2D magnetism in the metal-organic framework $Cu(C_4H_4N_2)_2(H_2O)_2Cr_2O_7$

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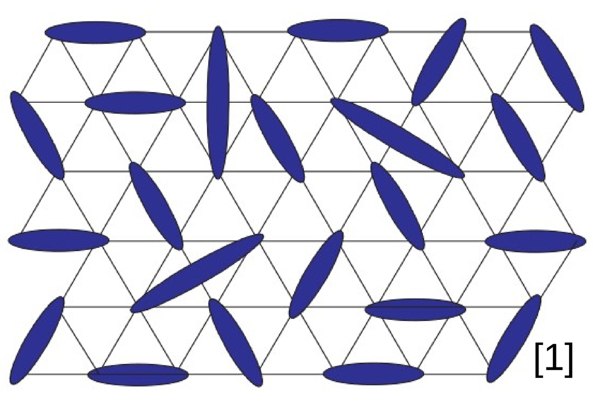
1. Why study low-dimenasional magnetism?

- Frustrated magnets
  - Quantum spin liquid
- High-T_c superconductivity
  - Fe-Pnictides
  - Cuprates
- Fractional excitations
  - 1D spinon continuum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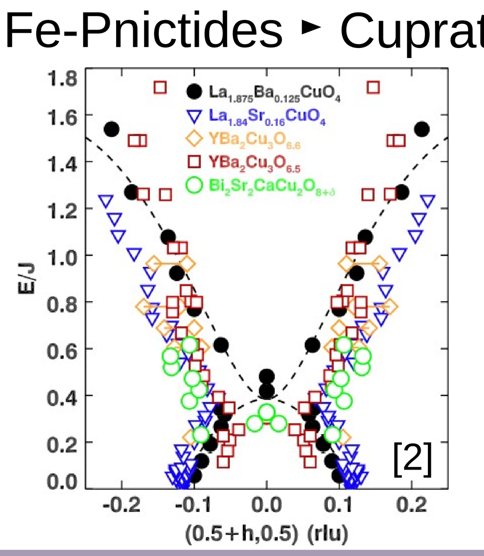
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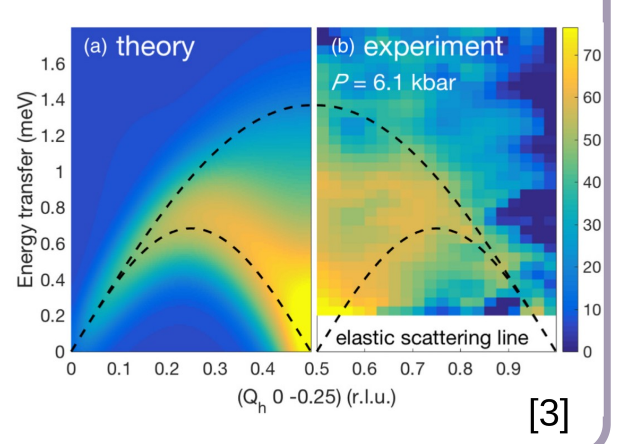
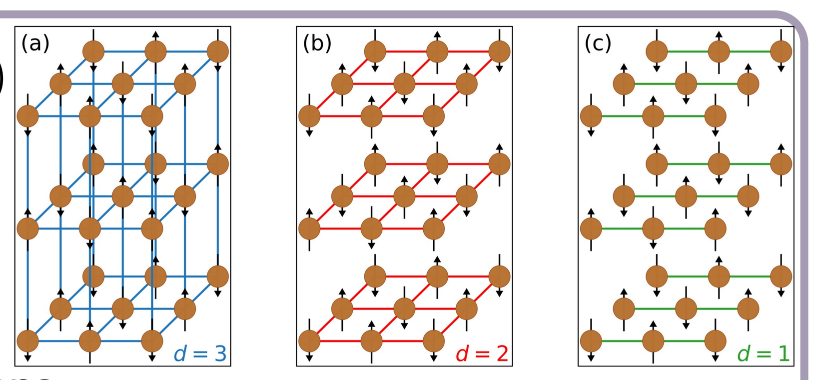


