B13 Investigation of spin systems via *ab initio* Quantum Monte Carlo and perturbation theory

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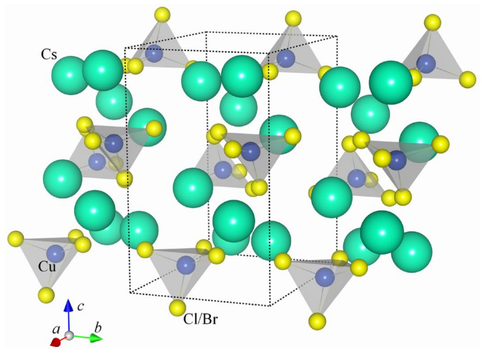
Introduction: Spin systems

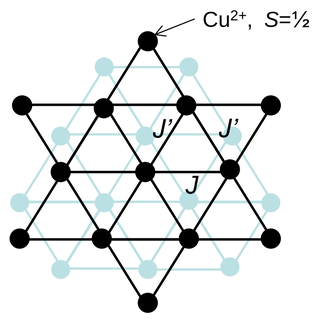
• Low-dimensional quantum spin systems offer a rich playground

$$H^{\text{Heis}} = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j$$

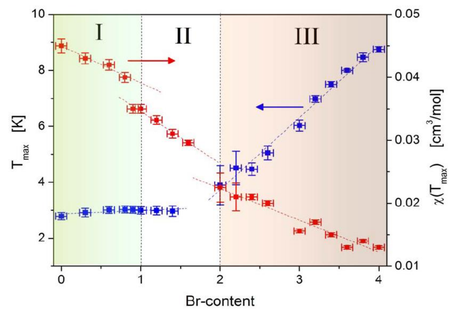
- ✓ Dimensionality interplaying with quantum effects
- ✓ Frustration effects
- ✓ Exotic collective excitations (spinons, triplons, BEC of magnons, …)
- ✓ Tunability through chemical doping or pressure

Copper halides $Cs_2CuCl_{4-x}Br_x$





- ✓ 2D AFM triangular lattice
- ✓ BEC of magnons
- ✓ Rich phase diagram: Spin liquid phase, magnetization plateaux, …
- ✓ Varying frustration (*J*/*J'*) via chemical doping: $Cs_2CuCl_{4-x}Br_x$
- ✓ Site-selective substitution



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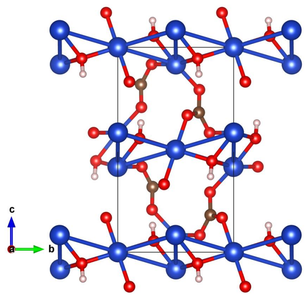
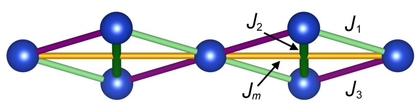
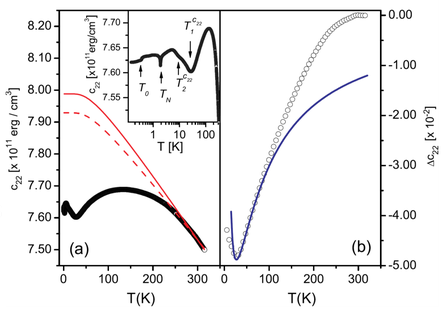
Cong *et al*., PRB **83**, 064425 (11)
van Well *et al*., PRB **91**, 035124 (15)

Azurite $Cu_3(CO_3)_2(OH)_2$





- ✓ 1D diamond chain with AFM couplings
- ✓ Concomitant structural and magnetic transitions
- ✓ Exceptionally strong magneto-elastic coupling



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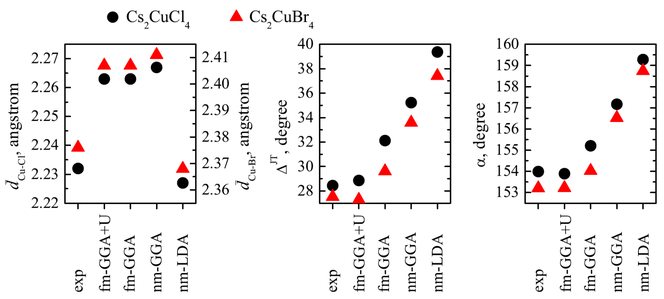
Jeschke *et al*., PRL **106**, 217201 (11)
Cong *et al*., PRB **89**, 174427 (14)

Ab initio approach: DFT and QMC

a) Achievements: DFT studies of copper halides

- Electronic structure of end members
- Structural relaxation
  - ✓ Equilibrium crystal sructure depends on the DFT approximation used



Foyevtsova *et al*., PRB **83**, 125126 (11)

