From spin chains to corner-sharing triangles in the compound family $ACuTe_2O_6$ A=Sr, Ba, Pb

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Outline

$ACuTe_2O_6$ family of magnets
Isostructural, cubic crystal symmetry
Quantum spin-1/2

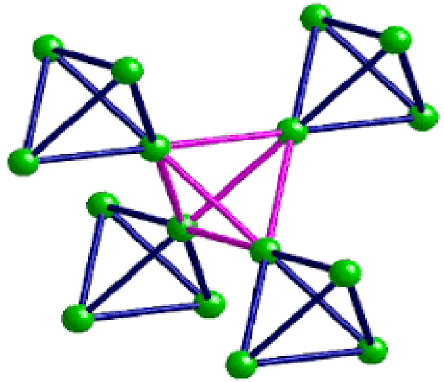
Explore the space group and possible magnetic lattices

$BaCuTe_2O_6$ – coupled spin chains

$SrCuTe_2O_6$ – coupled spin chains

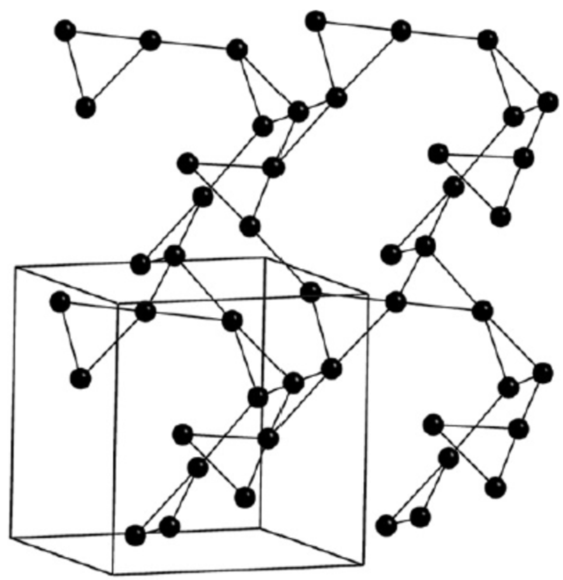
$PbCuTe_2O_6$ – corner-sharing triangles
– spin liquid candidate

3-dimensional arrangements of corner-sharing triangles



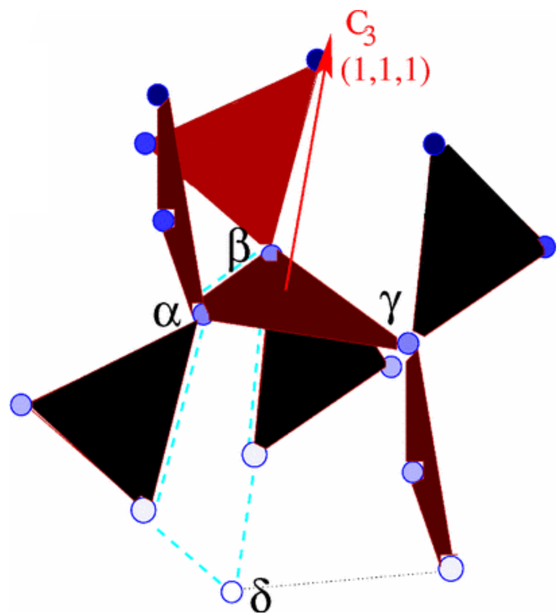
Pyrochlore

Hyperkagome lattice–
2 triangles at each corner
10 spins loops



M. V. Talanov, V. B. Shirokov, V. M. Talanov, Nanosystems: Physics, Chemistry, Mathematics, 6, 442 (2015)

Trillium lattice–
3 triangles at each corner
5 spin loops



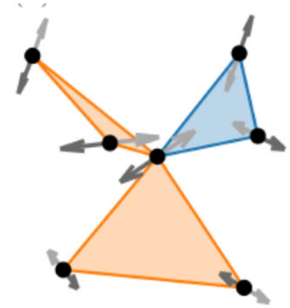
J. M. Hopkinson, H.-Y. Kee Phys. Rev. B 74, 224441 (2006)

Distorted Windmill Lattice

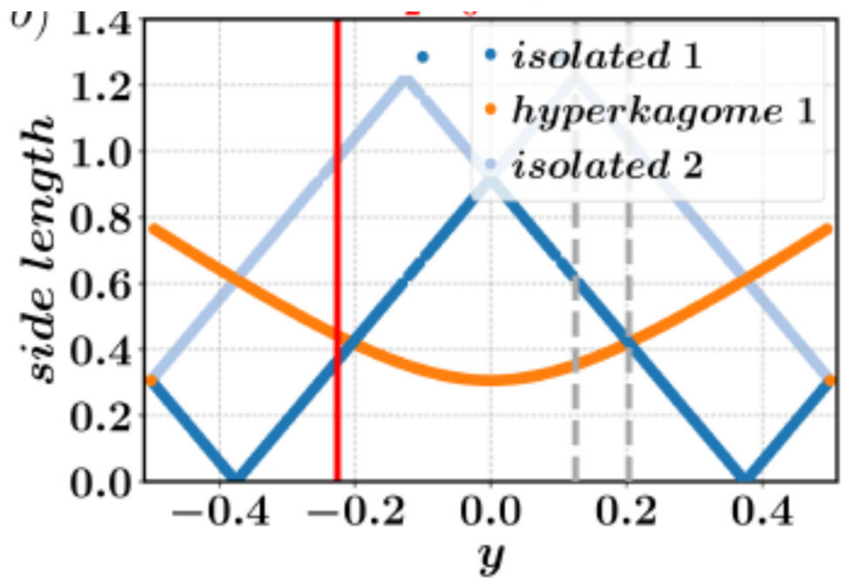
- Cubic space group $P4_132$
- Magnetic ions at Wyckoff 12d
- 12 equivalent sites per cell
- controlled by parameter *y*

Sublattice	Position
1	$(3/4 + y, 3/8, 1 - y)$
2	$(1/2 + y, 1/4 - y, 7/8)$
3	$(5/8, 1/2 - y, 3/4 - y)$
4	$(1/2 - y, 3/4 - y, 5/8)$
5	$(3/4 - y, 5/8, 1/2 - y)$
6	$(7/8, 1/2 + y, 1/4 - y)$
7	$(1 - y, 3/4 + y, 3/8)$
8	$(1/4 - y, 7/8, 1/2 + y)$
9	$(3/8, 1 - y, 3/4 + y)$
10	$(1/4 + y, 1/8, y)$
11	$(1/8, y, 1/4 + y)$
12	$(y, 1/4 + y, 1/8)$

A. Fancelli, J. Reuther, B. Lake
Phys. Rev. B 108, 184415 (2023)



For general *y*, each magnetic ion in
- 2 hyperkagome triangles
- 1 isolated triangle



$$y^* = \frac{9-\sqrt{33}}{16} \simeq 0.2035$$

For $-\frac{1}{2} < y < -y^*$ & $y^* < y < \frac{1}{2}$: isolated triangle

For $-y^* < y < y^*$ & $y = \pm\frac{1}{2}$: hyperkagome

For $y = \pm y^*$: hyperkagome & isolated triangles

For $y = \pm\frac{3}{8} = 0.375$: 4 sites, trillium lattice

From hyperkagome to trillium

$ACuTe_2O_6$ A=Sr, Ba, Pb - spin-1/2 windmill lattices

$P4_132$, Cu^{2+} S=1/2 @ 12d Wyckoff site

Neutron, powder by solid state reaction, 15 K

Atom	Mult.	x	y	z	B_{iso}
Te	24e	0.3371(3)	0.9180(3)	0.0640(3)	0.98(9)
Ba1	8c	0.0563(3)	0.0563(3)	0.0563(3)	0.36(19)
Ba2	4b	0.3750	0.6250	0.1250	0.96(24)
Cu	12d	0.4738(2)	0.8750	0.2761(2)	0.80(10)
O1	24e	0.5763(3)	0.9242(3)	0.3737(3)	1.02(10)
O2	24e	0.2662(2)	0.8115(5)	0.9900(3)	1.42(10)
O3	24e	0.2267(3)	0.9739(3)	0.1372(3)	1.39(9)

