Microscopic modelling of exotic properties in frustrated systems: triangular and kagome lattices

Project Report B2/B13, 9th Annual Retreat of SFB/TR49

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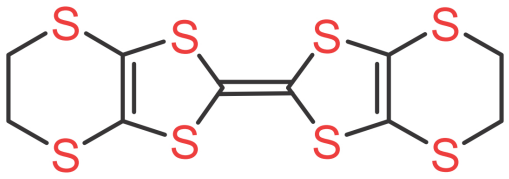
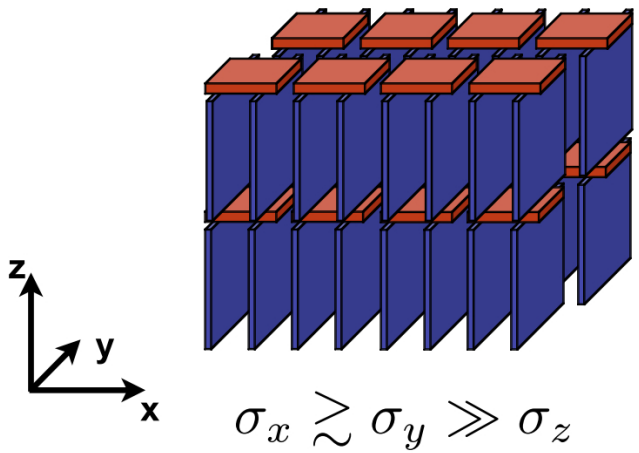
Organic charge transfer salts: Overview

B2

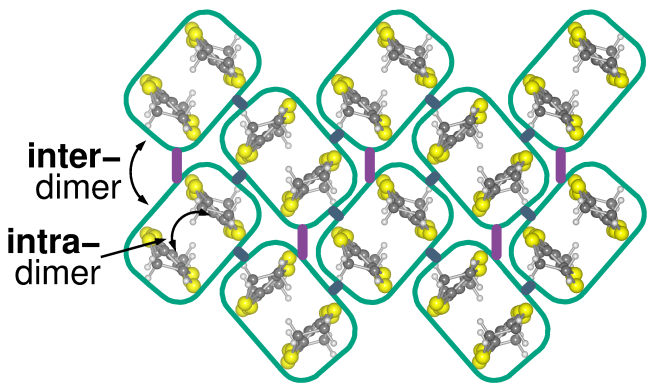
- ET = BEDT-TTF = bis(ethylene-dithio)-tetrathiafulvalene is the electron donor
- ET molecules can be packed in different patterns
- α'-phase charge-ordered insulator
- κ-phase often superconducting
- features $(ET)_2$ dimers that donate one electron to acceptor layer
- often modelled by 1/2-filled anisotropic triangular lattice of dimers
- alternative is 3/4-filled individual molecule model



ET (BEDT-TTF)



$$\sigma_x \gtrsim \sigma_y \gg \sigma_z$$



Figures from J. Müller, Chem. Phys. Chem. **12**, 1222 (2011) and Ferber *et al.*, PRB **89**, 205106 (2014)

