Overview of a Spin-Liquid Candidate, $Ba_3CuSb_2O_9$

Lara Thompson, MIT & KITP
Work done in collaboration with Patrick Lee

Massachusetts Institute of Technology

NSERC
CRSNG

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Zhou et. al., “**Spin liquid state in the S = 1/2 triangular lattice Ba3CuSb2O9**,” Phys. Rev. Lett. **106**, 147204 (2011)

- low T *linear* specific heat, C_m
- magnetic susceptibility, χ
- no magnetic order down to ~0.2 K

Nakatsuji et. al., “**Spin-Orbital Short-Range Order on a Honeycomb-Based Lattice**,” Science **336**, 559 (2012)

- dumbbell structure
- χ, C_m, inelastic scattering
- Jahn-Teller distortions

Balents, “**The Impact of Ionic Frustration on Electronic Order**,” Science **336**, 547 (2012)

- dumbbells save frustration (from orb ordering)

Quilliam et. al., “**Singlet Ground State of the Quantum Antiferromagnet Ba3CuSb2O9**,” Phys. Rev. Lett. **109**, 117203 (2012)

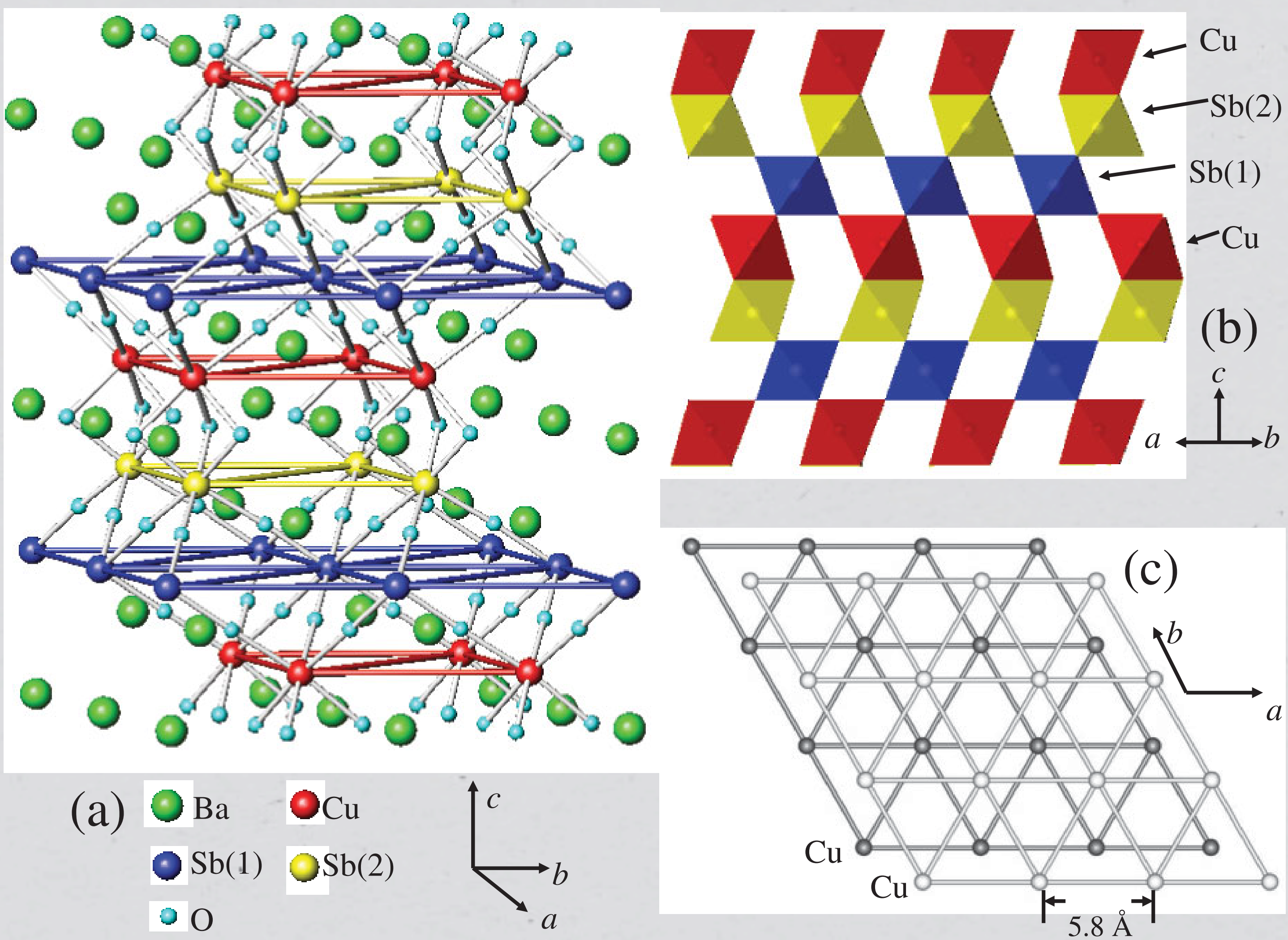
- NMR -> intrinsic χ
- relaxation $1/T_1$ ~ excitation gaps
- singlets and (intrinsic) ‘orphan spins’