$$(1/4 - y,\ 7/8,\ 1/2 + y)$$

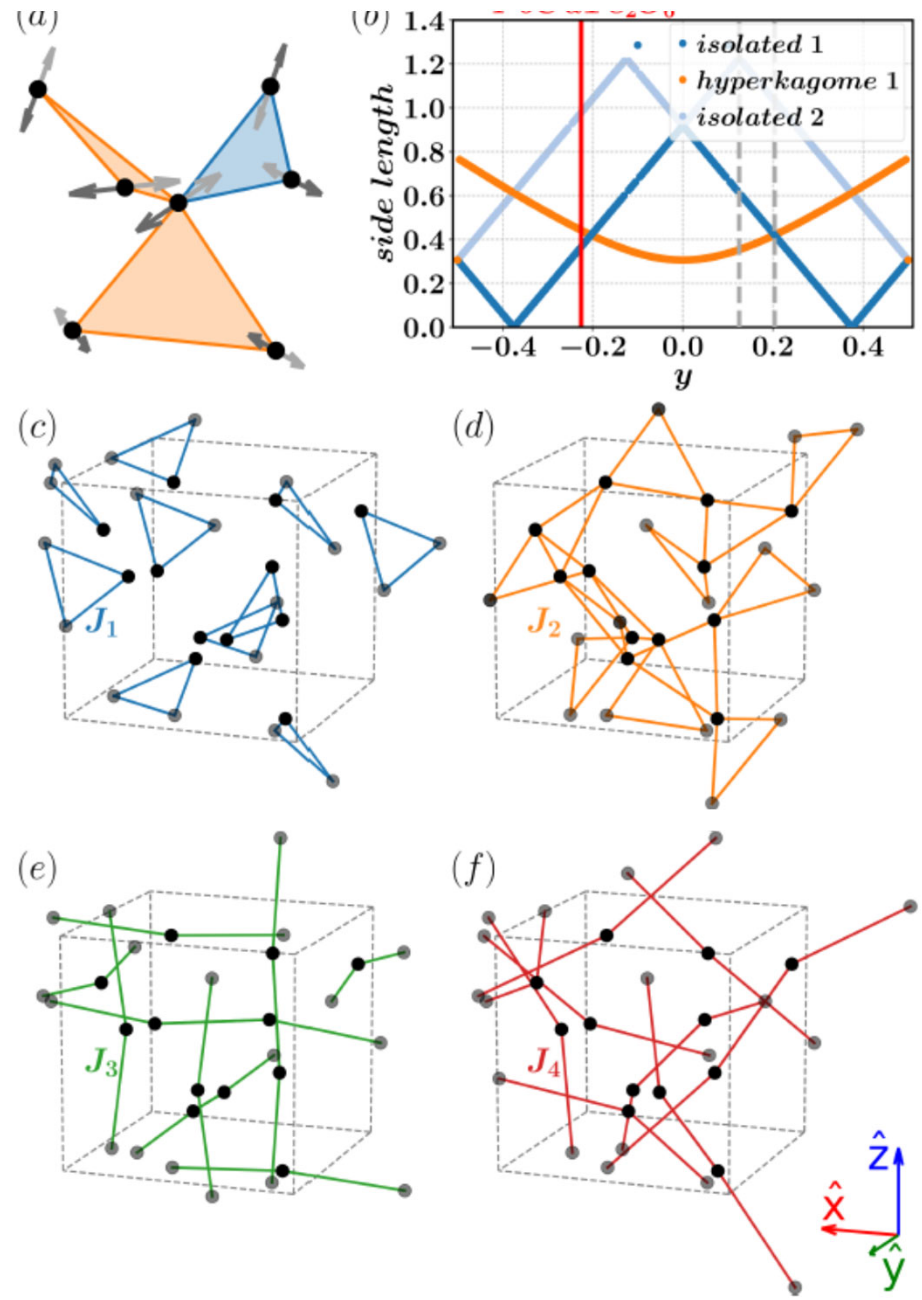
$PbCuTe_2O_6$, a=12.497 Å, y = −0.2258
$BaCuTe_2O_6$, a=12.829 Å, y = −0.2239
$SrCuTe_2O_6$, a=12.437 Å, y = −0.2255

For all three compounds

$$|y| > |y^*| = 0.2035$$

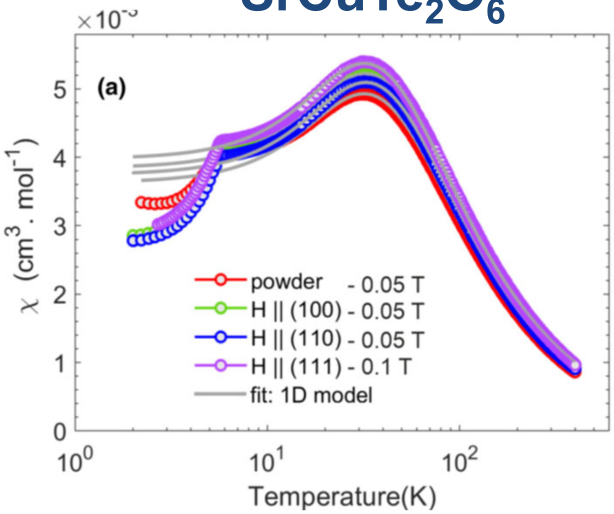
1st neighbor J_1 is the isolated triangle
2nd neighbor J_2 is the hyperkagome

But bond distances do not translate simply into interaction strengths
J_3 gives chains parallel to a, b, c
J_4 gives chains parallel to [1,1,0] etc



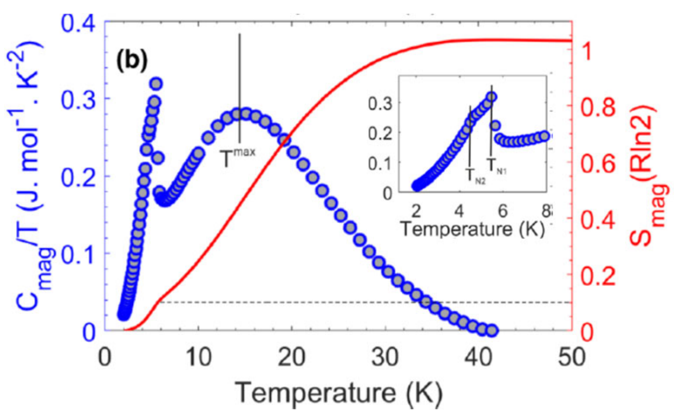
$ACuTe_2O_6$ A=Sr, Ba, Pb - DC susceptibility, heat capacity

$SrCuTe_2O_6$



θ_{CW} = -28$\pm$0.5K $\Rightarrow$ AFM

μ_{eff} = 1.85 μ_B $\Rightarrow$ S=1/2



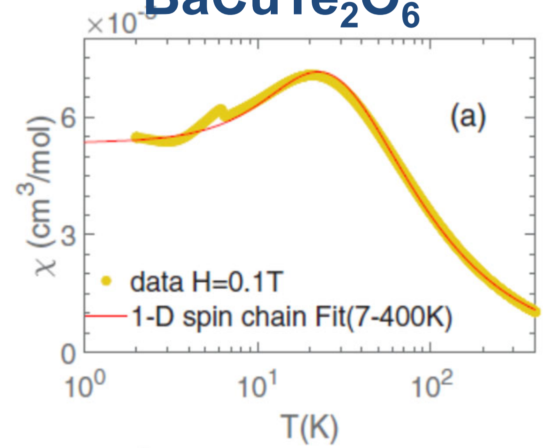
$T_{N1} \approx 5.5$ K & $T_{N2} \approx 4.5$ K

$\Rightarrow$ AFM order

S. Chillal et al

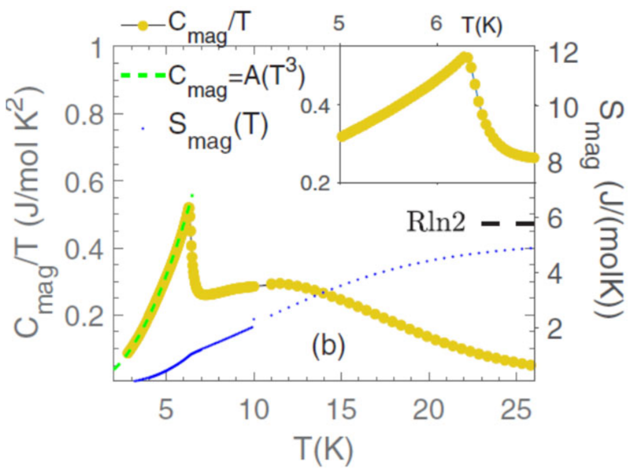
PRB 102, 224424 (2020)

$BaCuTe_2O_6$



θ_{CW} = -33$\pm$0.5K $\Rightarrow$ AFM

μ_{eff} = 1.96 μ_B $\Rightarrow$ S=1/2

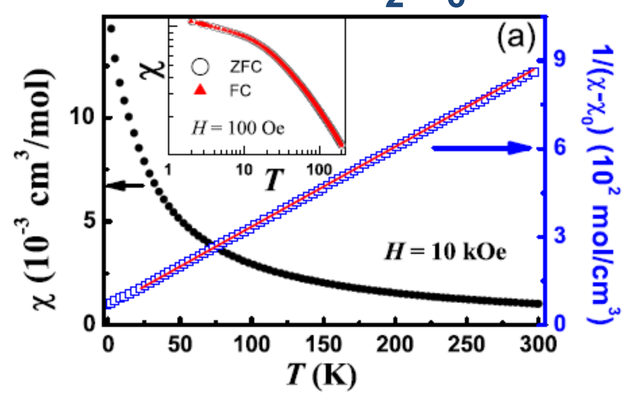


T_N = 6.31 K $\Rightarrow$ AFM order

A. Samartzis et al

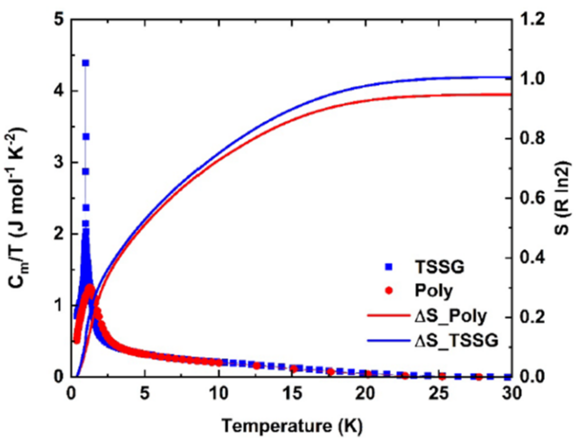
PRB 103, 094417 (2021)

$PbCuTe_2O_6$



θ_{CW} = -22$\pm$0.5K $\Rightarrow$ AFM

μ_{eff} = 1.75 μ_B $\Rightarrow$ S=1/2



$T_c < 1.0$ K $\Rightarrow$ **NO order**

B. Koteswararao et al

PRB 90 035141 (2014)