Microscopic models for polymorphs of the superconductor $(ET)_2Ag(CF_3)_4(TCE)$

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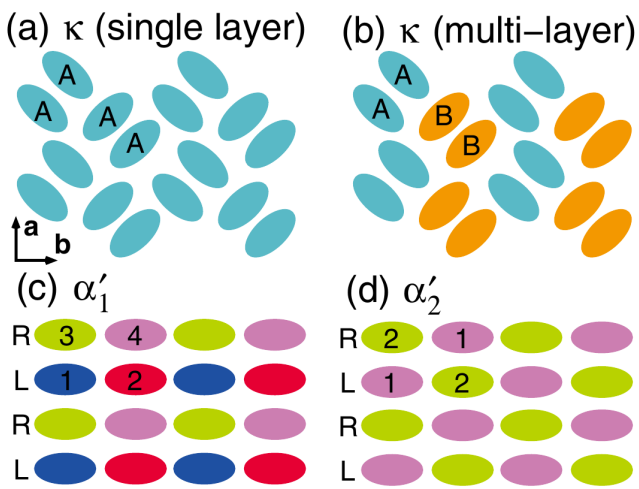
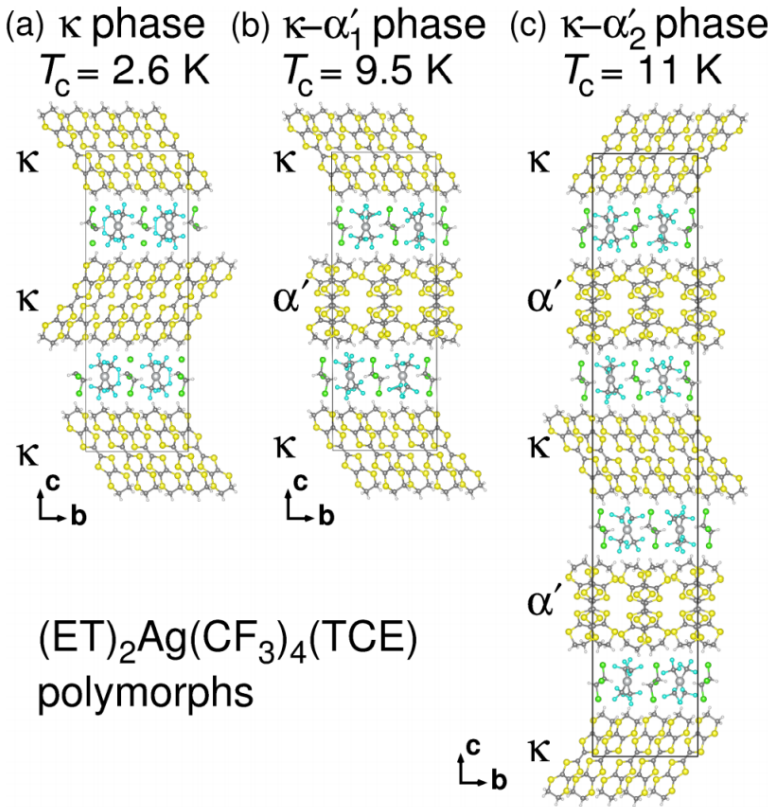
- Why does presence of α'-phase strongly enhance T_c ?
- microscopic hopping parameters from projective molecular Wannier functions
- charge ordering pattern in α'-layer influences electron hoppings in κ-layer
- degree of frustration significantly enhanced for higher-T_c compounds





Altmeyer, Valentí, Jeschke, PRB **91**, 245137 (2015)

Influence of molecular conformations on the electronic structure of organic charge transfer salts

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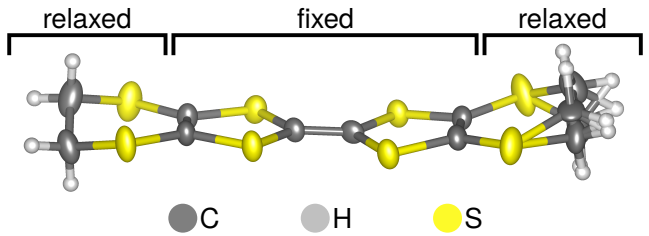
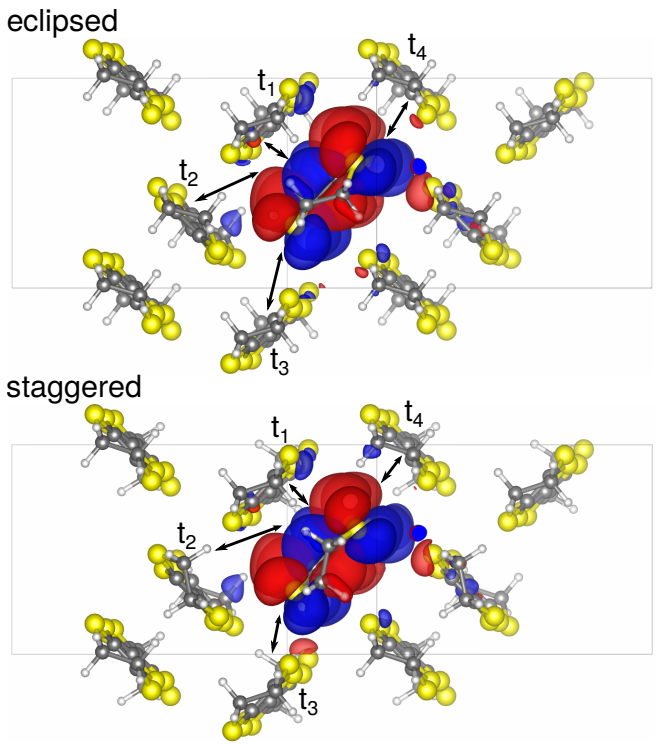
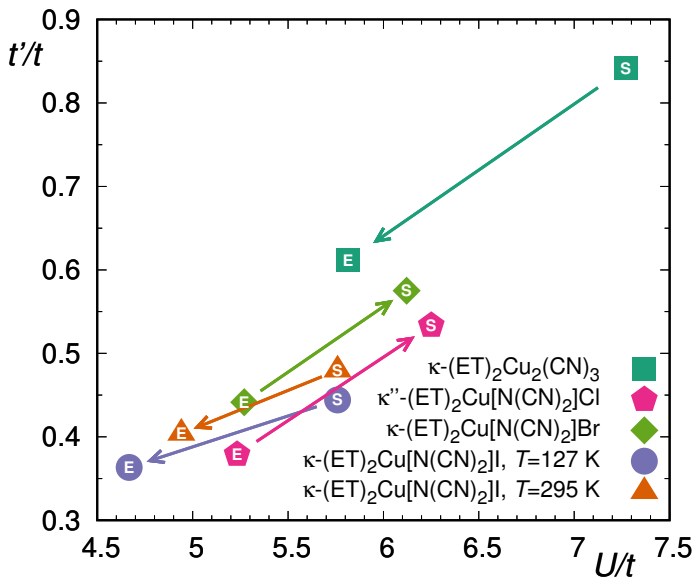
- ethylene endgroups in ET molecules can be in eclipsed or staggered configuration
- κ-$(ET)_2Cu[N(CN)_2]Br$ undergoes MIT with endgroup tuning (J. Müller *et al.*)
- endgroups influence hopping amplitudes and Hubbard repulsion
- staggered endgroups have larger t'/t, U/t







