Project B6

1) Universal properties of the Mott transition

Organic charge-transfer salts of the κ-(BEDT-TTF)$_2$X family offer exciting possibilities to directly access the Mott metal-insulator transition and to study its universal properties. A question of particular interest is whether the Mott transition belongs to a conventional, Ising-type universality class, or whether the distinct 2D character of these materials implies a novel, unconventional criticality, as proposed by transport measurements [1]. Based on our previous thermal expansion measurements on deuterated κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br single crystals [2], and a scaling analysis of the data [3], we found that the thermodynamic results are in perfect agreement with a 2D Ising universality class. In order to derive a more complete picture, we are presently performing thermal expansion measurements under variable He-gas pressure - an experiment now possible in our laboratory [4] – in order to tune the material in the vicinity of the Mott critical end point and to compare the results with theoretical predictions [4,5].

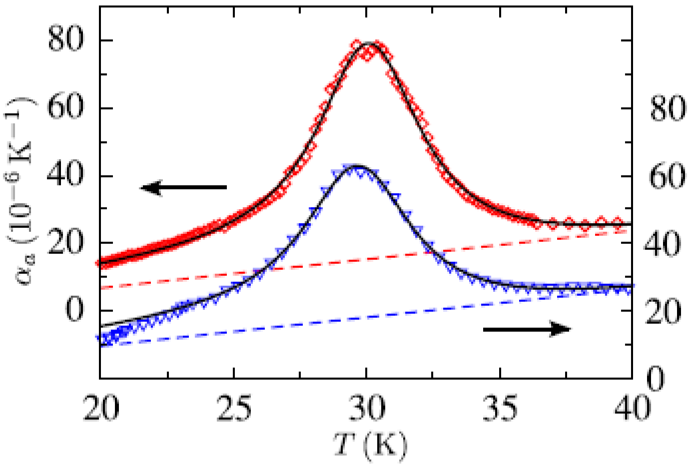
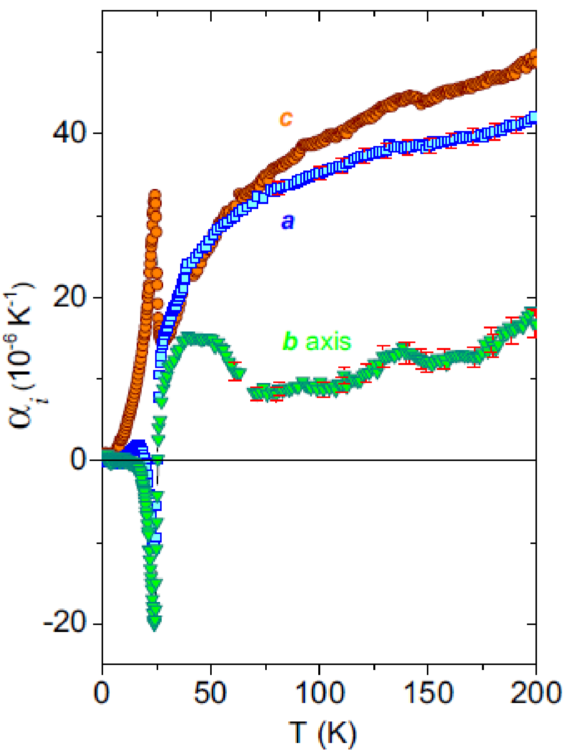


Fig. 1: Thermal expansion data fort two crystals of deuterated κ-(ET-TTF)$_2$Cu[N(CN)$_2$]Br; solid lines represent fits based on a 2D Ising universality class.

[1] F. Kagawa *et al.*, Nature **436**, 534 (2005).
[2] M. De Souza *et al.*, Phys. Rev. Lett. **99**, 0370031 (2007).
[3] L. Bartosch *et al.*, Phys. Rev. Lett. **104**, 245701 (2010).
[4] R. S. Manna *et al.*, Rev. Sci. Instr. **83**, 085111 (2012).
[5] M. Zacharias *et al.*, Phys. Rev. Lett. **109**, 176401 (2012).

2) Lattice effects in the 2D valence-bond-solid Mott insulator EtMe$_3$P[Pd(dmit)$_2$]

The title material belongs to the series of organic charge-transfer salts Et$_x$Me$_{4-x}$Z[Pd(dmit)$_2$]$_2$, where interesting ground states such as a spin liquid (Z = Sb, x = 1) or a valence-bond-solid phase (Z = P, x = 1) can be studied. In these compounds strongly dimerized [Pd(dmit)$_2$]$_2^-$ anions, carrying S = ½ spins, form a 2D, slightly distorted triangular lattice. We have studied the lattice effects around the phase transition at T_{VBS} = 25 K into the low-temperature valence-bond-solid phase and the paramagnetic regime above where effects of short range antiferromagnetic correlations are expected. The salient results of our study [1] include (i) the observation of strongly anisotropic lattice distortions at T_{VBS}, with an unexpectedly large out-



of-plane effect, and (ii) a distinct maximum in the expansion coefficient around 40 K. We argue that the latter feature together with a broad maximum in the magnetic susceptibility around 70 K, speaks in favour of a more anisotropic triangular-lattice scenario for this compound than previously thought [1].

[1] R. S. Manna et al., Phys. Rev. B **89**, 045113 (2014)

Fig. 2: Temperature dependence of the uniaxial expansivities α_i of $EtMe_3P[Pd(dmit)_2]_2$ measured along the in-plane *i* = *a* and *c* axis and along the out-of-plane *b* axis.