A. Hanna et al. PRM 5 113401 (2021)

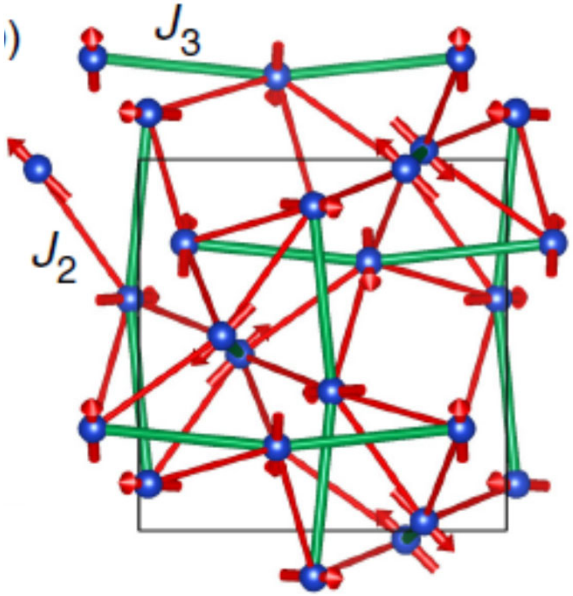
All compounds show suppression of magnetic order

$BaCuTe_2O_6$

Magnetic structure of $BaCuTe_2O_6$

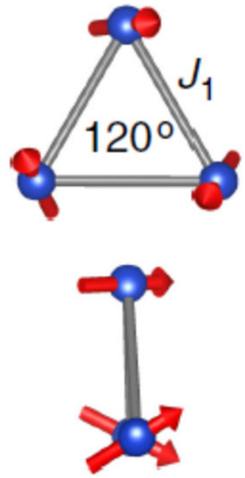
q=(0,0,1), m=0.46 μ_B/Cu^{2+} suppressed compared to 1 μ_B/Cu^{2+}

$\Rightarrow$ low-dimensional / frustrated



180° order along J_3 chains $\Rightarrow$ J_3 is AFM

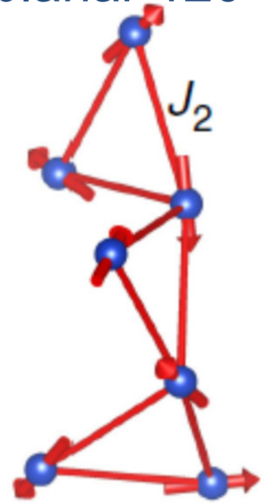
J_1 triangles canted 120°



isolated triangles

J_1 is very weak

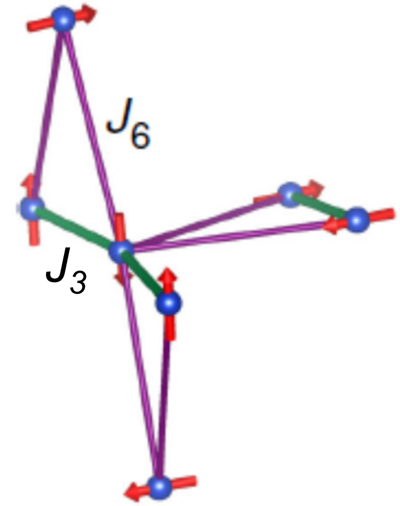
J_2 triangles planar 120°



hyperkagome triangles

$\Rightarrow$ J_2 is AFM

J_3/J_6 perpendicular order between chains



Neighboring parallel chains have 90° spins

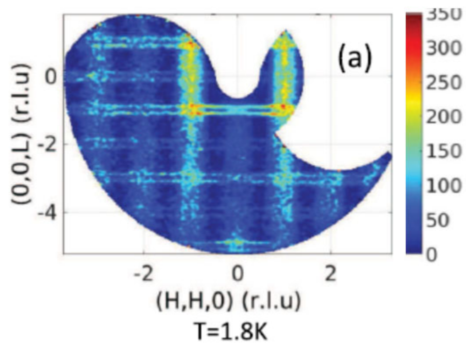
$\Rightarrow$ FM/AFM J_6 possible

Spin-1/2 Heisenberg AFM chains // a, b, c axes coupled by frustrated interchain interactions into a non-coplanar order

High energy magnetic excitations of $BaCuTe_2O_6$

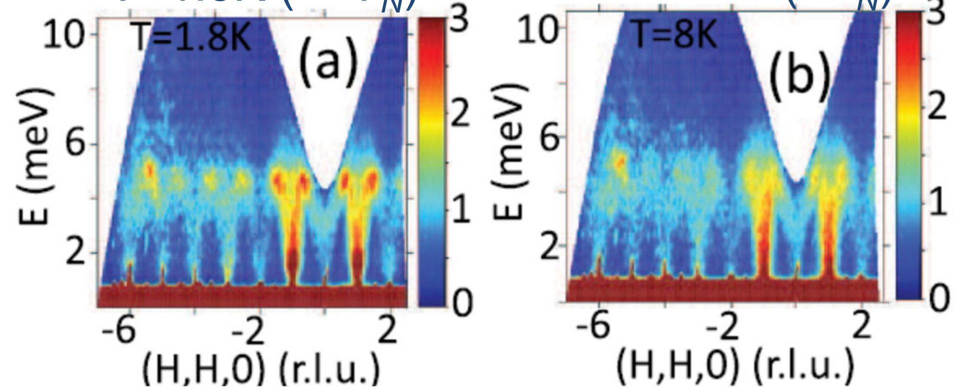
INS, [H,H,L] plane, 1.8 < *E* < 2.2 meV

T=1.8K (<<T_N)

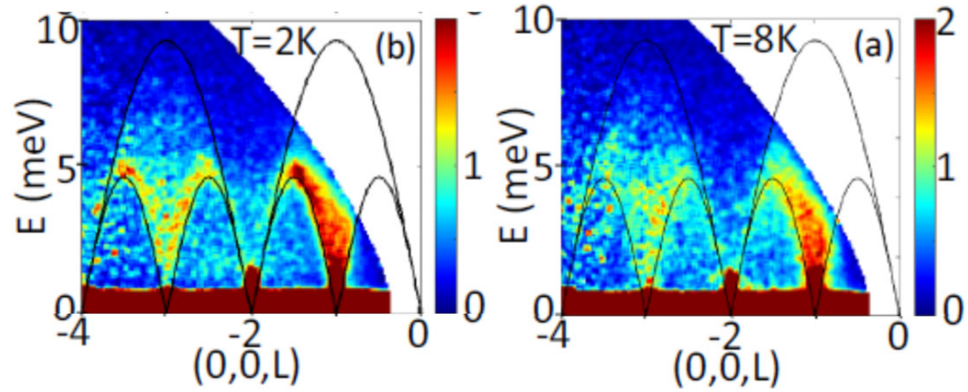


Energy-Wavevector maps

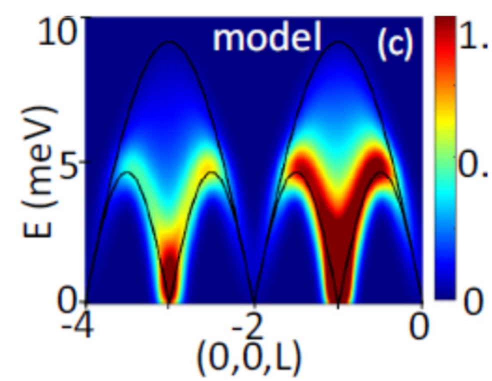
T=1.8K (<<T_N) *T*=8.0K (>T_N)



- Streaks indicate dispersionless scattering ⇒ 1D magnetism from J_3
- multi-spinon continuum typical of AFM S-1/2 chain
- Minor changes below T_N



- Comparison to the Bethe Ansatz
- des Cloiseaux-Pearson dispersion $(\pi J_3/2)|\sin(\pi L)|$ gives J_3 = 2.90(6) meV or 33.7(7) K

