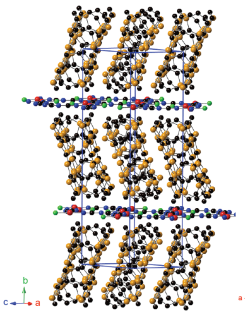
B11 Low-frequency electron dynamics of organic charge-transfer salts studied by fluctuation spectroscopy

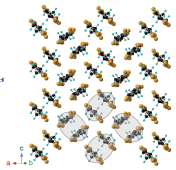
Jens Müller (Goethe University Frankfurt)

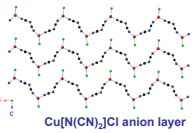
Introduction

Quasi-2D molecular metals κ-(BEDT-TTF)$_2$X



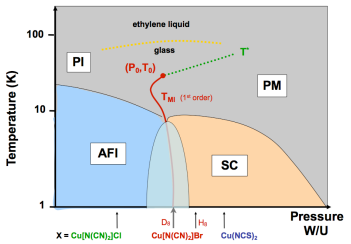
- strong dimerization: half-filled band
- $W \sim U_{eff}$: correlated π-carriers
- triangular lattice
- strong electron-phonon coupling





Generic phase diagram

- fine-tune electronic correlations by *p*, *X* and *q*
- access Mott criticality

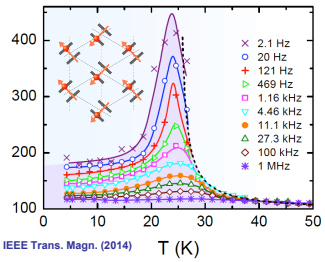


B2 B6 B8 B12

Achievements

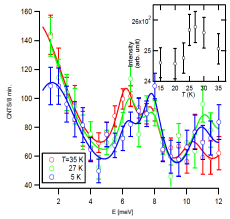
First multiferroic charge-transfer salt κ-(ET)$_2$Cu[N(CN)$_2$]Cl

- peak in dielectric constant characteristic for order-disorder-type ferroelectrics
- T_{FE} coincides with antiferro-magnetic order at $T_N \approx 27$ K
- sample-to-sample variation (relaxor vs. long range orde



Lunkenheimer et al., Nature Materials (2012), Lang et al., IEEE Trans. Magn. (2014)

→ electric-dipole driven magnetism ?! B6 U Augsburg



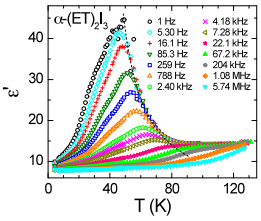
- spin-charge-lattice coupling?
  → first inelastic neutron results on 7 mg samples at ILL
- hardening of lowest-lying optical mode close to T_{FE}/T_N

Matsuura et al., unpublished

→ further studies in preparation

- cf. weakly-dimerized α-(BEDT-TTF)$_2$I$_3$
  → relaxor-ferroelectricity deep in the charge-ordered state
  → polar and non-polar stacks of organic molecules

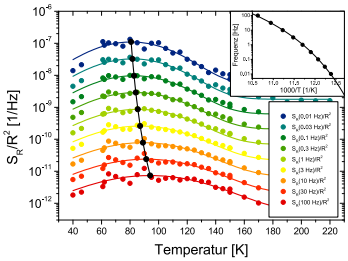
Lunkenheimer et al., arXiv:1407.0339 (2014)



→ study carrier dynamics in FE (charge-ordered) state

Achievements

Glass-like transition in κ-(BEDT-TTF)$_2$X



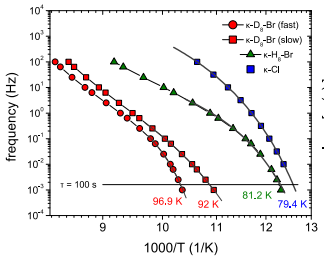
- 100 K maximum in low-*f* fluctuations ubiquitous in κ-(ET)$_2$X

→ importance of polymeric anion chains

→ Vogel-Fulcher-Tamman behavior

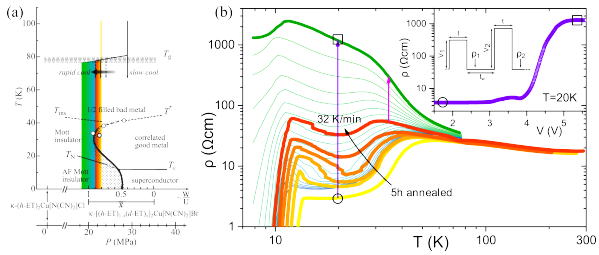
$$f = f_0 \exp\left(-\frac{D \cdot T_{VFT}}{T - T_{VFT}}\right)$$

- strength parameter *D* found to be 0.6 – 6.6 < 10 categorizes κ-(ET)$_2$X as fragile glass-formers
- fluctuation spectroscopy reveals typical glassy properties of α-relaxation (dieletric spectroscopy not applicable)



Rommel et al., to be submitted

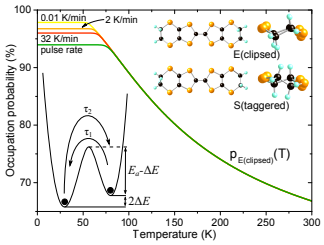
Utilizing the glasslike ordering to induce a Mott metal-insulator transition



- heat pulse (sample thermally coupled to low-temperature bath) induces Mott MIT → realize cooling rates of ~1000 K/min

→ fine-tune bandwidth *W*/*U* (persistent below T_g and reverible)

- simple model allows for:
  → determining amount of 'disordered' molecules (only 6% for 1000K/min)
  – estimate energy difference between ethylene endgroup confirmations: ΔE ~ 100 K (E_a ~ 2500 K)