[3]

2. Introduction to low-dimensional magnetism

$$H = -\sum_{i,j}(J_x S_{ix} S_{jx} + J_y S_{iy} S_{jy} + J_z S_{iz} S_{jz})$$



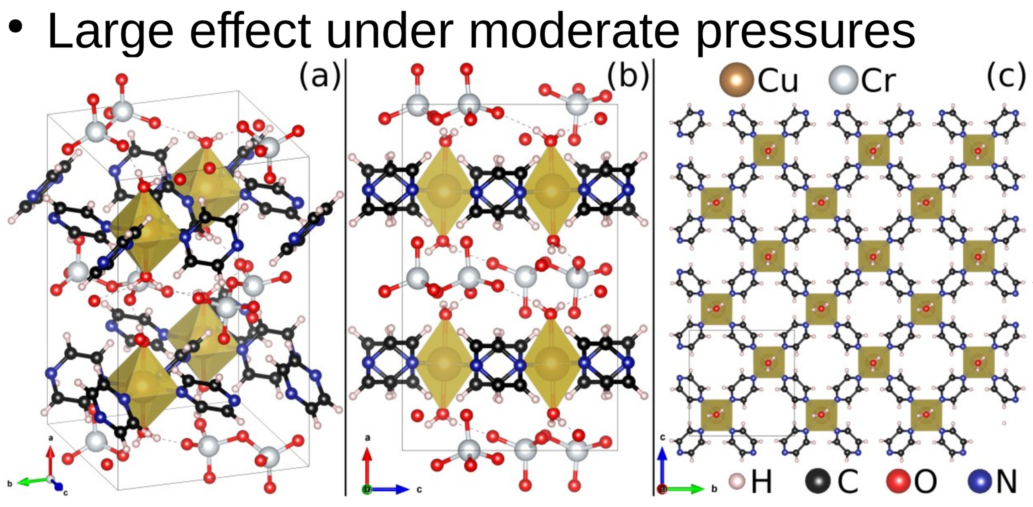
- Two ways to be low dimensional: lattice (*d*) and spin (*s*)
- Dimensionality fundamentally influences:
  - ▸ Phase transitions ▸ Ground state ▸ Excitations

Spin-dimensionality	Interaction	Model
n = 1: S_z^2	J_z	Ising
n = 2: $S_x^2 + S_y^2$	$J_x = J_y$ $J_x = 0$; J_y	Planar Planar Ising
n = 3: $S_x^2 + S_y^2 + S_z^2$	$J_x = J_y = J_z$ $J_z = 0$; $J_x = J_y$ $J_x = J_y = 0$; J_z	Heisenberg XY Z

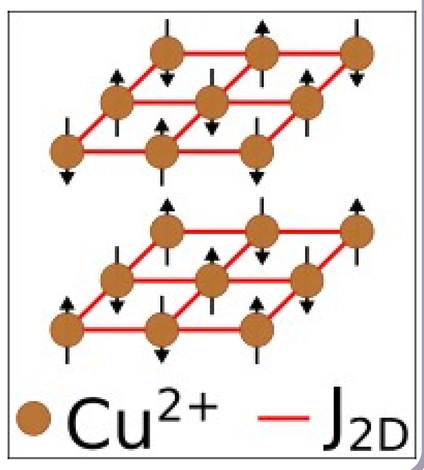
[4]

3. Cu-pyz a metal-organic model system

- Metal-organic compounds (MOC): A versatile platform for low-d magnetism
- Coordinate mag. ions with neutral ligands and countering anionic complexes
  - $Cu(C_4H_4N_2)_2(H_2O)_2Cr_2O_7$ (Cu-pyz) [5]
- MOCs are susceptible to pressure and allow tuning of mag. Interactions
  - High compressibility due to organic constituents
  - Anisotropic deformation due to different coordinating ligands
- Large effect under moderate pressures