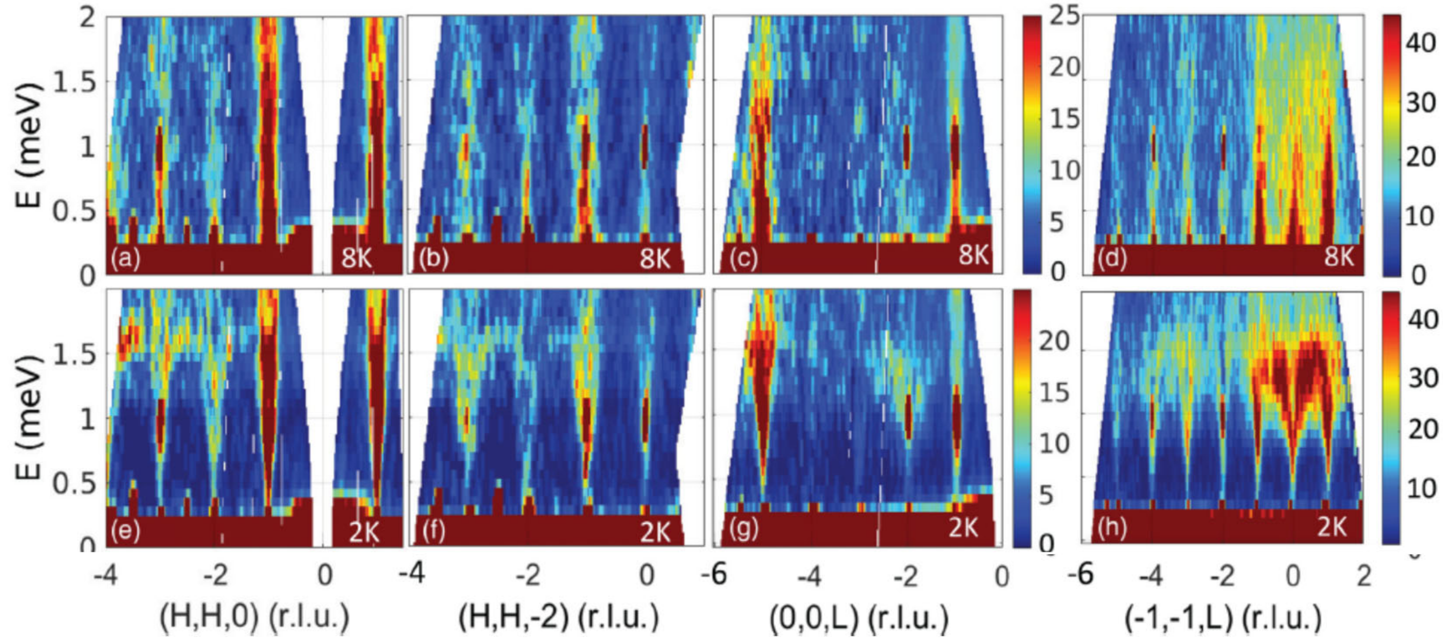


B. Lake; Budapest, **Spin chain physics – dominant AFM J_3**

Low Energy magnetic excitations of $BaCuTe_2O_6$

T = 8.0K (>T_N)
odd integer *H*, *L* – spinon continuum

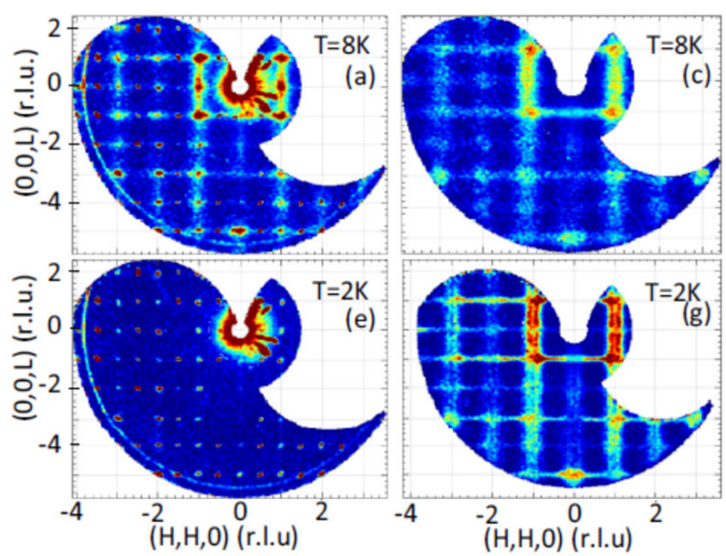
T = 1.8K (<<T_N)
Band at ≈1.6 meV, Spin-wave E<1 meV



T = 8.0K (>T_N)
Scattering streaks - uncoupled chains

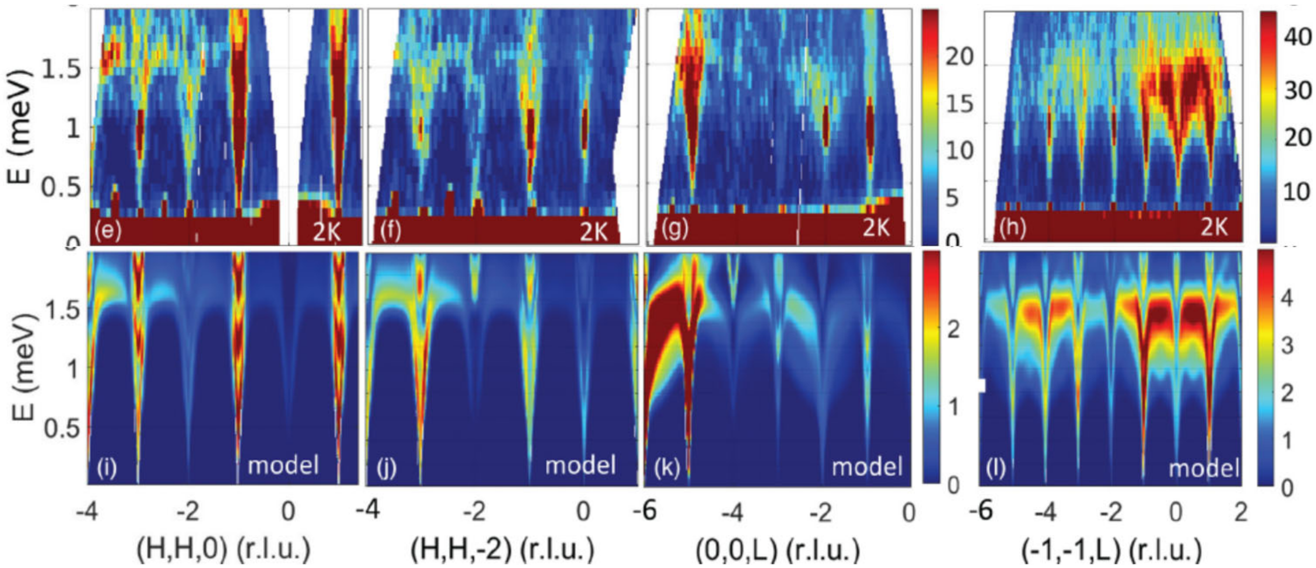
T =1.8K (<<T_N)
Scattering dots - Spin-wave dispersion

0.2<ΔE<0.6 meV 1.2<ΔE<1.6 meV



Confinement of spinon-pairs into spin-waves at low *E* and *T*

Hamiltonian of $BaCuTe_2O_6$

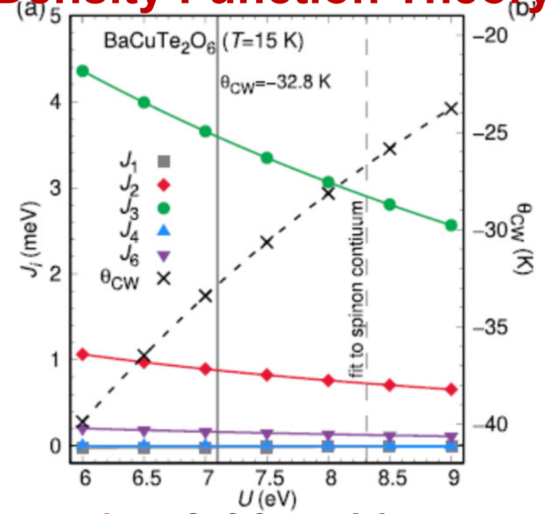


Data, T=1.8K

Spin-waves

J_3 = 2.90 meV, J_3'=4.56meV
J_2 = 0.30 meV,
J_1 = 0.05 meV,
J_6 = 0.03 meV

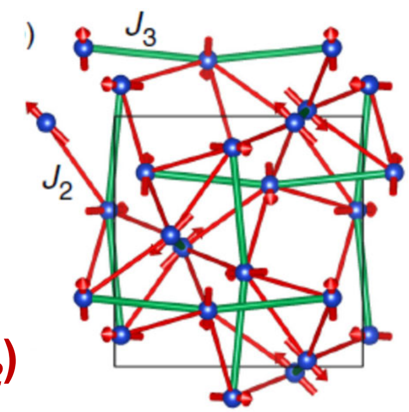
Density Function Theory



J_3 = 2.90 meV
0.44 < J_2 < 0.73 meV
J_1 = −0.02 meV
J_6 = 0.12 meV

Spin-wave theory, T=2K, E<2 meV
Fixed intrachain $J_3' = \pi J_3/2 = 4.56$ meV

J_3 = 2.90 meV AFM dominant
0.17< J_2 < 0.44 meV AFM hyperkagome subleading
J_1 = $2J_2$ - 0.54 meV, isolated triangle weak
J_6 undetermined



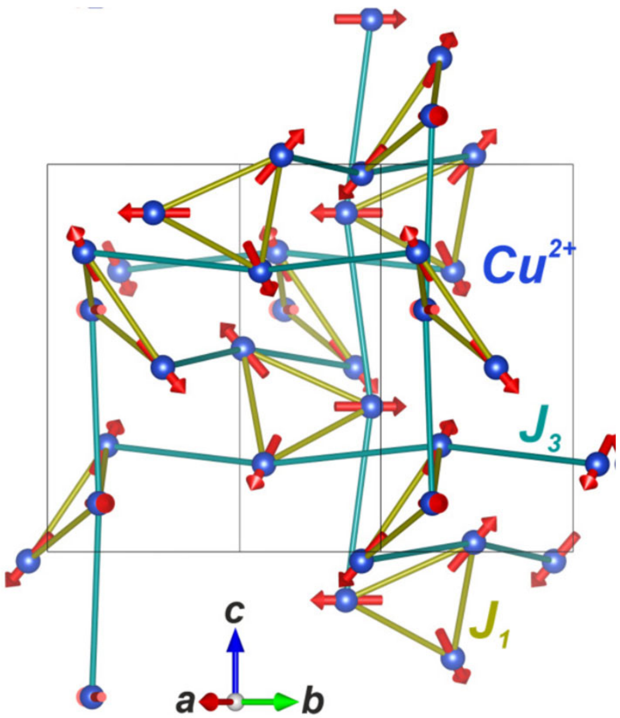
AFM spin chain // a, b, & c axes (J_3)
Weakly coupled by hyperkagome (J_2)

$SrCuTe_2O_6$

Magnetic structure of $SrCuTe_2O_6$

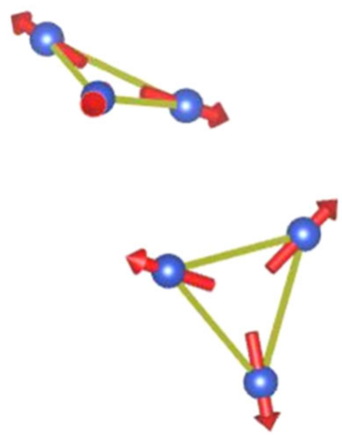
q=(0,0,1), m= 0.46 μ_B/Cu^{2+} compared to 1 μ_B/Cu^{2+}