Nasu, Ishihara, “**Dynamical Jahn-Teller Effect in Spin-Orbital Coupled System**,” arXiv: 1209.0239 [cond-mat]

- competing orbital/AFM orders ~> SO resonant states

$Ba_3CuSb_2O_9$ Structure

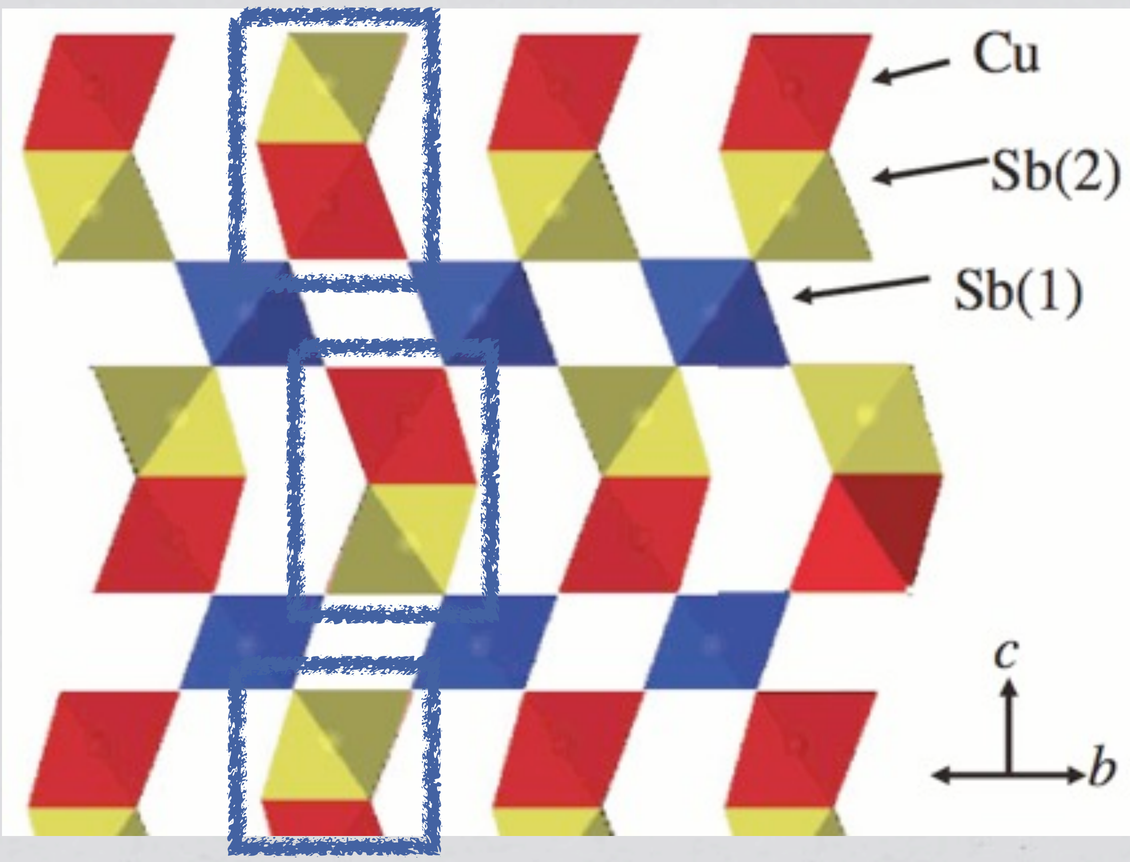
First, as proposed by Zhou et al:



- triangular layers of Cu^{2+}
- isolated by 2 non-magnetic Sb^{5+} layers
- Cu sits in an octahedron of O^{2-}: single hole is in degenerate e_g orbitals
- possible Jahn-Teller distortions may split the e_g orbitals

$Ba_3CuSb_2O_9$ Structure, cont'd

BUT! Cu^{2+}/Sb^{5+} 'dumbbells' are electric dipoles
Nakatsuji et. al. measure no (discernible) pyroelectricity

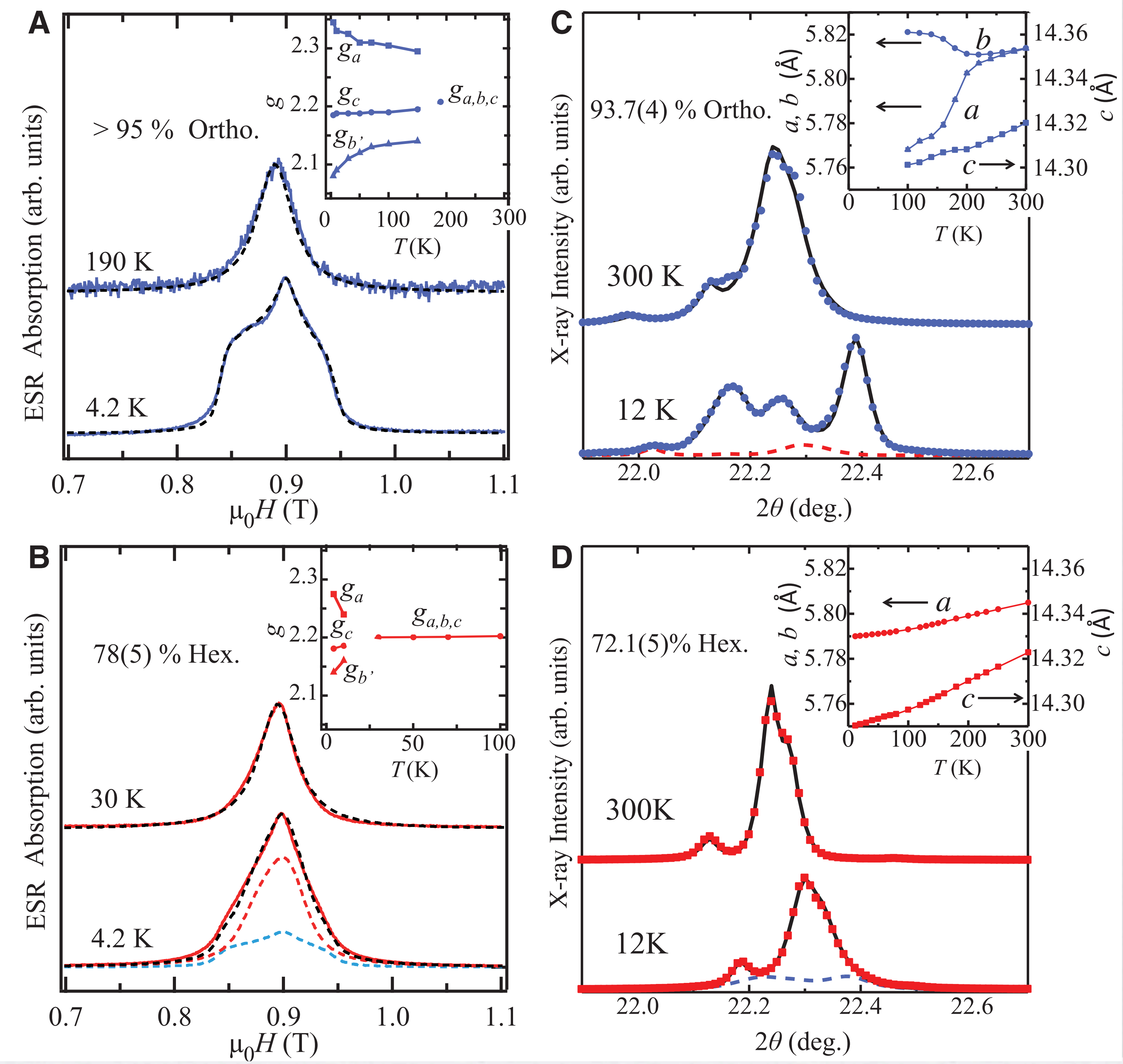


- Cu^{2+}/Sb^{5+} dumbbells ~ Ising 'spins' on a triangular lattice: frustrated!
- are magnetic layers still isolated?
- what is the superexchange for same-plane vs out-of-plane Cu's?

