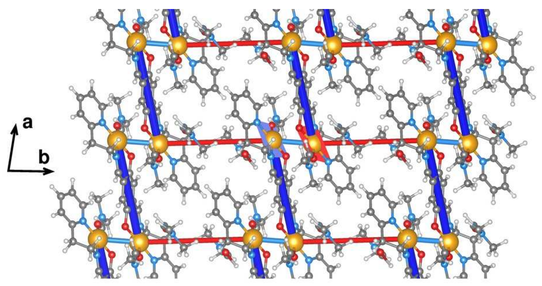
# B2

# *Ab initio* modelling and design of correlated materials

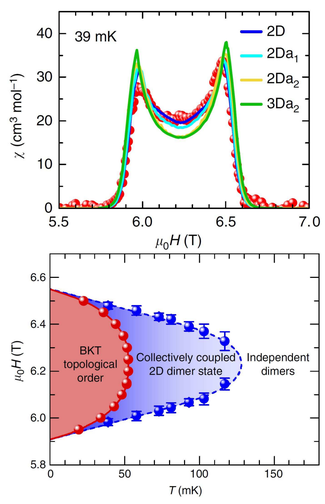
Roser Valentí (Universität Frankfurt)

## Quantum spin systems: Achievements

### Cull-p-hydroquinonate coordination polymer TK91: coupled dimer-based system




*ab initio* DFT + QMC

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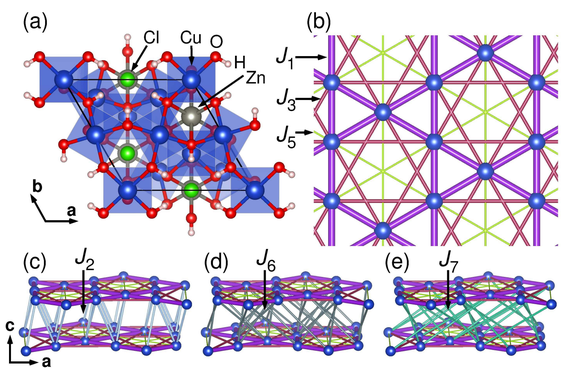
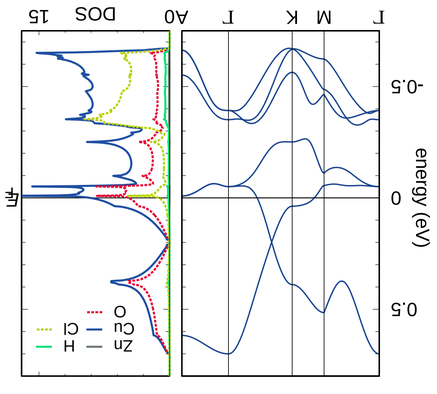
- Possible field-induced Berezinskii-Kosterlitz-Thouless topological order

U. Tutsch *et al.*, Nature Communications **5**, 5169 (2014).

### Herbertsmithite $ZnCu_3(OH)_6Cl_2$: Kagome system / spin liquid

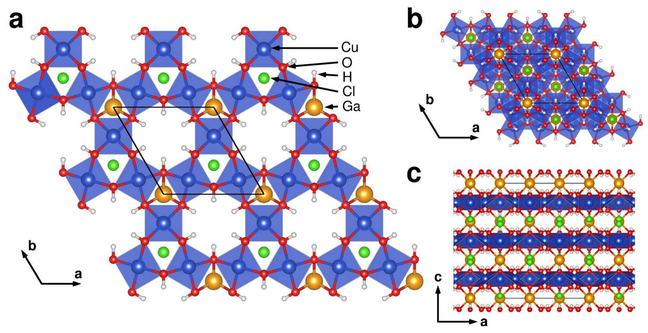



- Determination of exchange couplings via *ab initio* DFT
- Inclusion of correlations opens gap at $E_F$: Mott insulator

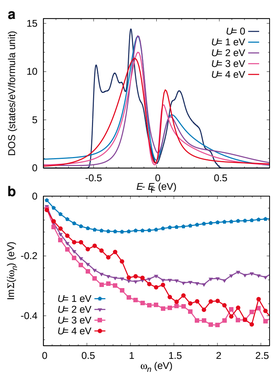
H. O. Jeschke, F. Salvat-Pujol, R. Valentí, Phys. Rev. B **88**, 075106 (2013).

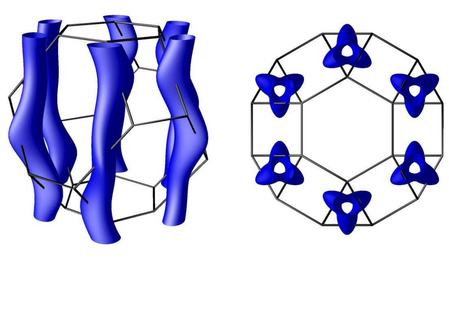
### Prediction of a strongly correlated Dirac metal




- DFT + many-body methods
- Exotic behavior by doping Herbertsmithite
- Change filling $n = 1$ to $n = \frac{4}{3}$: Replace $Zn^{2+}$ by $Ga^{3+}$
- $GaCu_3(OH)_6Cl_2$

- Dirac points at the Fermi surface
- $H = \sum_{i,j}\sum_{\sigma} t_{ij} c^{\dagger}_{i,\sigma} c_{j,\sigma} + \mu \sum_{i,\sigma} c^{\dagger}_{i,\sigma} c_{i,\sigma} + U \sum_i n_{i,\uparrow} n_{i,\downarrow} + \frac{V}{2} \sum_{\langle i,j \rangle} \sum_{\sigma,\sigma'} n_{i,\sigma} n_{j,\sigma'}$
- Possible instabilities to charge order, ferromagnetism, *f*-wave superconductivity → dynamical cluster approximation, functional RG



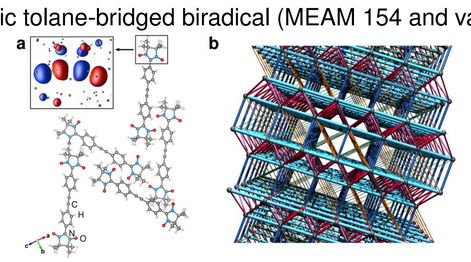



I. I. Mazin, H. O. Jeschke, F. Lechermann, H. Lee, M. Fink, R. Thomale, R. Valentí, Nature Communications **5**, 4261 (2014).