$\Rightarrow$ low-dimensional / frustrated



180° order along J_3 chains $\Rightarrow$ J_3 is AFM

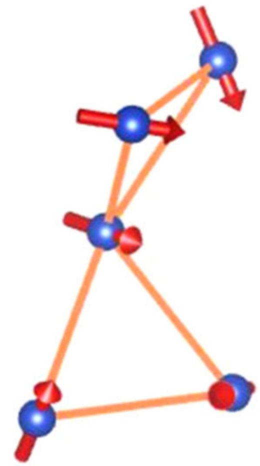
J_1 triangles planar 120°



isolated triangles

$\Rightarrow$ J_1 is AFM

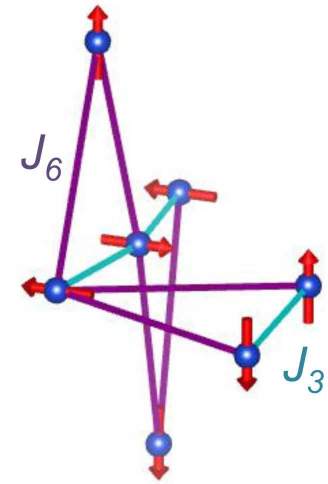
J_2 triangles canted 120°



hyperkagome triangles

$\Rightarrow$ J_2 is very weak

J_3 / J_6 perpendicular order between chains

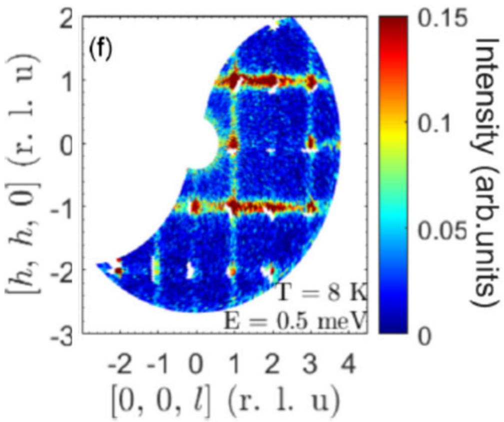


Neighboring parallel chains have 90° spins

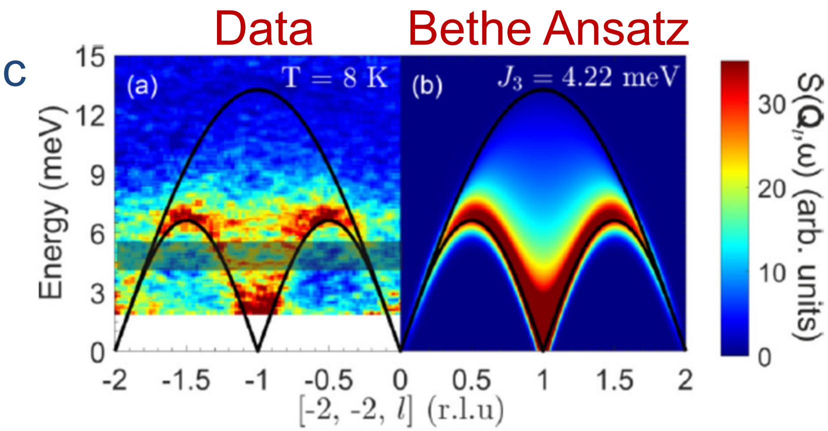
$\Rightarrow$ FM/AFM J_6 possible

Spin-1/2 Heisenberg AFM chains // a, b, c axes coupled by frustrated interchain interactions into a non-coplanar order

Hamiltonian of $SrCuTe_2O_6$



- Spin chains // a, b, c
- Spinon continuum
- $(\pi J_3/2)|\sin(\pi H)|$
- $\Rightarrow J_3 = 4.22$ meV



Spin-wave theory: T=2K, E<2 meV

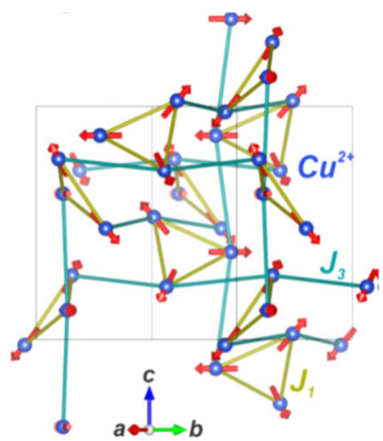
Fixed intrachain $J_3' = \pi J_3/2 = 6.62$ meV

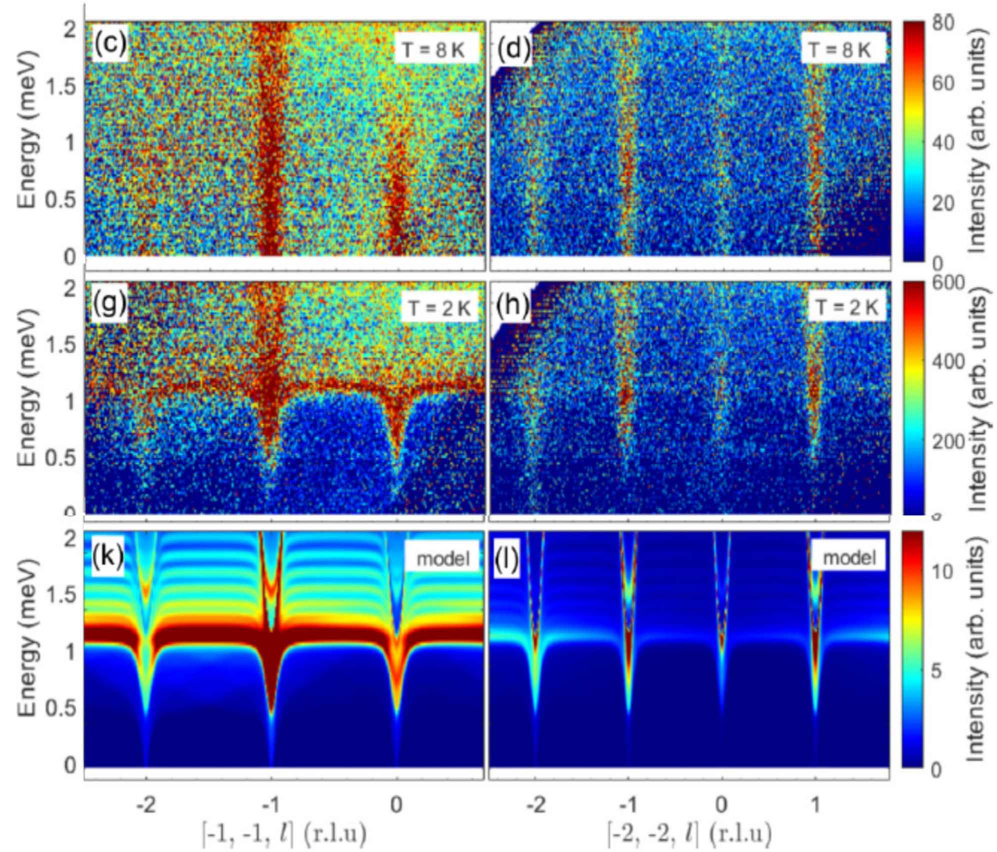
J_3 = *4.22* meV AFM dominant

J_2 −0.037 ± 0.012 meV very weak

J_1 = 0.17 ± 0.025 meV, AFM subleading

J_6 undetermined



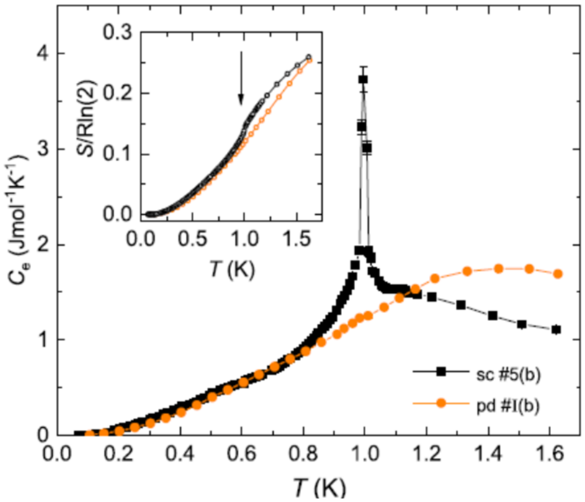


AFM spin chain // a, b, & c axes (J_3)

Weakly coupled by triangular (J_1)

