B2

Ab initio modelling and design of correlated materials

Roser Valentí (Universität Frankfurt)

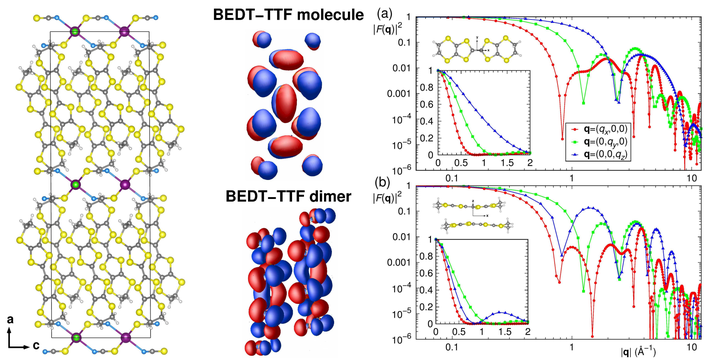
Charge-transfer salts: Achievements

Magnetic form factors for charge-transfer salts

- Magnetic cross section in inelastic neutron scattering:

$$\frac{d^2\sigma}{d\Omega d\omega} = (\gamma r_0)^2 \frac{k'}{k} |F(\mathbf{q})|^2 e^{-2W(\mathbf{q})} \sum_{\alpha\beta} \left(\delta_{\alpha\beta} - \frac{q_\alpha q_\beta}{q^2}\right) S^{\alpha\beta}(\mathbf{q},\omega) \quad F(\mathbf{q}) = \int \mathrm{d}^3 r \; e^{i\mathbf{q}\cdot\mathbf{r}} \rho_s(\mathbf{r})$$

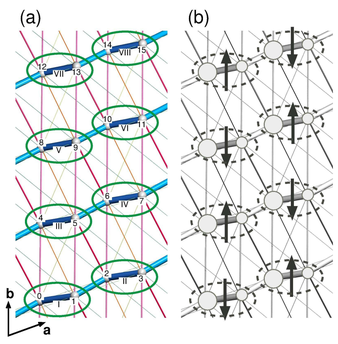
Form factor? ⇒ *ab initio* DFT + molecular Wannier projections

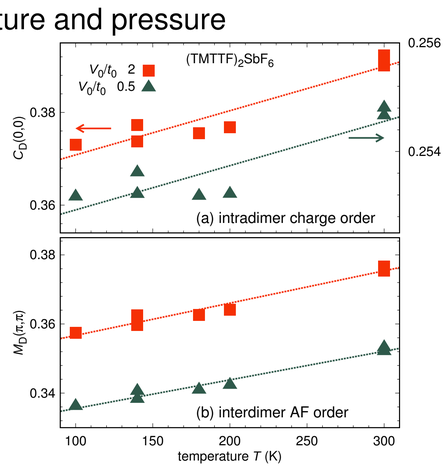


F. Salvat-Pujol, H.O. Jeschke, R. Valentí, Phys. Rev. B **90**, 041101 (2014)

Quasi-1D Fabre charge-transfer salts

ab initio DFT + extended Hubbard model → electronic properties as a function of temperature and pressure



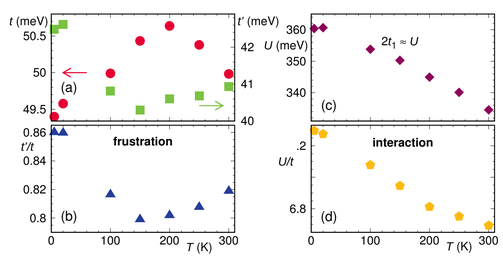


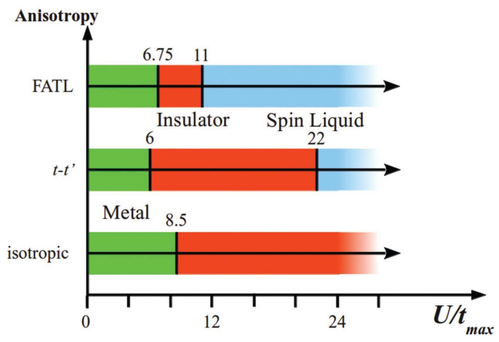
A.C. Jacko, H. Feldner, E. Rose, F. Lissner, M. Dressel, R. Valentí and H.O. Jeschke, PRB **87**, 155139 (2013)

2D triangular spin-liquid candidates

ab initio-derived Hubbard parameters

κ- $(BEDT\text{-}TTF)_2Cu_2(CN)_3$: **B6** $Me_3EtSb[Pd(dmit)_2]_2$: **B7** DFT+VMC temperature dependence

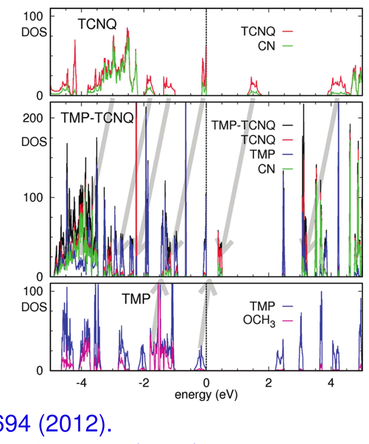




A.C. Jacko, L.F. Tocchio, H.O. Jeschke and R. Valentí, PRB **88**, 155139 (2013) H.O. Jeschke, M. de Souza, R. Valentí, R.S. Manna, M. Lang, J.A. Schlueter, PRB **85**, 035125 (2012) L.F. Tocchio, H. Feldner, F. Becca, R. Valentí, C. Gros, PRB **87**, 035143 (2013)

New donor-acceptor system TMP/HMP-TCNQ/F_4TCNQ

- Synthesis in Project **B10** **B9**
- Nature of charge transfer: combined NEXAFS **B8** **B12** and density functional theory calculations.



