

Observation of multiferroicity in the Mott insulator κ -(ET)₂Cu[N(CN)₂]Cl

M. Lang, J. Müller, B. Hartmann, R. Rommel, M. de Souza ¹,
P. Lunkenheimer, S. Krohns, A. Loidl,²
and J. A. Schlueter³

¹*Institute of Physics, Goethe-University Frankfurt, Germany*

²*Experimental Physics V, University of Augsburg, Germany*

³*Materials Science Division, Argonne National Laboratory, Argonne, Illinois, USA*

Email: milang@physik.uni-frankfurt.de

The organic charge-transfer salt κ -(ET)₂Cu[N(CN)₂]Cl has a Mott-insulating ground state where localized spins, residing on a triangular lattice of ET dimers, give rise to antiferromagnetic (afm) order below $T_N \approx 27$ K followed by a weak ferromagnetic canting around 23 K [1]. Very recently, by means of a comprehensive dielectric study, we have found that the system not only exhibits magnetic order but also undergoes a ferroelectric transition at T_{FE} , making this material the first multiferroic charge-transfer salt [2]. Our proof of ferroelectricity is based on (i) a peak in the dielectric constant $\epsilon'(T)$, reaching values of several hundred, (ii) the switching of the macroscopic polarization $P(E)$ of the sample probed by measurements of the current response through so-called positive-up-negative-down (PUND) sweeps of the electric field E , and (iii) measurements of the polarization-electric field hysteresis curve. Most remarkably, the measurements reveal $T_{FE} \approx T_N$, suggesting a close interrelation between both types of ferroic order. From studies in a magnetic field of 9 T, which leaves the ferroelectric transition unaffected but alters the spin structure significantly, a standard spin-driven ferroelectric order can be ruled out. Here we summarize our results of dielectric measurements, including studies on four crystals with different geometries and contact materials, and discuss a possible charge-order driven mechanism to account for the observations.

[1] K. Miyagawa *et al.*, Phys. Rev. Lett. **75**, 1174 (1995).

[2] P. Lunkenheimer *et al.*, Nature Mater. **11**, 755 (2012).