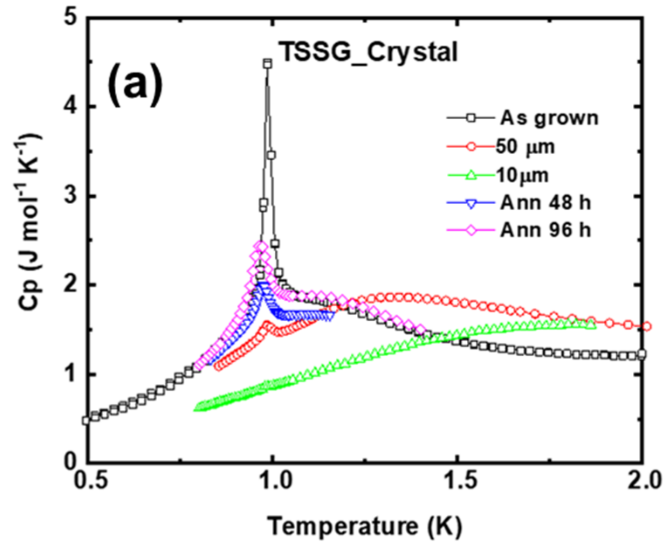
$PbCuTe_2O_6$

$PbCuTe_2O_6$ – Ferroelectric transition

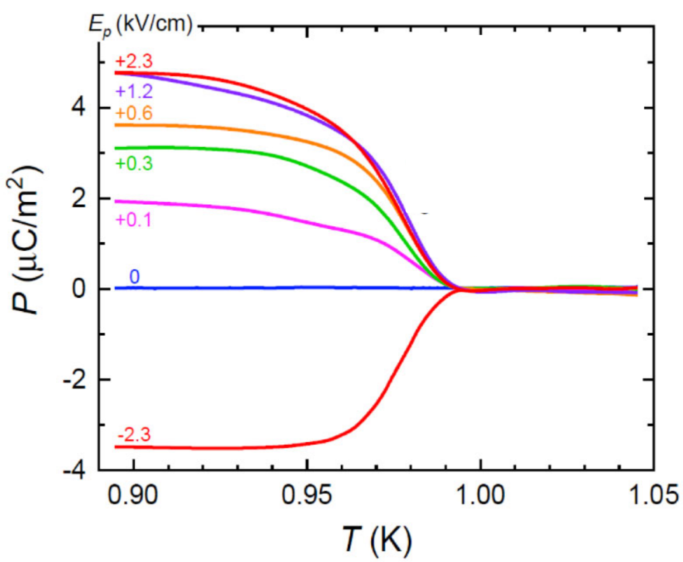


Transition Tc =1.1K

Present in crystal
but absent in powder



A.R. N. Hanna, A. T. M. N. Islam, R. Feyerherm, K. Siemensmeyer, K. Karmakar, S. Chillal, B. Lake Phys. Rev. Mat. 5 113401 (2021)



- Electric field applied on cooling - monodomain sample
- Electric field switched off Polarization measured on heating
- Clear spontaneous polarization below T_{FE}.

Ch. Thurn, P. Eibisch, A. Ata, M. Winkler, P. Lunkenheimer, I. Kézsmárki, U. Tutsch, Y. Saito, S. Hartmann, J. Zimmermann, A. R. N. Hanna, A. T. M. N. Islam, S. Chillal, B. Lake, B. Wolf, M. Lang npj Quantum Materials 6 95 (2021)

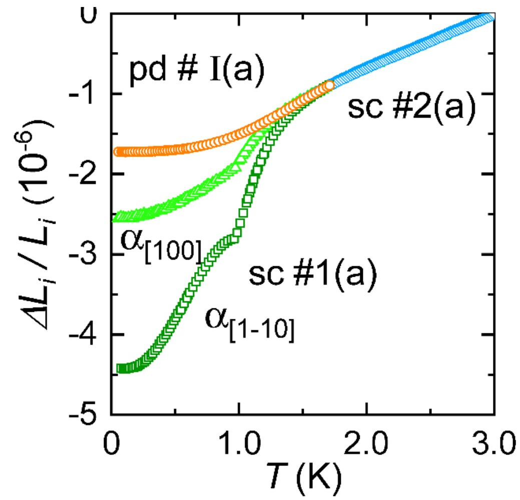
The transition is ferroelectric

$PbCuTe_2O_6$ – Structural & magnetic changes at T_{FE}

- Thermal expansion reveals structural distortion at T_{FE} to non-cubic phase.
- However no distortion observed by neutron diffraction (HRPT, PSI and WISH, ISIS)

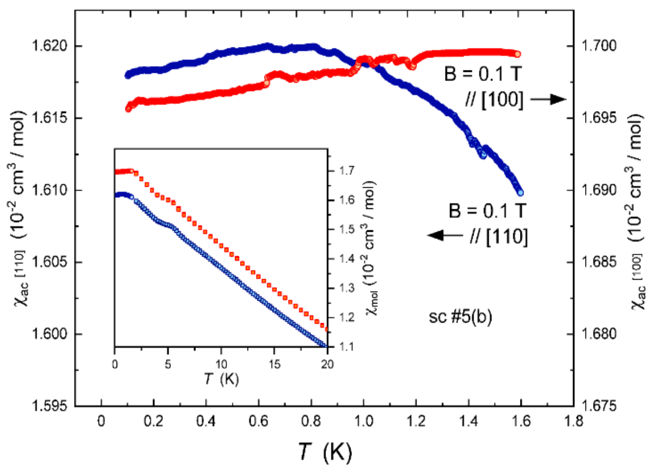
Ferroelectric transition accompanied by weak structural distortion

Symmetry lowering might lift frustration & induce magnetic order



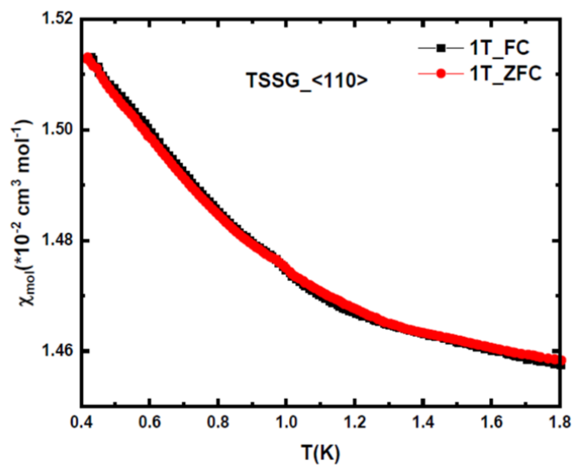
Ch. Thurn, npj Quantum Materials 6 95 (2021)

AC susceptibility – no transition



Ch. Thurn, npj Quantum Materials 6 95 (2021)

DC susceptibility – very weak signature



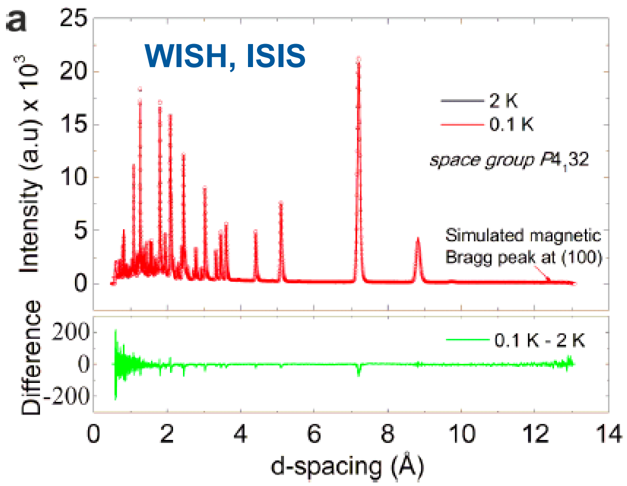
A.R. N. Hanna, Phys. Rev. Mat. 5 113401 (2021)

