Field-induced Ferroelectricity in an Organic Quantum Magnet

V. S. Zapf, F. Balakirev (NHMFL, LANL); F. Fabris, M. Kenzelmann (Laboratory for Solid State Physics, ETH Hönggerberg, Zurich); Y. Chen, C. Broholm (Johns Hopkins U., Physics)

Introduction
Multiferroic behavior is generally rare and has never been observed before in an organic quantum magnet. The compound CDC (CuCl2 •2(CH3)2SO) however exhibits the necessary symmetry for certain applied magnetic fields H to allow an electric polarization P to coexist with antiferromagnetic (AFM) order. This compound contains chains of Cu S=1/2 spins [1] that are weakly coupled via superexchange and exhibit AFM order below TN = 0.93 K, observed in specific heat and neutron diffraction measurements [2]. For H applied along the crystallographic a- and c-axes, the AFM order is suppressed, most probably due to a competition between interchain interactions and field-induced staggered fields [2]. For H along the orthorhombic c-axis, AFM order is suppressed by H ~ 4 T (see Fig 1). A spin-flop transition above Hsf = 0.35 T leads to a magnetically ordered state (AFM B) that breaks inversion symmetry along the b-axis for 0.35 T < H < 4 T.

Experimental
We have measured the pyroelectric current of single crystals of CDC using an electrometer for H || c with the polarization P || b down to 3He temperatures in the 50 T short pulse magnet at NHMFL-LANL. The rapid rise time of this magnet was necessary to increase the signal to noise of the pyroelectric current in this sample.

Results and Conclusion
Our pyroelectric measurements (Fig 2) indicate that ferroelectricity occurs in the same region of H-T space as the AFM B phase between 0.35 T < H < 4 T and below T = 0.93 K. Hysteresis is observed in the polarization depending on the direction of the previous two field sweeps. Interestingly, we find the same results with and without electrically poling the sample. The spin polarization calculated from the pyroelectric current closely tracks the magnetic order parameter. While the magnetically-induced ferroelectricity in CDC is far from practical temperatures and fields, it nevertheless demonstrates that this phenomenon can occur in a whole new class of compounds.

Fig 1: Phase diagram of CDC showing regions of AFM with inversion symmetry (AFM A), AFM without inversion symmetry (AFM B) ferroelectric polarization, and paramagnetism (PM). Neutron and specific heat data is from Refs. 3,4.

Fig 2: pyroelectric current measured on the upsweep of a 50 T short pulse magnet. Consecutive pulses A – F are shown, where the sign of P depends on whether the field sweep is in the same or opposite direction as the previous sweep.

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References