Observation of multiferroicity in the Mott insulator $\kappa$-(ET)$_2$Cu[N(CN)$_2$]Cl

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The organic charge-transfer salt $\kappa$-(ET)$_2$Cu[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.