No evidence of a magnetic transition

$PbCuTe_2O_6$ – No observable magnetic order

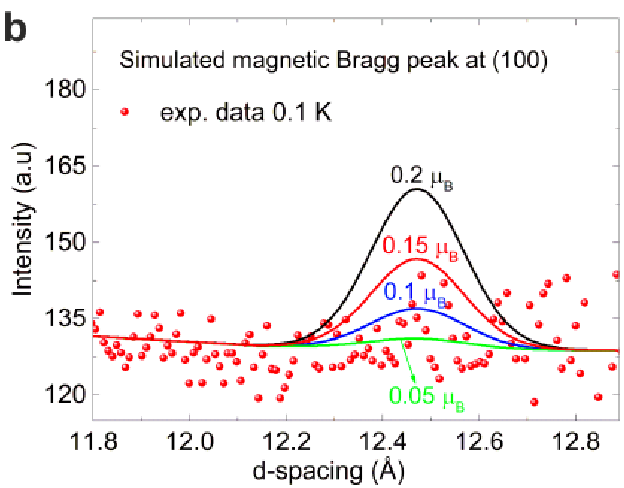
Neutron diffraction

No new Bragg peaks below T_{FE}=1K



- Different magnetic structures simulated

⇒ Magnetic order must be <0.05 μ_B



Muon Spectroscopy

- No oscillations down to 20mK

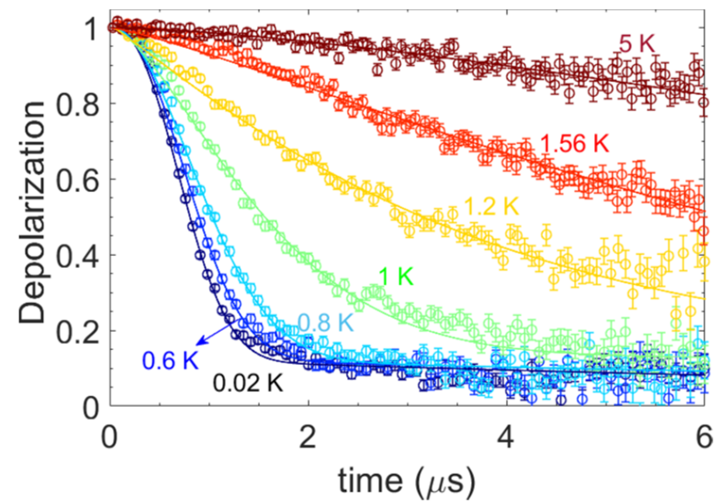
⇒ No long-range magnetic order

- Polarization at long times
  $P(t \rightarrow \infty,\ T \rightarrow 0) < 1/3$

⇒ No static magnetism, no spin glass

- Slowing of spin dynamics

Zero Field LTF, GPS, PSI

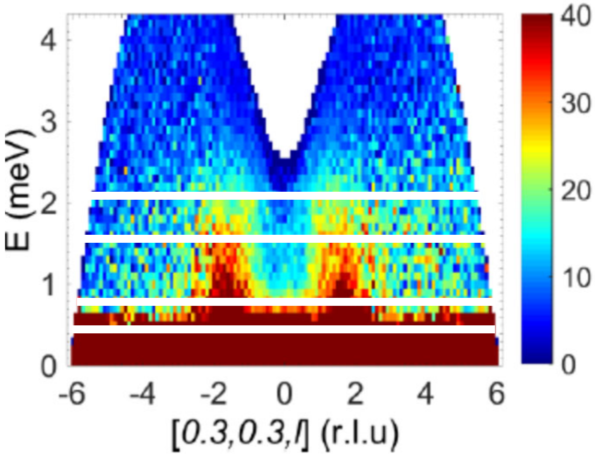


Long-range order and static magnetism is suppressed

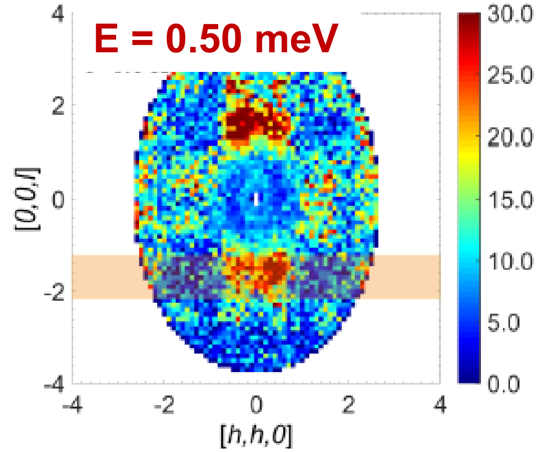
S. Chillal, Y. Iqbal, H.O. Jeschke, J.A. Rodriguez-Rivera, R. Bewley, D. Khalyavin, P Steffens, R. Thomale, A.T.M.N Islam, J. Reuther, B. Lake
Nature Communications 11 2348 (2020)

PbCuTe$_2$O$_6$ – Inelastic Neutron Scattering

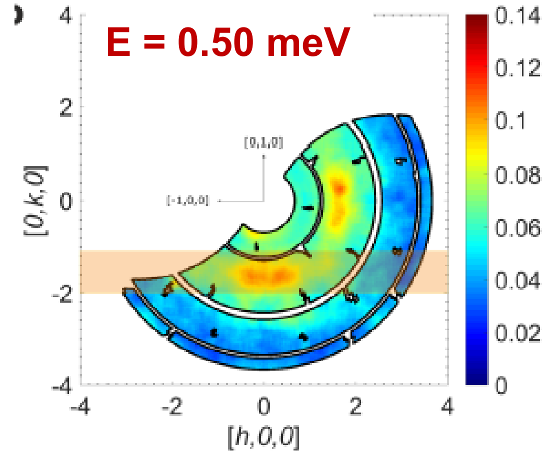
LET, ISIS; T=0.1K



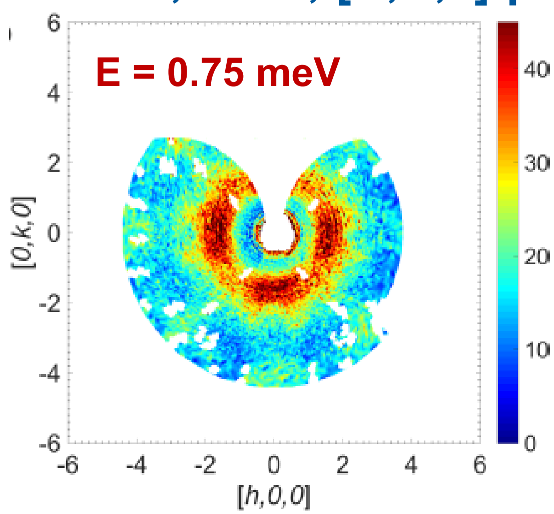
LET, ISIS; [H,H,L]-plane

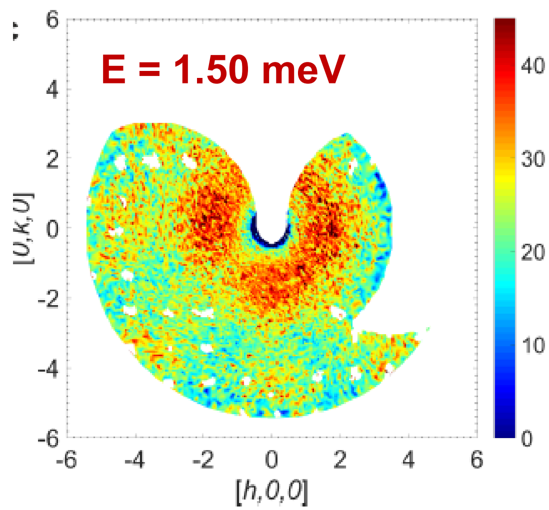


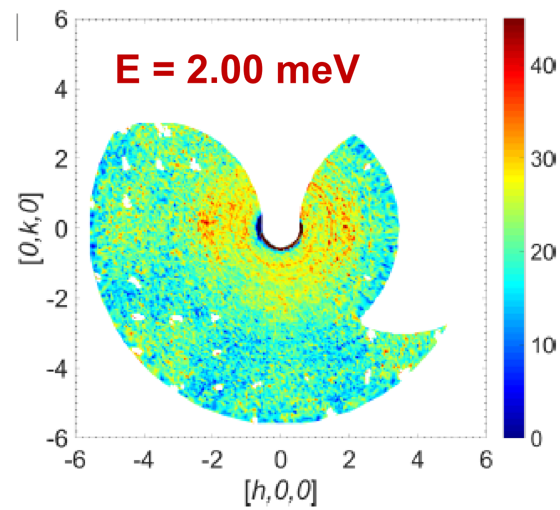
Thales, ILL; [H,K,0]-plane



MACS, NIST; [H,K,0]-plane T=0.1K







- For all energies diffuse sphere at Q=0.8Å^{-1}
- No sharp features, no spin-waves

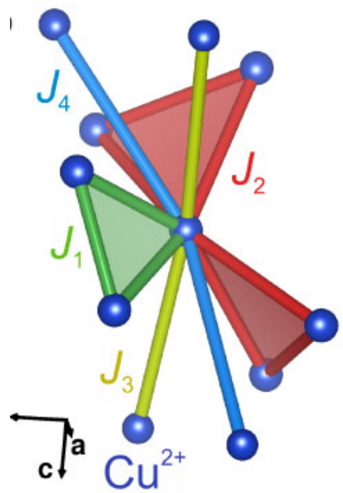
consistent with spinon excitations

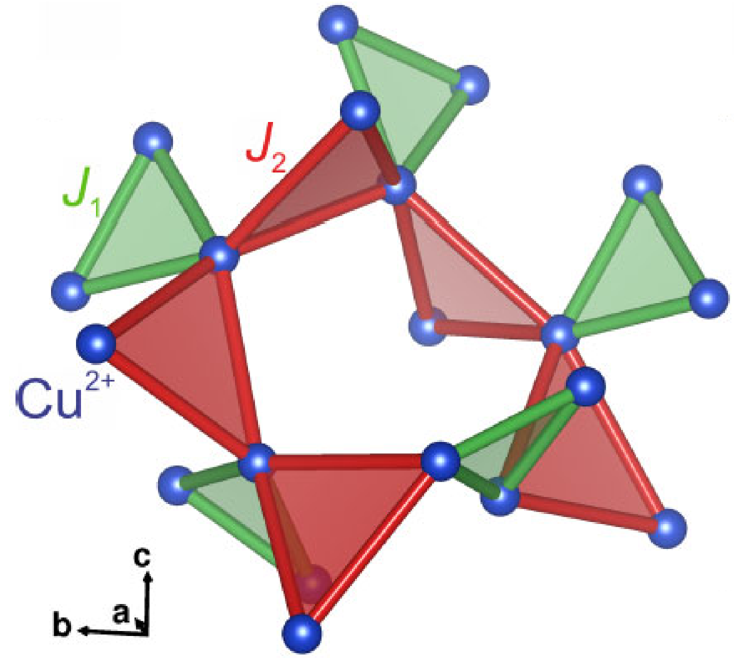
S. Chillal, Y. Iqbal, H. O. Jeschke, J.A. Rodriguez-Rivera, R. Bewley, D. Khalyavin, P. Steffens, R. Thomale, A.T.M.N Islam, J. Reuther, B. Lake Nature Communications 11 2348 (2020)

$PbCuTe_2O_6$ – Exchange interactions

Density Functional Theory (FPLO) (GGA)
Function of onsite potential U (5.5-8.0eV)
Hund's rule coupling J_H=1 eV

All antiferromagnetic
J_1 = 1.13 meV
J_2 = 1.07 meV
J_3 = 0.59 meV
J_4 = 0.12 meV
J_1:J_2:J_3:J_4=1:1:0.5:0.1





Harald Jeschke

- Distorted windmill lattice with **$J_1 \approx J_2$**
- 3D network of corner-sharing triangles
- 3 triangles per spin,
  - 2 hyperkagome J_2
  - 1 isolated triangle J_1

- **Strong frustrated interactions, $J_1 \approx J_2$**
- **Chain interaction J_3 is significant**

S. Chillal, Y. Iqbal, H. O. Jeschke, J.A. Rodriguez-Rivera, R. Bewley, D. Khalyavin, P Steffens, R. Thomale, A.T.M.N Islam, J. Reuther, B. Lake Nature Communications 11 2348 (2020)

$PbCuTe_2O_6$ – Comparison to theory PFFRG

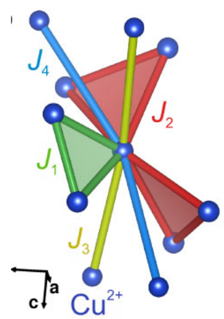
Pseudo-Fermion Functional Renormalisation Group