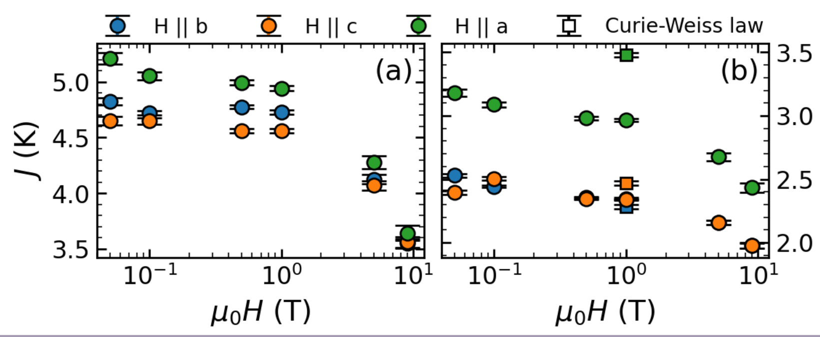


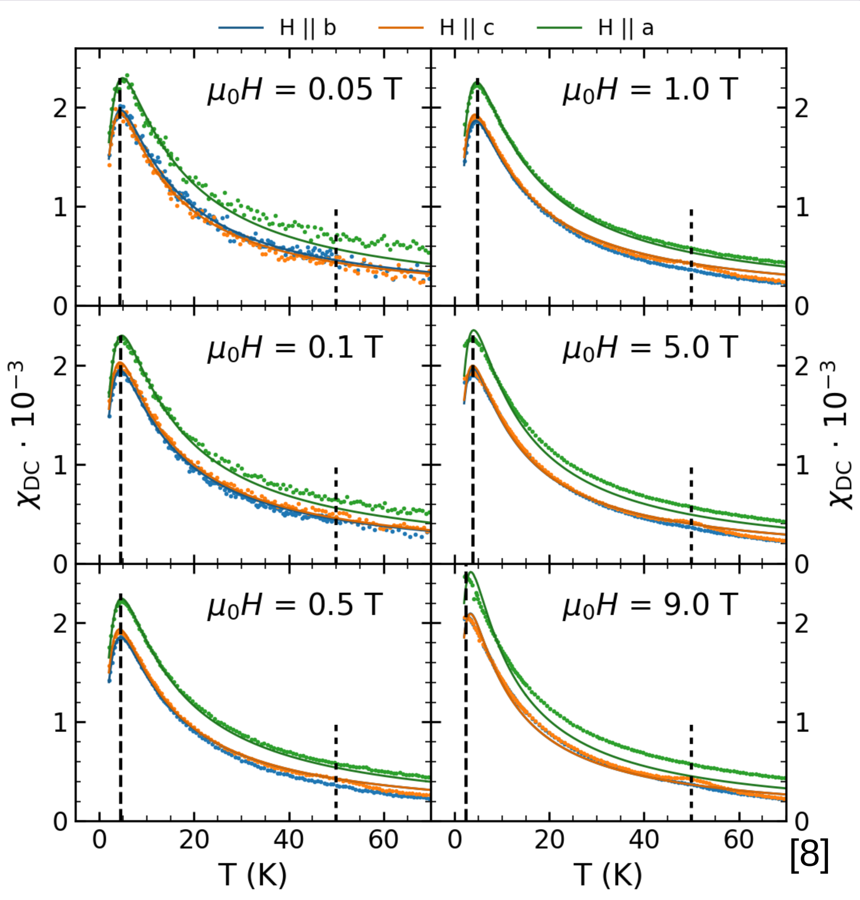
Abstraction of the mag. square lattice



4. Static susceptibility – Signature of 2D AFM

- Broad peak corresponds to intralayer exchange coupling strength J_{2D}
- Fit Padé approximation for a 2D AFM square lattice [6] → $J_{2D} \approx 0.5$ meV
- No sign of mag. LRO below $T < 2$ K
- Good realization of a 2D magnet [7]
- Octahedral constituents introduce *g*-factor anisotropy