3) Multiferroicity in κ-$(BEDT\text{-}TTF)_2Cu[N(CN)_2]Cl$

The organic charge-transfer salt κ-$(BEDT\text{-}TTF)_2Cu[N(CN)_2]Cl$ has a Mott-insulating ground state where localized spins S = 1/2, residing on a triangular lattice of BEDT-TTF dimers, order antiferromagnetically below T_N ~ 27 K [1]. In a recent comprehensive study of the material's dielectric properties in collaboration with the Augsburg group (P. Lunkenheimer et al.) we 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]. Surprisingly, we found that $T_{FE} \approx T_N$, which suggests a close interrelation between both types of ferroic order.

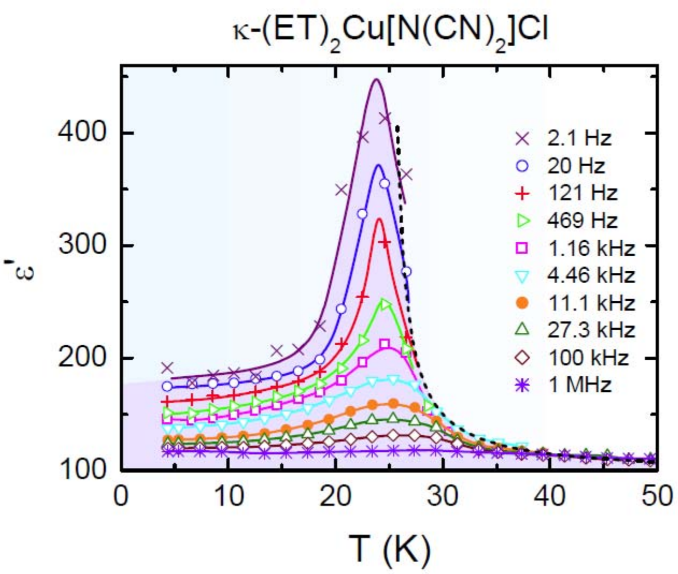


Fig. 3: The anomaly in the dielectric constant ε' measured at different frequencies is indicative of an order-disorder-type ferroelectric transition.

From studies in a magnetic field of 9 T, which leaves the ferroelectric transition unaffected but alters, via a spin-flop transition, the spin structure significantly, a standard spin-driven ferroelectric order can be ruled out. In a recent work [3] we study sample-to-sample variations by performing both magnetic and dielectric measurements on the same single crystal: whereas some crystals (three out of eight crystals studied) show relaxor-type anomalies, indicative of short-range ferroelectric correlations, others (five out of eight) reveal clear indications for an order-disorder type transition to long-range ferroelectric order.

[1] K. Miyagawa *et al.*, Phys. Rev. Lett. **75**, 1174 (1995)
[2] P. Lunkenheimer *et al.*, Nature Mater. **11**, 755 (2012)
[3] M. Lang *et al.*, arXiv: 1311.2715 (accepted for publication).

4) Investigations of the spin liquid candidate system κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

The title compound is a half-filled Mott insulator, where ET dimers form a quasi-2D triangular lattice with hopping amplitudes t'/t ≈ 0.8. Due to the strong frustration the material lacks long range order down to mK temperatures and is thus considered a promising candidate for a spin liquid ground state. In a previous thermal expansion and specific heat study on single crystalline material we have demonstrated that the mysterious 6 K anomaly - a potential instability of the spin liquid – manifests a second order phase transition. By comparison with various spin models we found that the spin entropy alone is not sufficient to account for the entropy release, indicting an important role of charge degrees of freedom in this instability. In a more recent work we have shown that there is a striking effect of a magnetic field on the low-temperature state: While the 6 K anomaly is found to be insensitive to magnetic fields $B \leq 10$ T, the maximum field applied, surprisingly strong B-induced effects are observed for magnetic fields applied along the in-plane b-axis. Above a threshold field of 0.5 T < Bc ≤ 1 T, a jump-like anomaly is observed in the b-axis lattice parameter. This anomaly, which is located at 8.7 K at B = 1 T, grows in size and shifts to lower temperatures with increasing the magnetic field. Although the anomaly bears resemblance to a first-order phase transition, the lack of hysteresis suggests otherwise.

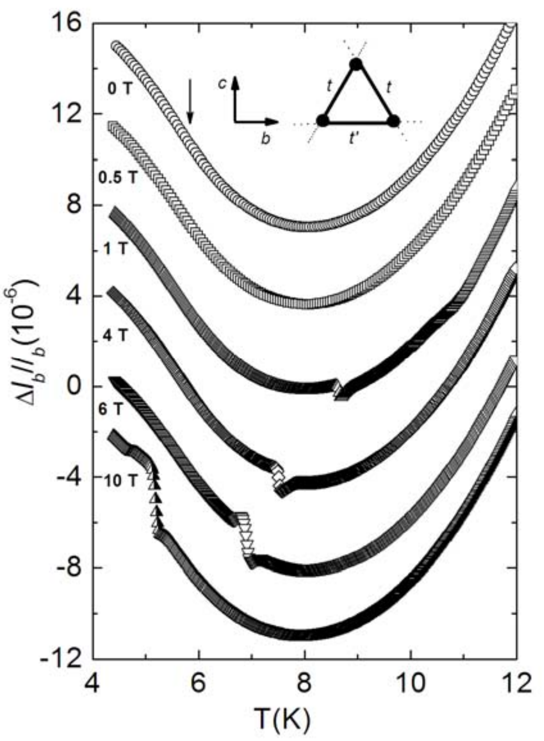


Fig. 4: Relative length changes of single crystalline κ-(BEDTTTF)$_2$Cu$_2$(CN)$_3$ measured along the in-plane b-axis as a function of temperature for varying magnetic fields $B || b$.

[1] R. S. Manna *et al.*, Phys. Rev. Lett., 104, 016403 (2010).
[2] R. S. Manna *et al.*, Phys. Stat. Solidi C9, 1180 (2012).