B6 Collective phenomena in organic charge-transfer salts close to the Mott transition

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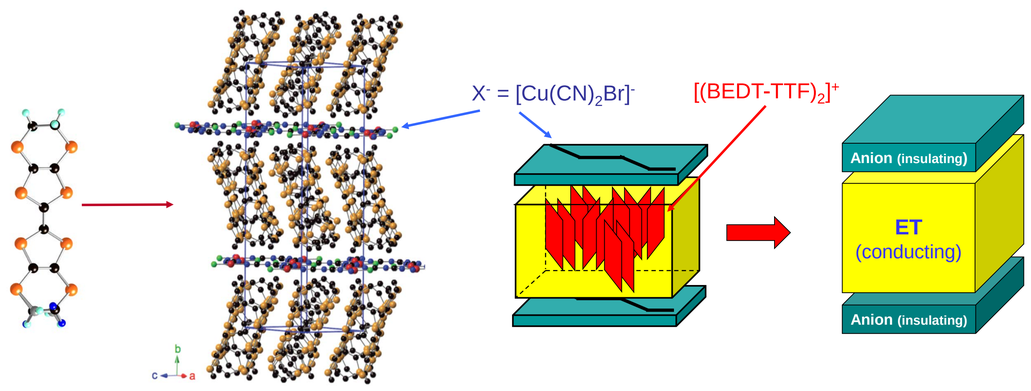
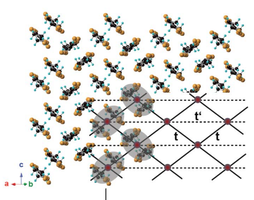
Introduction

κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]X, X = Br, Cl

Quasi-2D electronic structure





Top View
BEDT-TTF layer

1 hole/dimer: half-filled conduction band

• Narrow bands: $W \approx U_{eff} \sim 0.5$ eV ⇒ correlated π-electron system

• Frustrating magnetic dimer-dimer interactions $t'/t \sim 0.7$ -1, depending on X
cf. Kandpal *et al.*, PRL **103**, 067007 (09)

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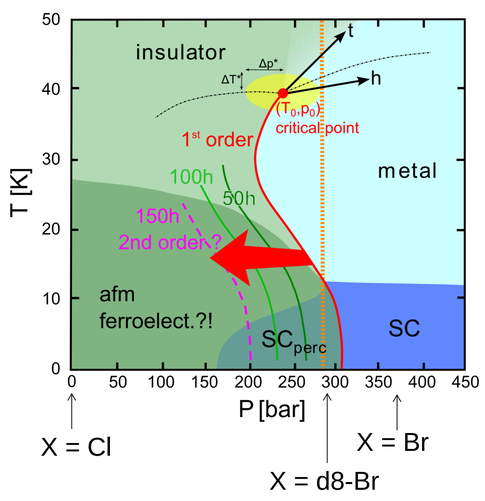
• afm ordered Mott insulator
• superconductivity under pressure
• S-shaped Mott transition line $T_{MI}(P)$

Cf. Lefebvre *et al.*, PRL **85**, 5420 (00)
Limelette *et al.*, PRL **91**, 016401 (03)
Fournier *et al.*, PRL **90**, 127002 (03)
Kagawa *et al.*, PRB **69**, 064511 (04)
S. Köhler, PhD Thesis (in preparation)



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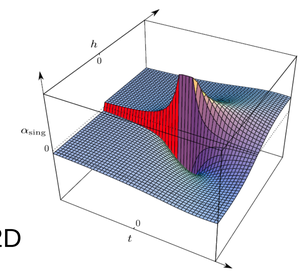
Mott transition and criticality

a) Achievements

Previous results on κ-(d8-BEDT-TTF)$_2$Cu[N(CN)$_2$]Br,

Cf. M. de Souza *et al.*, PRL 101, 216403 (08)
L. Bartosch *et al.*, PRL 104, 016403 (10)



- Large lattice effects arising from critial behaviour
- Application of a scaling Ansatz: Results consistent with 2D Ising universality class
- Sign change for $\alpha(T,P)$ at (P_0,T_0) predicted by scaling Ansatz!

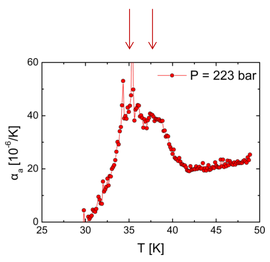
$\alpha(T,P)$ provides a most sensitive thermodynamic tool for probing Mott criticality!

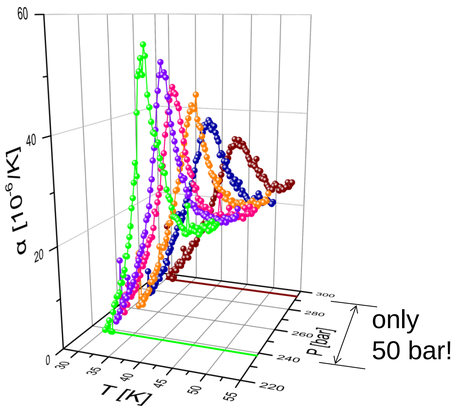
Important new developments

***P*-dependent thermodynamic study of Mott criticality!**
Use of novel technique: thermal expansion under pressure $\alpha(T,P)$

κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl:
$P > P_0$ metallic regime:

Within 50 bar, α anomaly changes drastically
in size by a factor of 3!
in position by 5 K!