- Identifies long-range magnetic order
- Calculates spin-spin correlations

Johannes Reuther, Yasir Iqbal

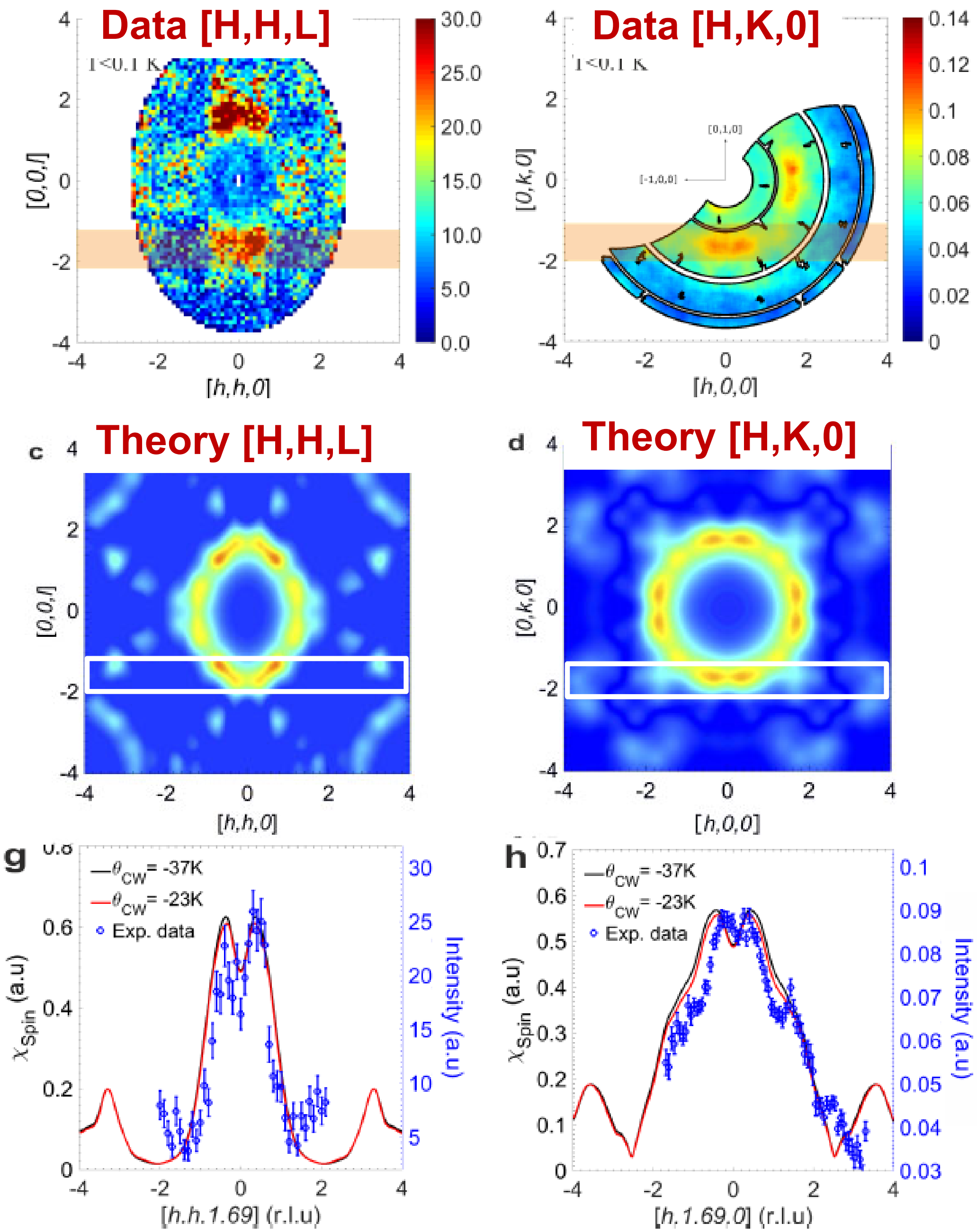
Using the Hamiltonian from DFT

All antiferromagnetic
J1 = 1.13 meV
J2 = 1.07 meV
J3 = 0.59 meV
J4 = 0.12 meV
J1:J2:J3:J4=1:1:0.5:0.1



PFFRG confirms
⇒ no long-range order
⇒ diffuse scattering pattern

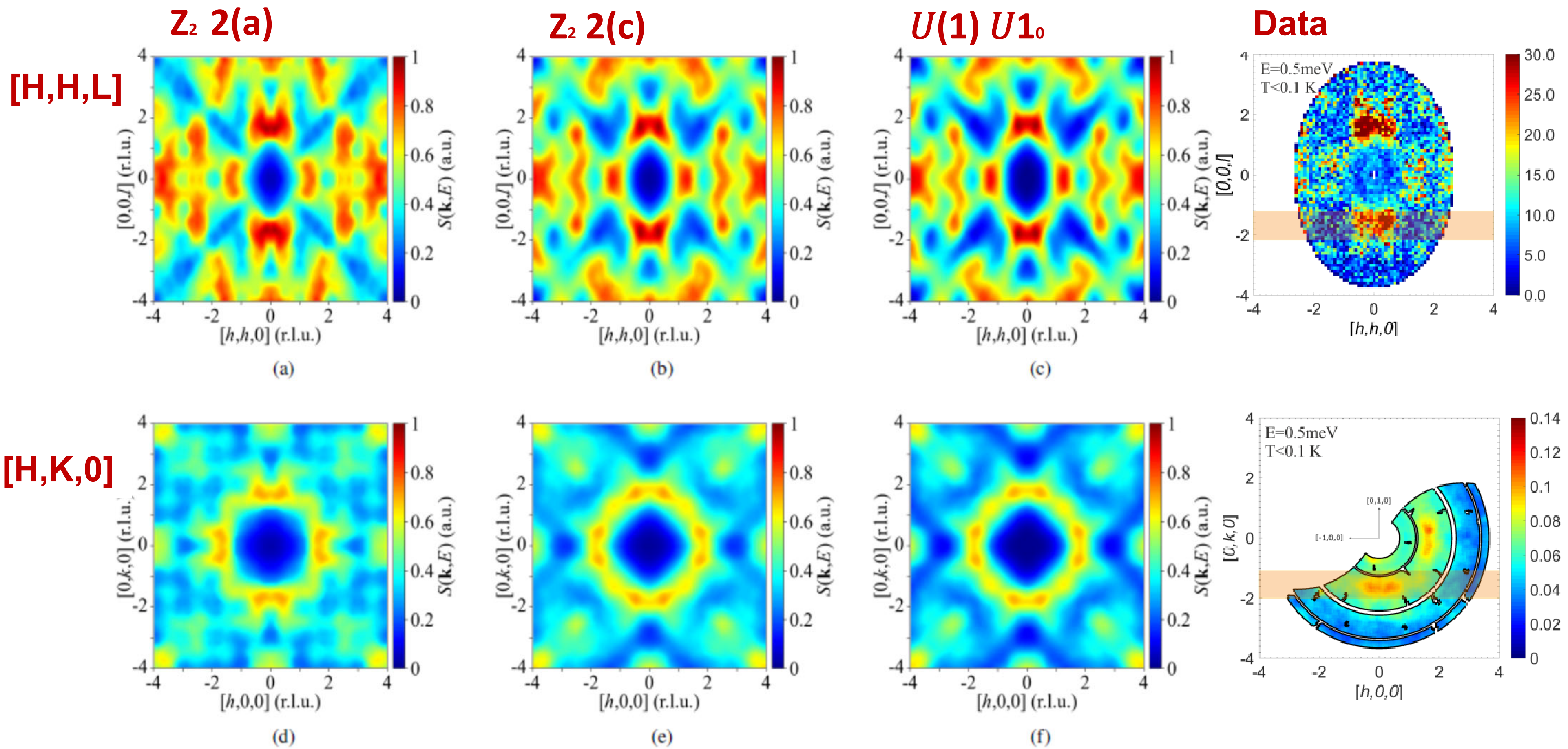
S. Chillal, Y. Iqbal, H.O. Jeschke, J.A. Rodriguez-Rivera, R. Bewley, D. Khalyavin, P Steffens, R. Thomale, A.T.M.N Islam, J. Reuther, B. Lake Nature Comm. 11 2348 (2020)



$PbCuTe_2O_6$ – Projective symmetry group analysis

- Parton construction and projective symmetry group
- identify 3 spin liquids compatible with lattice of $PbCuTe_2O_6$ (J_1-J_2 model)
  - 2 Z_2 spin liquids (gapped and ungapped)
  - 1 $U(1)$ spin liquid

Li Ern Chern and Yong Baek Kim
Phys. Rev. B 104, 094413 (2021)



General agreement with data, but difficult to distinguish between them

Summary

$P4_132$ with magnetic site 12d spans a range of geometries
spin chains, distorted windmill, hyperkagome and trillium

Explore the space group and possible magnetic lattices

$BaCuTe_2O_6$ – spin chains coupled by hyperkagome interaction

$SrCuTe_2O_6$ – spin chains coupled isolated triangles

$PbCuTe_2O_6$ – corner-sharing triangles 3 triangles at each site
– spin liquid candidate

The $ACuTe_2O_6$ Teams

HZB Helmholtz Zentrum Berlin

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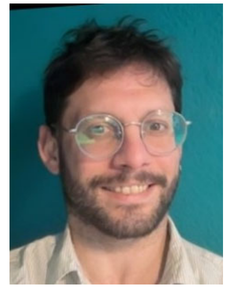
Koushik Karmakar
Konrad Siemensmeyer

Shravani Chillal

Abanoub Hanna



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