Guterding, Valentí, Jeschke, PRB **92**, 081109(R) (2015)

Unconventional superconductivity in realistic models for organic charge transfer salts

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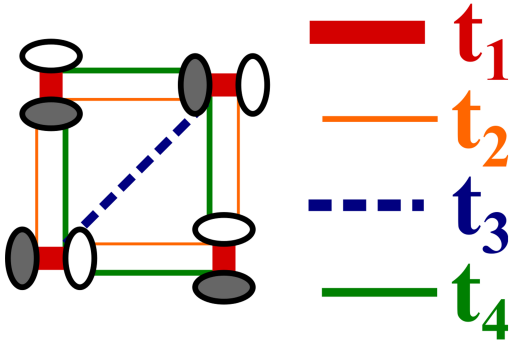
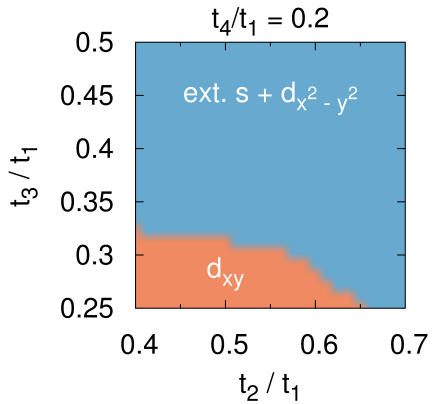
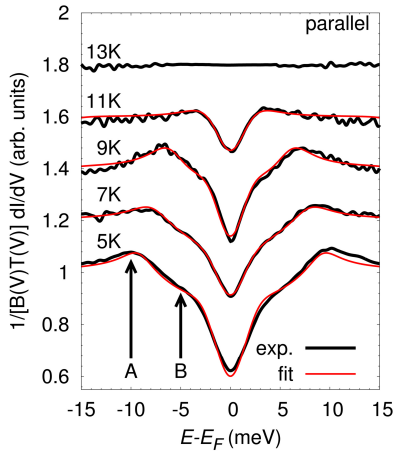
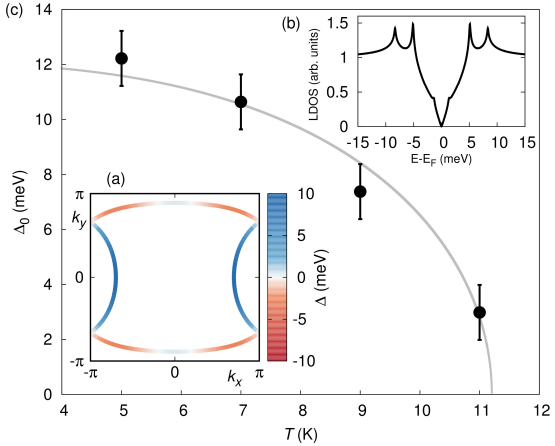
- superconductivity likely mediated by spin-fluctuations
- symmetry of the order-parameter from RPA for the Hubbard model
- dimer model gives d_{xy}
- molecule model either d_{xy} or ext. $s + d_{x^2-y^2}$
- STS from Elmers group shows two-peak structure in LDOS of superconducting κ-Br
- naturally explained by $s + d_{x^2-y^2}$ symmetry in molecule model