Net electric dipoles should cancel.

Jahn-Teller distortions

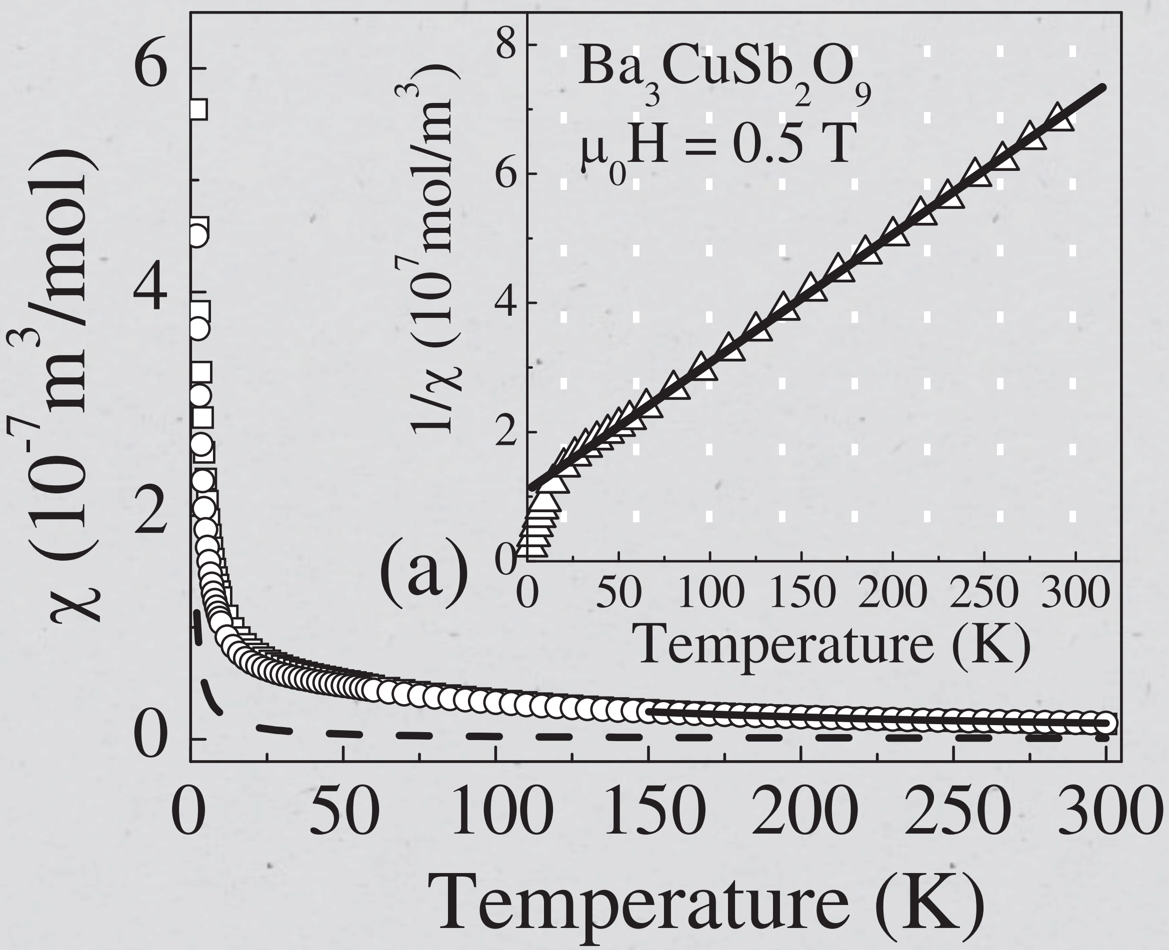
- for >8% off-stoichiometric Cu/Sb samples, find a 200K JT transition
- otherwise, JT 'remnants' <30K