Closer to P_0: occurence of double-peak structure, interference of another phase transition (intrinsic) or bicrystal (extrinsic)?

b) Project goals and work programme

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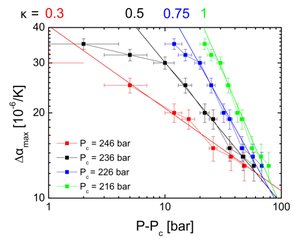
1) Solve controversy on criticality

2D Ising vs. "unconventional" criticality

cf. Kagawa *et al.*, Nature **436**, 534 (05)

- Prediction by scaling ansatz: $\alpha_{max} \propto (P\text{-}P_0)^{\kappa}$, $\kappa = \frac{1-\beta}{\beta+\gamma}$

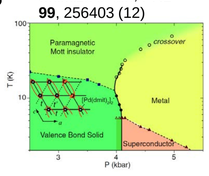


- Preliminary results incompatible with "unconventional" criticality (κ = 0)!
- More detailed knowledge about P_0 required: check higher-quality single crystals

2) Effect of electron-phonon-coupling on critical behaviour:

- Crossover from 2D Ising (κ ≈ 0.5) to mean-field (κ ≈ 0.3) criticality?
cf. Zacharias *et al.*, PRL **109**, 097206 (12)

Shimizu *et al.*, PRL **99**, 256403 (12)



3) Critical behaviour of $EtMe_3P[Pd(dmit)_2]_2$

- Nonmagnetic VBS ground state: stronger coupling to lattice expected

Charge degrees of freedom close to the Mott transition

a) Achievements

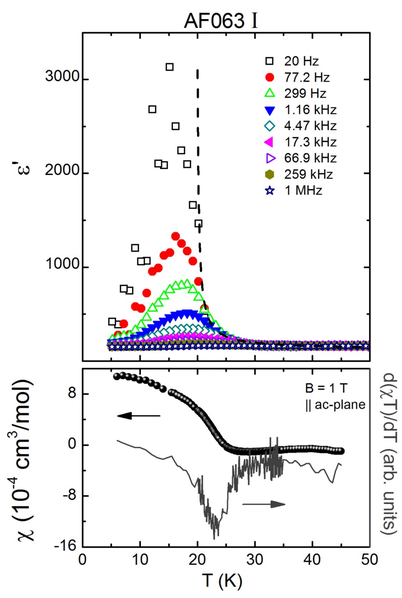
Discovery of first multiferroic charge-transfer salt κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl

P. Lunkenheimer, J. Müller, … , M. Lang, Nature Materials **11**, 755 (12)
M. Lang *et al.*, IEEE Transactions on Magnetics **6**, 2700107 (14)

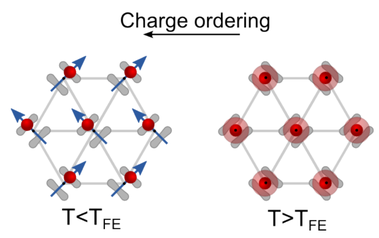
P. Lunkenheimer, Augsburg B11

Important new insights:

- Intra-dimer degrees of freedom active!
- Order-disorder type ferroelectric (5 out of 8 crystals)
- $T_N \approx T_{FE}$ suggests close interrelation
- Studies at finite *B*: no spin-driven mechanism



Proposed mechanism: afm driven by charge order!



- amount of charge disproportionation? cf. Sedlmeier *et al.*, PRB **86**, 245103 (13)
- Consistent with relaxor-type ferroelectricity in α-(BEDT-TTF)$_2$I$_3$

P. Lunkenheimer *et al.*, arXiv: 1407.0339

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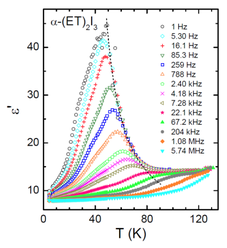


b) Project goals and work programme

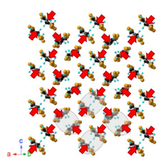
1) Study the interrelation between T_N and T_{FE} under pressure

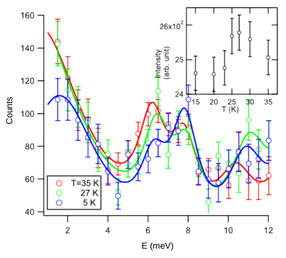
- ε'(*T*, *P*) in combination with χ(*T*, *P*)
- Dielectric measurements in preparation

2) Study of the coupling between spin, charge and lattice degrees of freedom

- Inelastic neutron scattering

International collaboration: Japan, Germany, France B11

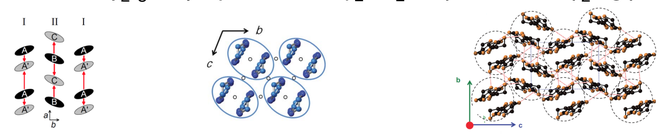




3) Effects due to magnetic order vs. effects due to charge order

- Comparative study of several systems with different dimer topology

α-(BEDT-TTF)$_2$I$_3$ β'-(BEDT-TTF)$_2$ICl$_2$ κ-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl



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