Guterding, Altmeyer, Jeschke, Valentí, in preparation

Diehl, Methfessel, Tutsch, Müller, Lang, Huth, Guterding, Altmeyer, Jeschke, Valentí, Jourdan, Elmers, in preparation

Prediction of a strongly correlated Dirac metal

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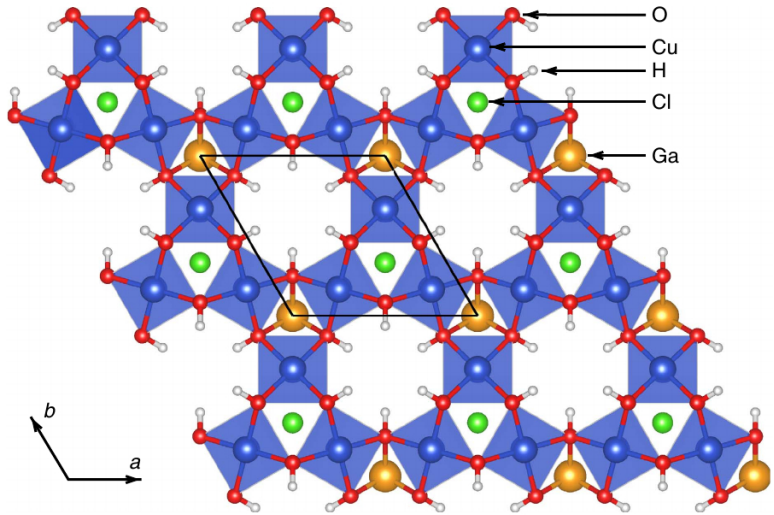
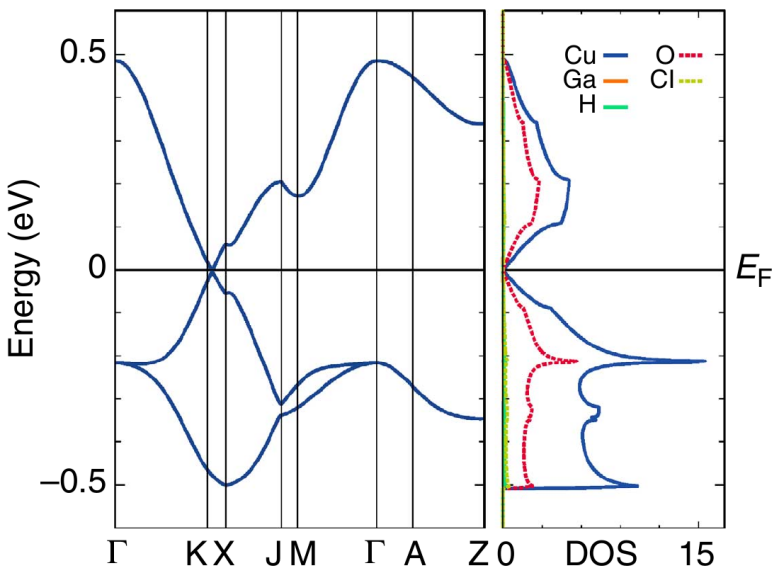
- Herbertsmithite $ZnCu_3(OH)_6Cl_2$ is a spin-liquid candidate
- replacing Zn^{2+} by Ga^{3+} shifts Fermi level to Dirac point
- Mott-transition suppressed by electron doping
- no charge-ordering present in DCA calculation
- competition between FM and f-wave SC predicted in vicinity of Dirac point
- material synthesis in Krellner group

Poster by Pascal Puphal, B4





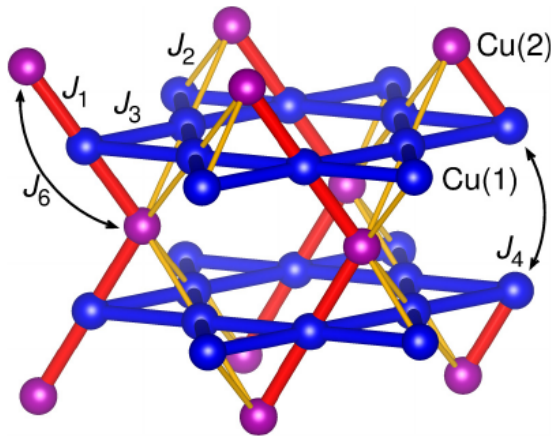
Jeschke, Salvat-Pujol, Valentí, PRB **88**, 075106 (2013)