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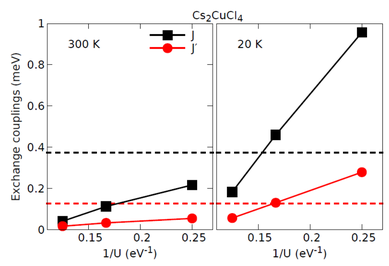


- Spin superexchnage for end members:
  - ✓ Strong bond-geometry dependence

Possible temperature dependence?

van Well *et al*., PRB **91**, 035124 (15)

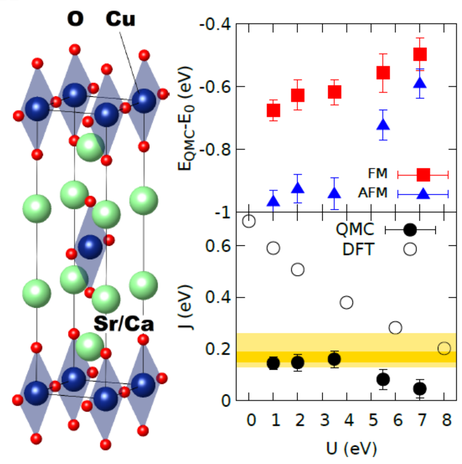
b) Achievements: QMC studies of a 1D chain antiferromagnet

- ✓ Ca_2CuO_3: The best representation of a 1D AFM chain
- ✓ The largest *J*
- ✓ The *U* value from LDA+U used as variational parameter
- ✓ The best trial WF determined using the variational nature of the DMC energy
- ✓ A good starting point for further QMC studies of cuprates!

Spin superexchange from QMC:

excellent agreement with experiment!

Ca_2CuO_3



Foyevtsova *et al*., PRX **4**, 031003 (14)

c) Goals and work program

Copper halides and azurite

1) **QMC** equilibrium crystal structure and phonons
   - ✓ Frozen phonon approximation
   - ✓ Specific heat
   - ✓ Thermal expansion
   - ✓ Elastic constants

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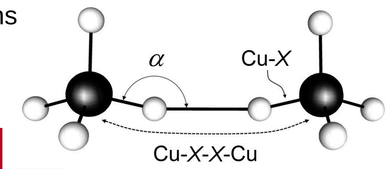
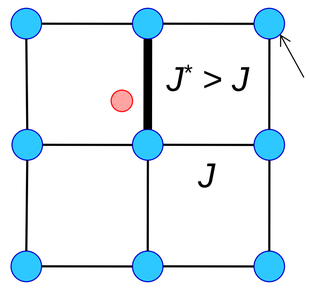


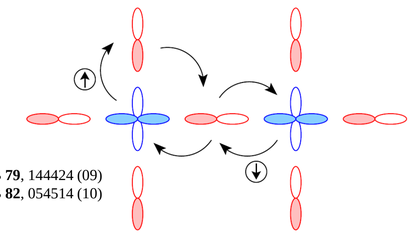
2) **QMC** spin superexchange couplings
   - ✓ Total energy difference approach
3) Combining **QMC** and **DFT**: use QMC to single out the best DT functional — B2
4) Benchmarking **QMC** calculations on simple systems
   - ✓ $CuCl_2$, CuF_2
   - ✓ Organic molecules — B6

Many-body perturbation theory

a) Achievements

• Perturbation theory for a 3-band model of a CuO_2 plane in the cuprates





Foyevtsova *et al*., PRB **79**, 144424 (09)
Foyevtsova *et al*., PRB **82**, 054514 (10)

- Superexchange as a function of doping-induced inhomogeneities
- Relates to STS measurements on the cuprates' surfaces
- Results qualitatively depend on the complexity of the model → importance of ligand atoms (oxygen)
- DFT used to parametrize the Hubbard model

b) Goals and work program

Copper halides and azurite

• By combining DFT/QMC and perturbation theory calculations, to design a code that efficiently and accurately calculates spin superexchange as a function of

- ✓ Doping and disorder
  - ❖ Mixed copper halides
- ✓ Fine structural details
  - ❖ Temperature effects
  - ❖ Spin-phonon coupling (magneto-elastic measurements)
  - ❖ Electron-phonon coupling

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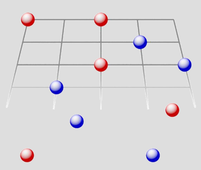
• Advantages:
  - ✓Efficient, expecially for evaluating interactions with phonons (avoids lengthy *ab initio* calculations)
  - ✓ Flexible (accuracy is controlled by the expansion order, number of parameters, *etc.*)
  - ✓ Model parameters are provided by *ab initio* calculations



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