Hartmann et al., PRB (2014)

- theoretical calculations in progress

B2

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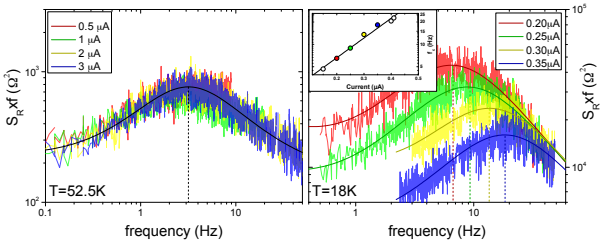
Project goals and work program

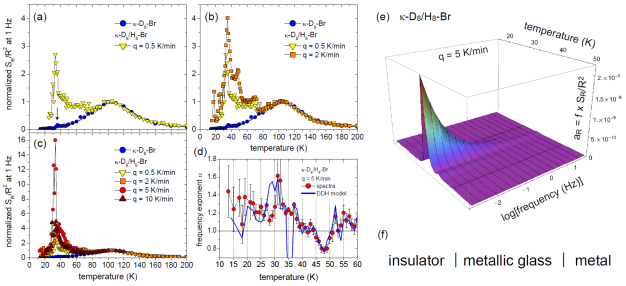
Multiferroicity in κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl



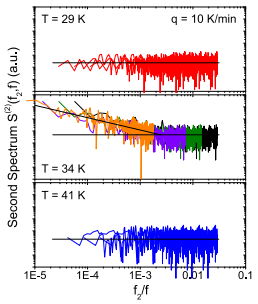
- 1/*f* noise resolved into indiviudual Lorentzian
  → single enhanced process/fluctuator
- cluster/domains switching with characteristic frequency
- determine volume from $f_c(E)$ → nanoscale cluster size

→ Ferroelectric-Domain Wall Dynamics in TTF-CA **B9**

Dynamic Criticality at the Finite-Temperature Mott Transition



- κ-(d$_8$-ET)$_x$(h$_8$-ET)$_{1-x}$Cu[N(CN)$_2$]Br (close to Mott critical region)
- correlation-induced increase of the low-frequency fluctuations



- *q* = 5 K/min: divergent fluctuations & strong shift of spectral weight to low frequencies
  → **critical slowing down**
- *q* = 10 K/min: noise peak decreases
  → rules out disorder effect
- deviation of α(*T*) from phenomenological model of non-exponential kinetics (DDH) in the vicinity of T_{cr} & correlations in second spectrum

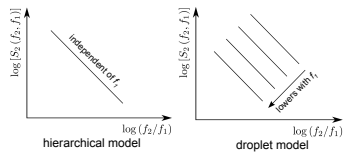
→ onset of non-Gaussian fluctuations

→ **possible glassy freezing of the electron system**

- spin glasses: hierarchical/droplet model for $S^{(2)}$

→ spectral wandering indicates correlated fluctuations
→ many metastable states connected by kinetic hierarchy

see Weissman et al., JMMM (1992)

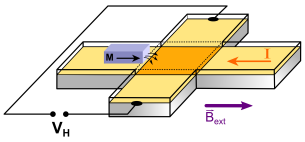


Techniques

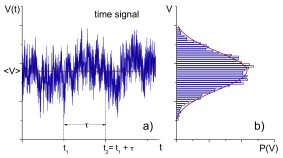
- growth of organic charge-transfer salts by electrochemical crystallization
- **micro-Hall magnetometry** **B6**

  → magnetic field-induced effects related to '6K anomaly' in spin-liquid candidate κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

  Manna et al., Phys. Stat. Sol. C (2012)



- electrical (magneto-)resistivity and Hall effect
- time-resolved el. transport: **fluctuation (noise) spectroscopy**



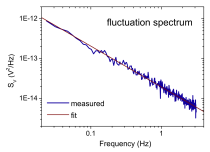
- a) time signal with b) Gaussian probability distribution and c) 1/*f*-type frequency spectrum

$$S_V(\omega) \equiv \langle |\partial \tilde{V}(\omega)|^2 \rangle$$

$$\propto \int \psi(\tau)\cos(\omega\tau)d\tau \propto \frac{\chi''(\omega)}{\omega}$$

- access correlation function:

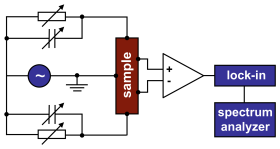
$$\psi(\tau) = \langle \partial V(t) \cdot \partial V(t+\tau) \rangle$$

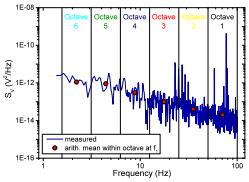


- evaluate higher-order correlation functions by second spectrum

$S_V^{(2)}(\omega_2,\omega)$, i.e. power spectrum of fluctuations of $S_V(\omega)$

- new versatility with self-written python software for control and analysis
  → measure second spectrum
  → automatized noise measurements (!)
  → DC/current noise for high-*R* samples

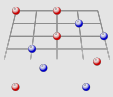




see review article:
J. Müller, ChemPhysChem (2011)

Role within the SFB/TR 49

B6	• critical behavior at finite-temperature Mott transition • noise spectroscopy under ^{4}He-gas pressure • electrocrystallization of single crystals • dielectric spectroscopy (with P. Lunkenheimer, Augsburg)
B2	• band structure and DFT calculations
B9	• noise spectroscopy on TTF-CA thin films w/ Pt(C) interface
B12	• local density of states: low-temperature STM/STS
B8	• charge-order/ferroelectric domains: momentum microscopy



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