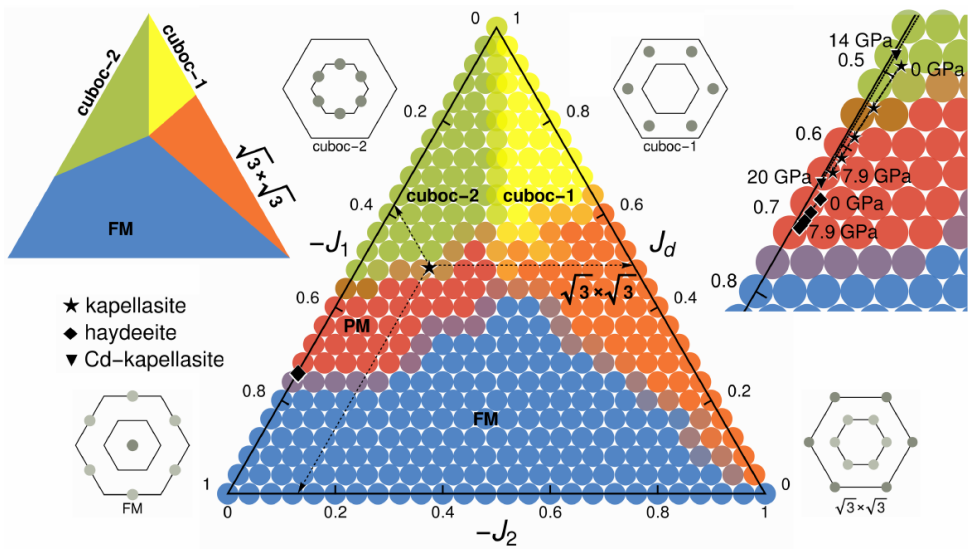
Mazin, Jeschke, Lechermann, Lee, Fink, Thomale, Valentí, Nat. Commun. **5**, 4261 (2014)

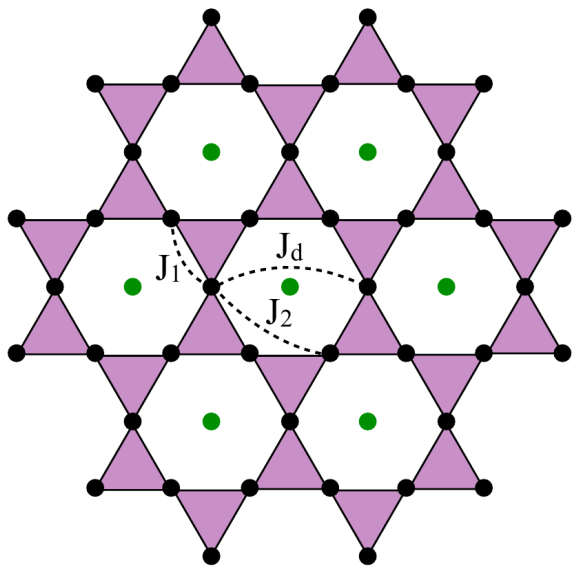
Search for Quantum spin liquids

B2

- Barlowite $Cu_4(OH)_6FBr$ has structure similar to Herbertsmithite, but Cu also on interlayer site
- no spin liquid because of significant interlayer couplings
- Kapellasite $ZnCu_3(OH)_6Cl_2$ predicted to be quantum paramagnet at $p \approx 8$ GPa







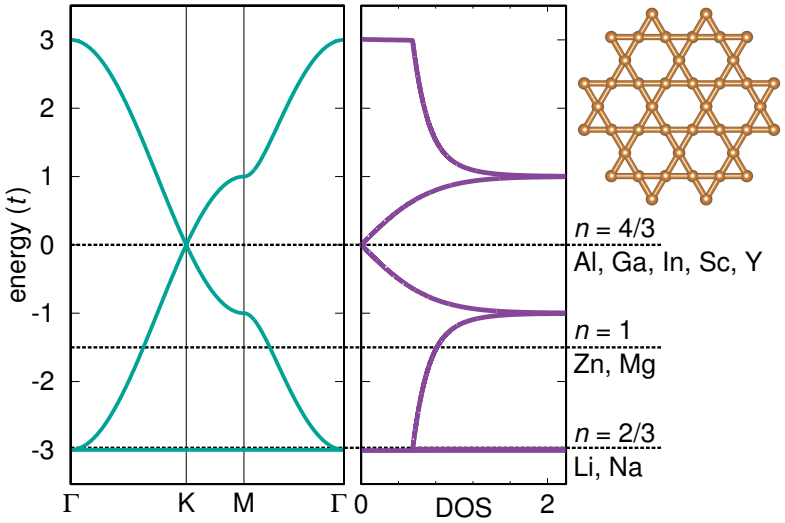
Iqbal, Jeschke, Reuther, Valentí, Mazin, Greiter, Thomale, arxiv:1506.03436