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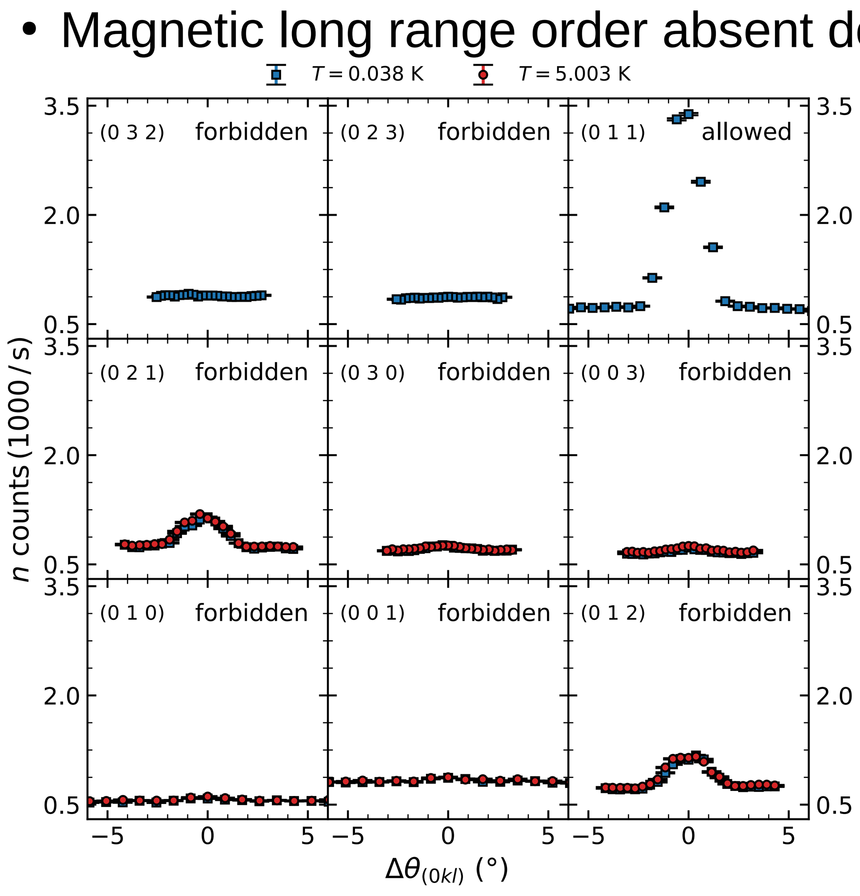
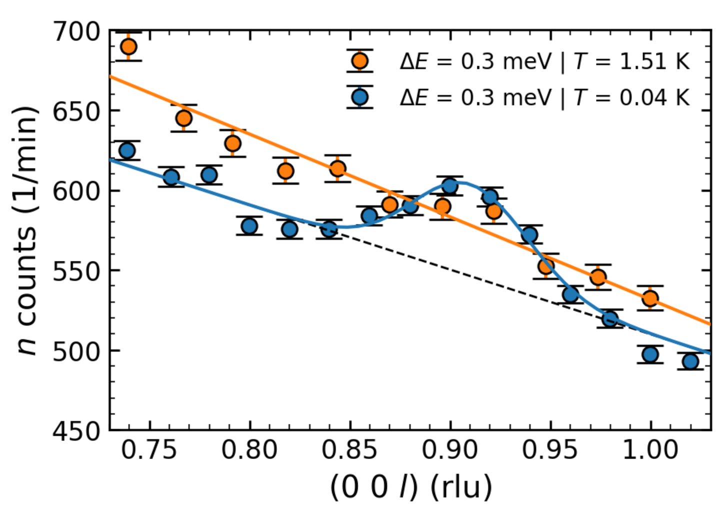
5. Neutron scattering – Emergent spin waves