## Project goals and program

### Coupled dimer-based systems

Organic tolane-bridged biradical (MEAM 154 and variations)




- resolve the underlying important interactions via *ab initio* DFT total energy calculations
- thermodynamics via QMC

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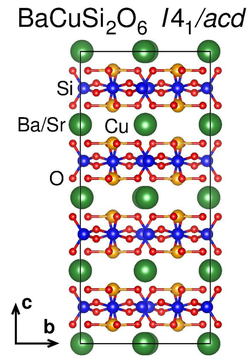
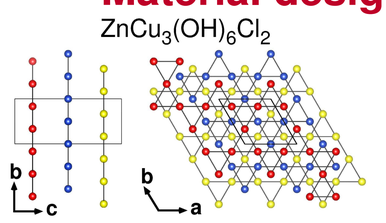
$BaCuSi_2O_6$ $I4_1/acd$

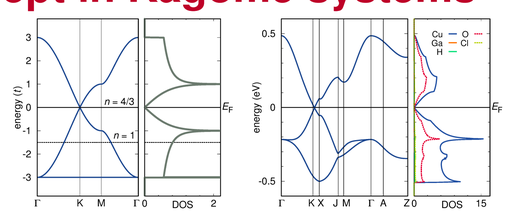



- Exchange couplings: high T ↔ low T structures
- field-induced 2D quantum critical point, origin?

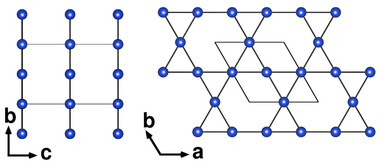
### "Material design" concept in Kagome systems

$ZnCu_3(OH)_6Cl_2$




$Cu_4(OH)_6FBr$




- prediction of structures at different fillings: use of genetic algorithms
- Dzyaloshinski-Moriya? full relativistic non-collinear *ab initio* calculations
- new frustrated geometries
- role of correlations? mapping to extended Hubbard model

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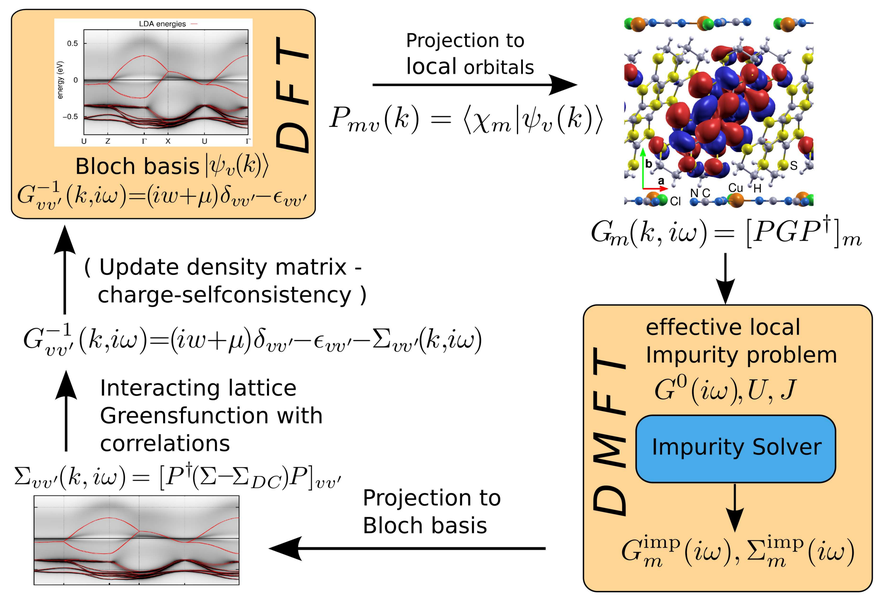
## Methods

- *ab initio density functional theory* with various basis sets as implemented in WIEN2k, ELK, FLEUR, FPLO, VASP, QUANTUM ESPRESSO and GPAW




- finite pressure structure prediction using the strain tensor (M. Tomić *et al.* Phys. Rev. B **85**, 094105 (2012), S.A.J. Kimber *et al.* PNAS 111, 5106 (2014))
- crystal structure prediction with genetic algorithms (USPEX) (A.R. Organov *et al.* J. Chem. Phys. **124**, 244704 (2006)).
- functionals: LDA, GGA, LDA+U, HSE. Van der Waals corrections
- phonons/lattice stability/finite temperature: Born-Oppenheimer Molecular Dynamics (K. Muthukumar *et al.* J. Chem. Phys. **140**, 184706 (2014).
- polarization/multiferroic properties: Berry phase formalism in DFT + effective model calculations. R.D. King-Smith, D. Vanderbilt, Phys. Rev. B **47**, 1651 (1993)
- many-body approaches: exact diagonalization, variational Monte Carlo, DMFT and fluctuation exchange theory.

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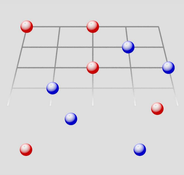

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