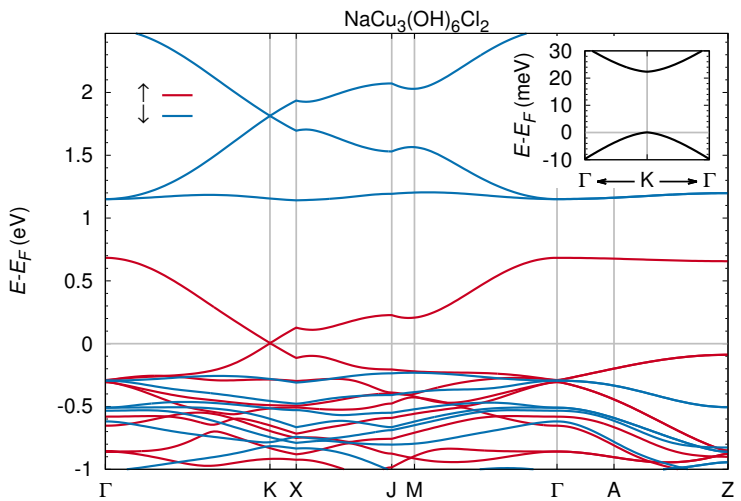
Jeschke, Salvat-Pujol, Gati, Hoang, Wolf, Lang, Schlueter, Valentí, PRB **92**, 094417 (2015)

Exotic states in new Kagome lattice materials: doping of Herbertsmithite and Barlowite

B2

- all proposed modifications for Herbertsmithite are stable in DFT
- hole-doping generates ferromagnet with Fermi level at Dirac point
- spin-orbit coupling opens a gap, strongly correlated topological insulator
- prospect for Quantum Anomalous Hall effect at elevated temperatures
- doping Barlowite interlayer sites with nonmagnetic atoms could generate Quantum Spin Liquid
- only Mg-Barlowite is a stable modification (unstable: Li, Na, K, Ca, Zn, Cd, Hg)



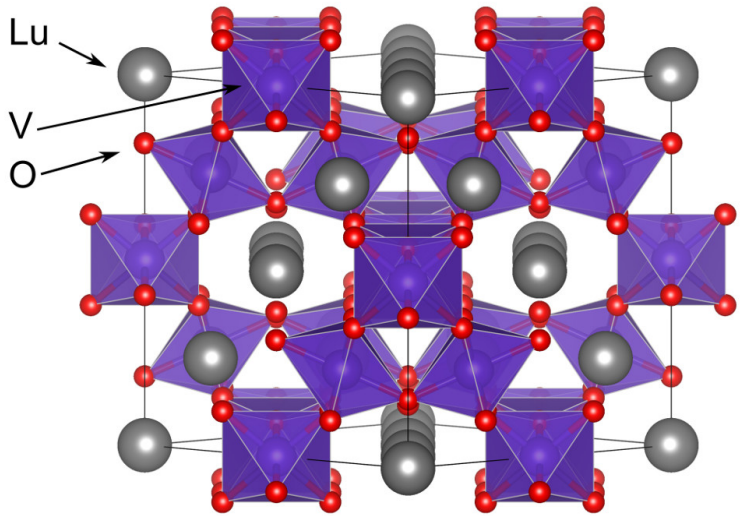


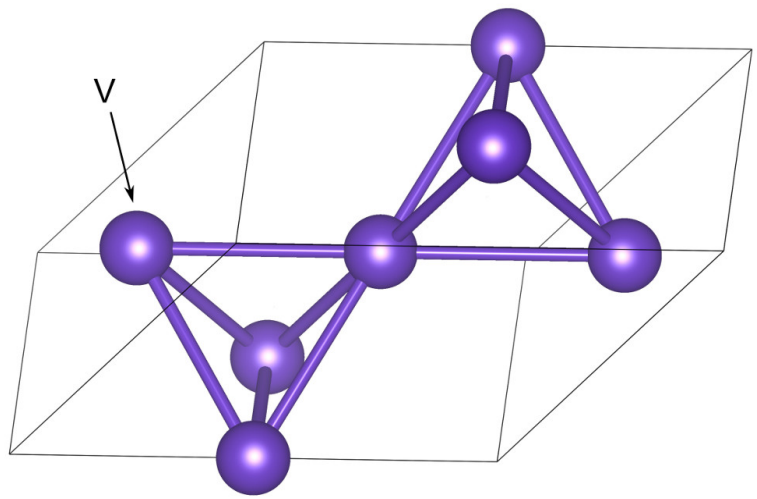
Guterding, Jeschke, Valentí, in preparation