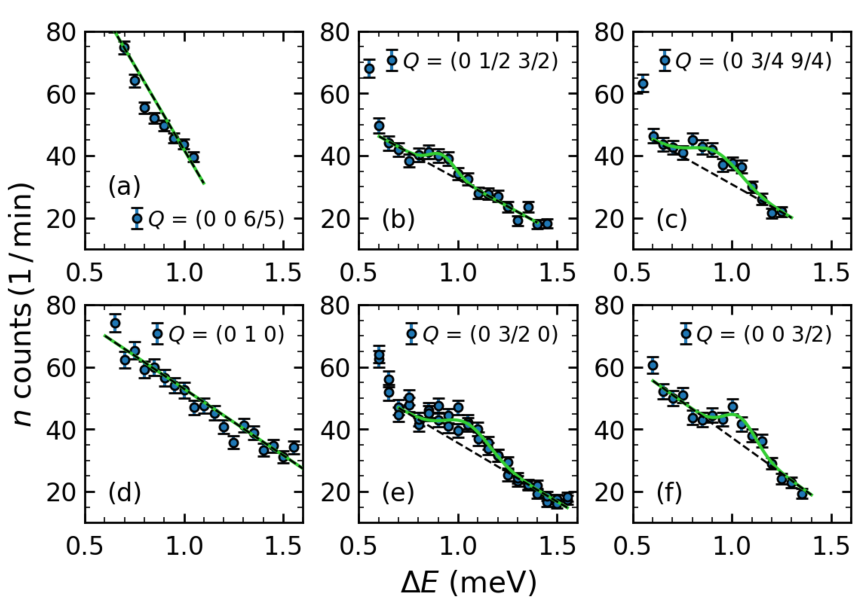
- No magnetic Bragg scattering observed at integer and half-integer positions
- Magnetic long range order absent down to $T = 0.04$ K

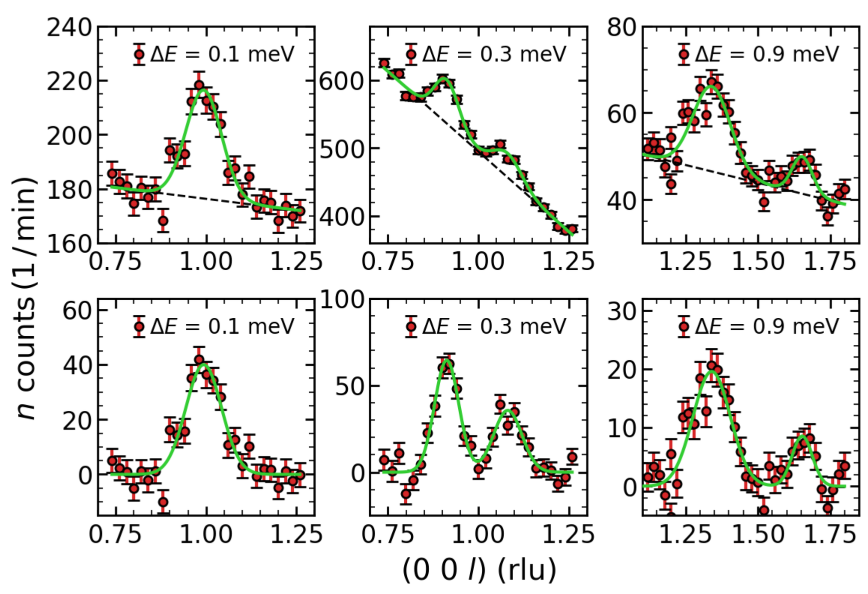


- Inelastic magnetic signal emerges around the (010) and (001) positions
- Spin waves propagate within the ordered, individual bc-planes



