeze into a polar space group. We present the phase diagram of multiferroic behavior in Mn(taa), discuss underlying physics and present an accompanying mean field theory.