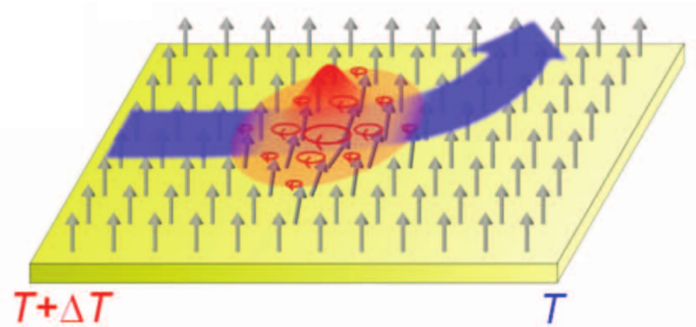
Low-energy model for pyrochlore $Lu_2V_2O_7$: *ab-initio* Spin Hamiltonian

B13

- Anomalous Thermal Hall effect of magnons measured
- hard to calculate effective spin-Hamiltonian directly with DFT
- new approach using exact diagonalization and subspace projection
- hopping parameters $t_{i\alpha,j\beta}$ from projective Wannier functions
- spin-orbit coupling strength λ from fit to relativistic band structure





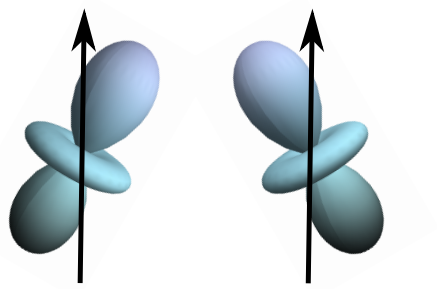


Onose, Ideue, Katsura, Shiomi, Nagaosa, Tokura, Science **329**, 297 (2010)

Low-energy model for pyrochlore $Lu_2V_2O_7$: exact diagonalization and projection

B13

$$H = \sum_{\langle ij\rangle}\sum_{\alpha\beta}\sum_{\sigma} t_{i\alpha,j\beta}\, d^{\dagger}_{i\alpha\sigma} d_{j\beta\sigma} + \lambda \sum_{i}\sum_{\alpha\beta}\sum_{\sigma\sigma'} [\langle i\alpha\sigma|\vec{L}\cdot\vec{S}|i\beta\sigma'\rangle d^{\dagger}_{i\alpha\sigma} d_{i\beta\sigma'} + \text{h.c.}]$$
$$+ U\sum_{i}\sum_{\alpha} n_{i\alpha\uparrow} n_{i\alpha\downarrow} + U'\sum_{i}\sum_{\alpha\neq\beta} n_{i\alpha\uparrow} n_{i\beta\downarrow} + (U' - J_H)\sum_{i}\sum_{\alpha<\beta}\sum_{\sigma} n_{i\alpha\sigma} n_{i\beta\sigma}$$
$$+ J_H \sum_{i}\sum_{\alpha\neq\beta} (d^{\dagger}_{i\alpha\uparrow} d^{\dagger}_{i\beta\downarrow} d_{i\alpha\downarrow} d_{i\beta\uparrow} + d^{\dagger}_{i\alpha\uparrow} d^{\dagger}_{i\alpha\downarrow} d_{i\beta\downarrow} d_{i\beta\uparrow}),$$



- scalar product $\vec{L}\cdot\vec{S}$ depends on site, different local coordinate systems
- diagonalize one bond exactly
- ground state is ferromagnet with one electron per site in d_{z^2} orbital
- evaluate J_{ij}, $\vec{D}_{ij}$, $\hat{K}_{ij}$ in low-energy subspace
- ratio of DM and Heisenberg (for $U = 2.25$ eV, $J_H = 0.7$ eV): 0.25
- experiment between 0.32 and 0.18

Poster by Kira Riedl, B2/B13

$$H = \sum_{i<j} [J_{ij}\hat{S}_i\hat{S}_j + \hat{S}_i\hat{K}_{ij}\hat{S}_j + \vec{D}_{ij}(\hat{S}_i\times\hat{S}_j)] + E_0$$

Summary and Outlook

B2 B13

Organic charge transfer salts

- realistic tight-binding models, unconventional SC
- investigate magnetic ordering in κ-Cl, ferroelectricity

Kagome lattice systems

- predicted new spin-liquid candidates, correlated Dirac metal, correlated ferromagnetic topological insulator
- investigate interplay of correlations and topology