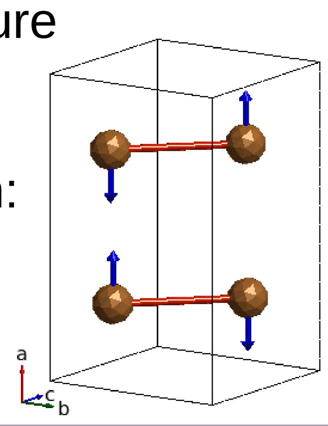
- Mapping spin wave dispersion in [010] x [001] scattering plane at IN12 (ILL)
  - Constant-*Q* scans
  - Constant-*E* scans

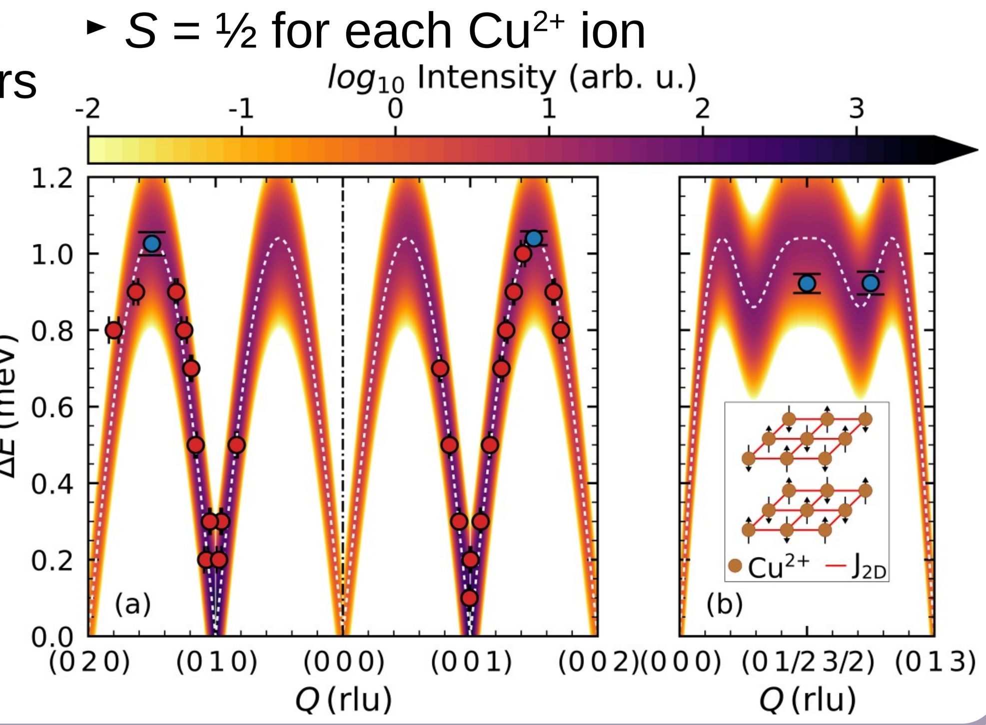




6. Modeling the spin wave dispersion

- Model building and calculation of spin wave dispersion using *spinW* [9]
- Initial model parameters
  - AFM exchange $J_{2D} = 0.5$ meV between next neighbors
  - Magnetic structure compatible with putative mag. zone centers (010) and (001)
  - No out-of-plane coupling $J_\perp = 0$ meV
  - $S = ½$ for each Cu^{2+} ion
- Fitting the dispersion yields $J_{2D} = 0.52(3)$ meV
- Magnetic structure consistent with measured spin wave dispersion:





7. Conclusion

- Susceptibility and magnetization agrees well with the model of a 2D quantum Heisenberg antiferromagnet
- Magnetic long range order is absent down to $T = 0.038$ K
- Spin wave dispersion refines the exchange constant: $J_{2D} = 0.52(3)$ meV and suggests G-type AFM order

8. References

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