K. Medjanik *et al.* JACS **134**, 4694 (2012).
M. Rudloff *et al.* Phys. Chem. Chem. Phys. **17**, 4118 (2015).

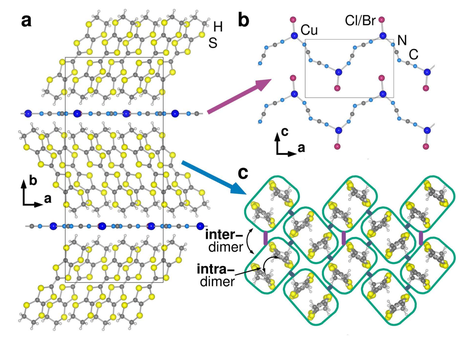
Project goals and program

Electronic effects beyond the effective dimer triangular lattice model

Multiferroic behavior in AFM κ-$(BEDT\text{-}TTF)_2Cu[N(CN)_2]Cl$ **B6** **B11**

- magnetism ↔ ferroelectricity ↔ intradimer charge degrees of freedom?

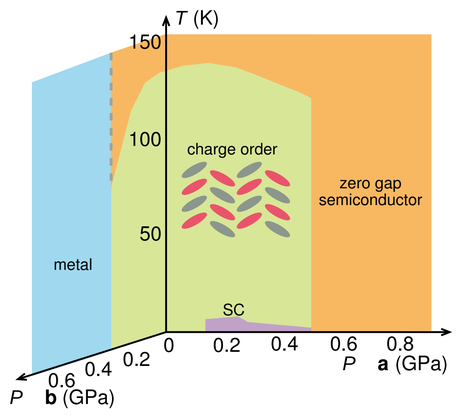
→ *ab initio* DFT investigation + effective models
→ dielectric response



α $(ET)_2I_3$ under strain

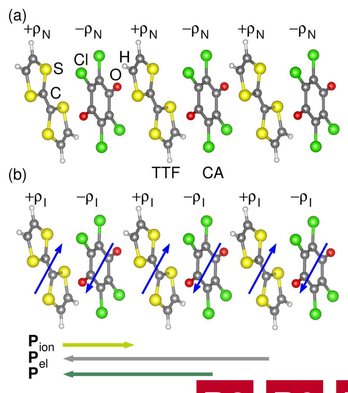
- Phase diagram: simulation of pressure/strain from *ab initio*
- Evaluation of the *ab-initio* derived extended Hubbard model:
  - Mean-field theory, DMFT, DCA, VMC
- Behavior near the Dirac-metal region?

B6 **B9** **B11**



Neutral-Ionic transition in TTF-CA under strain

- Polarization opposite to ionic displacement → Ferroelectricity caused by charge transfer processes
- Polarization from *ab initio* via Wannier representation and Berry phase formalism
  $\mathbf{P} = \mathbf{P_{ion}} + \mathbf{P_{el}} =$
  $-\frac{2ie}{(2\pi)^3} \sum_n \int_{BZ} \mathrm{d}^3k e^{-i\mathbf{kR}} < u_{n\mathbf{k}} | \nabla_{\mathbf{k}} | u_{n\mathbf{k}} >$
- Strain effects on charge transfer?

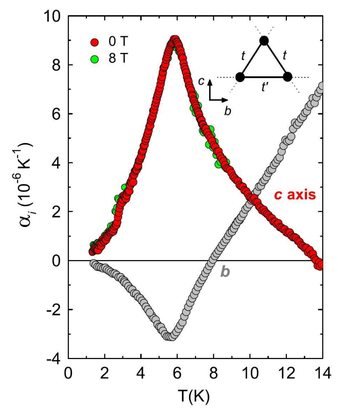


B8 **B9** **B11**

Electron-lattice couplings in spin-liquid candidates

Spin liquid candidates:

- $EtMe_3Sb[Pd(dmit)_2]_2$:
  - Anomalous uniaxial expansion
  - Strong electron-lattice coupling?
- κ-$(ET)_2Cu_2(CN)_3$:
  - Anomalous thermal expansion behavior at $T_c = 6$ K
  - Coupling of lattice, spin and charge degrees of freedom? → *ab initio* molecular dynamics simulations.

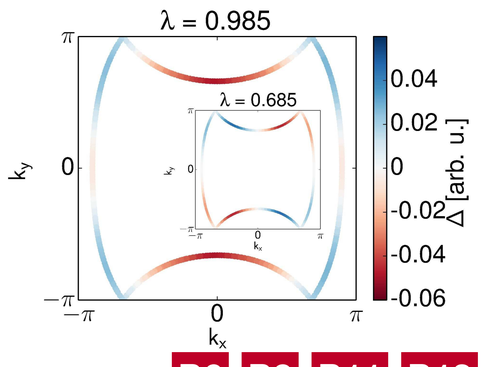


R.S. Manna, M. de Souza, A. Brühl, J.A. Schlueter, and M. Lang, PRL **104, 016403**

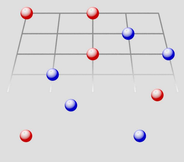
B6

Microscopic nature of superconductivity in organic systems

- Vicinity to Mott insulating state → Hint towards spin-fluctuation-mediated SC
- Role of frustration, dimerization?
- Dimer vs. molecule description
- DFT + spin fluctuation theory + VMC



B6 **B8** **B11** **B12**



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