Pyrochlore lattice systems

- developed method to parametrize effective Hamiltonians
- extend to larger system size, compare to experiment

Contributors

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Appendix

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Conversion from four-band to dimer model

- $t = \frac{1}{2}(t_2 + t_4)$
- $t' = \frac{1}{2}t_3$
- $U = 2t_1$

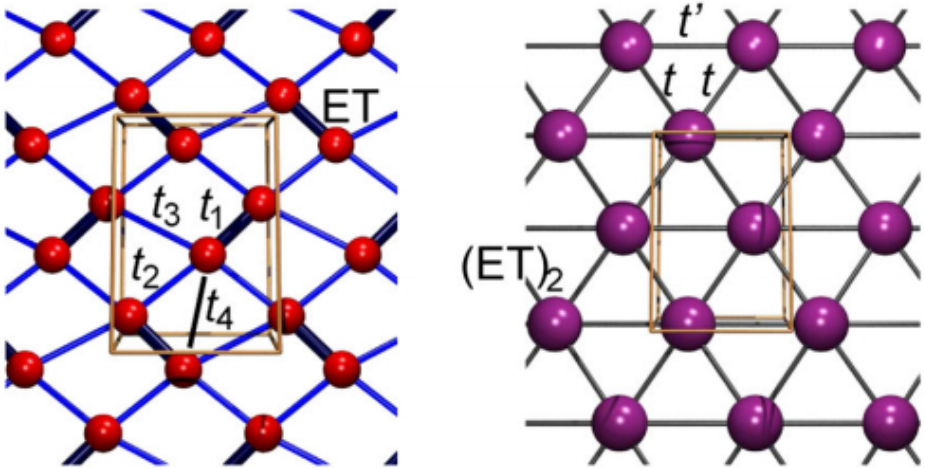


Figure : Kandpal et al., PRL **103**, 067004 (2009)

Tight binding+RPA formalism in a nutshell

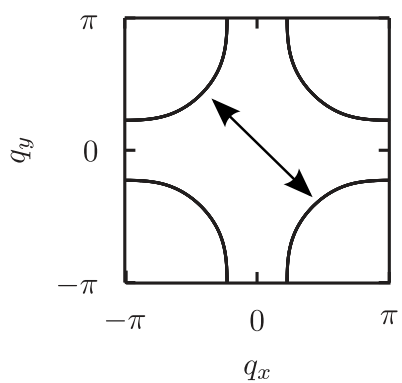
$$\chi^{pq}_{st}(\vec{q}) = -\frac{1}{N}\sum_{\vec{k},\mu,\nu} a^{s}_{\mu}(\vec{k})a^{p*}_{\mu}(\vec{k})a^{q}_{\nu}(\vec{k}+\vec{q})a^{t*}_{\nu}(\vec{k}+\vec{q})\frac{f(E_{\nu}(\vec{k}+\vec{q})) - f(E_{\mu}(\vec{k}))}{E_{\nu}(\vec{k}+\vec{q}) - E_{\mu}(\vec{k})}$$

$$\left[(\chi^{RPA}_{spin})^{pq}_{st}\right]^{-1} = \left[\chi^{pq}_{st}\right]^{-1} - (U_{spin})^{pq}_{st}$$

$$\Gamma^{pq}_{st}(\vec{k},\vec{k}') = \left[\frac{3}{2}U_s\,\chi^{RPA}_{s}(\vec{k}-\vec{k}')\,U_s + \frac{1}{2}U_s - \frac{1}{2}U_c\,\chi^{RPA}_{c}(\vec{k}-\vec{k}')U_c + \frac{1}{2}U_c\right]^{tq}_{ps}$$

$$\Gamma_{ij}(\vec{k},\vec{k}') = \sum_{stpq} a^{t*}_{i}(-\vec{k})a^{s*}_{i}(\vec{k})\mathrm{Re}\left[\Gamma^{pq}_{st}(\vec{k},\vec{k}')\right]a^{p}_{j}(\vec{k}')a^{q}_{j}(-\vec{k}')$$

$$-\sum_{j}\oint_{C_j}\frac{dk'_{\parallel}}{2\pi}\frac{1}{4\pi v_F(\vec{k}')}\left[\Gamma_{ij}(\vec{k},\vec{k}') + \Gamma_{ij}(\vec{k},-\vec{k}')\right]g_j(\vec{k}') = \lambda_i g_i(\vec{k})$$



Graser, Maier, Hirschfeld, Scalapino, New Journal of Physics **11**, 025016 (2009)