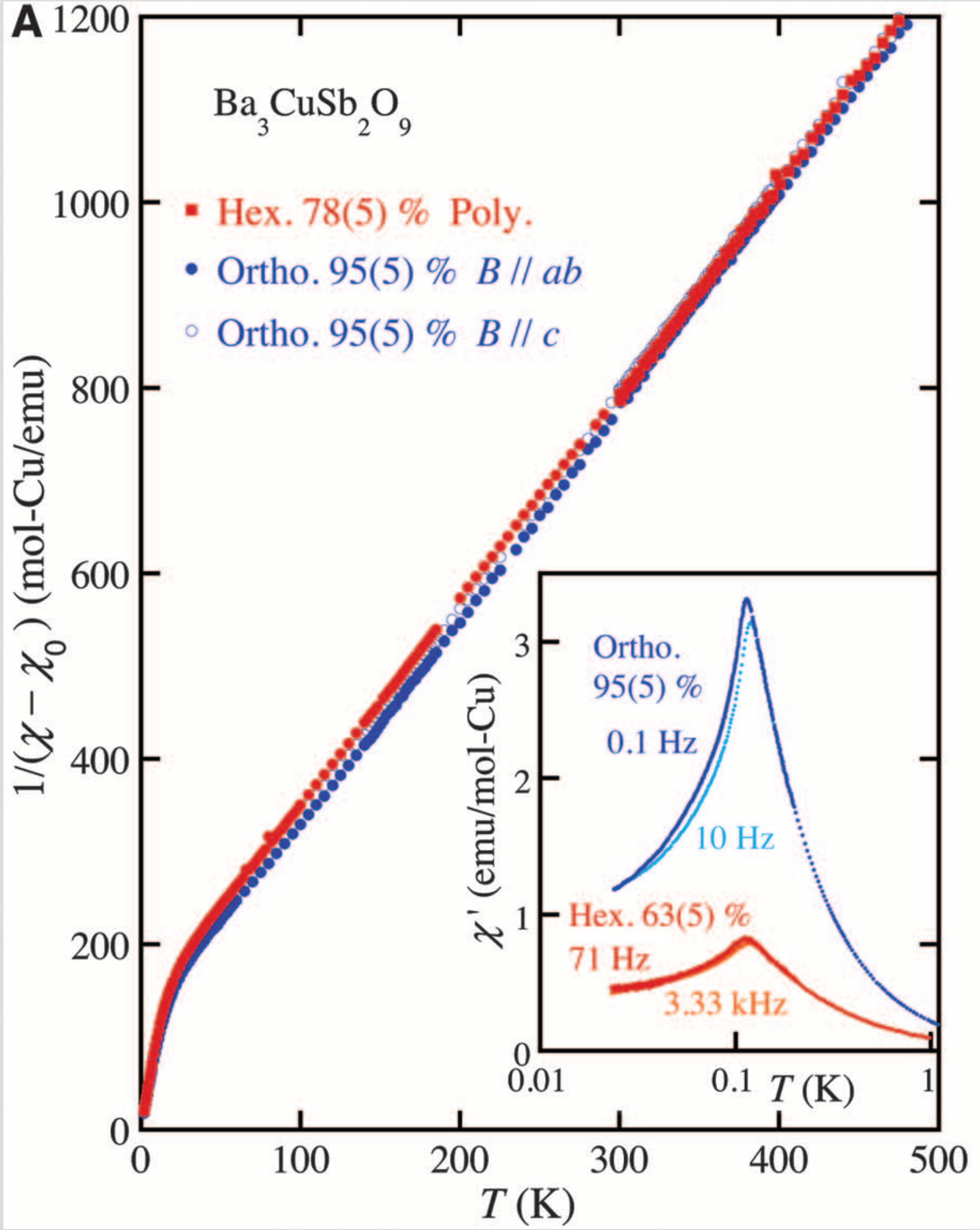
Nakatsuji et al (2012)

Magnetic Probes

- No magnetic ordering
- high T CW fit: θ_{CW} ~ -55K (afm)
- possible low T freezing of ortho phase orphan spins



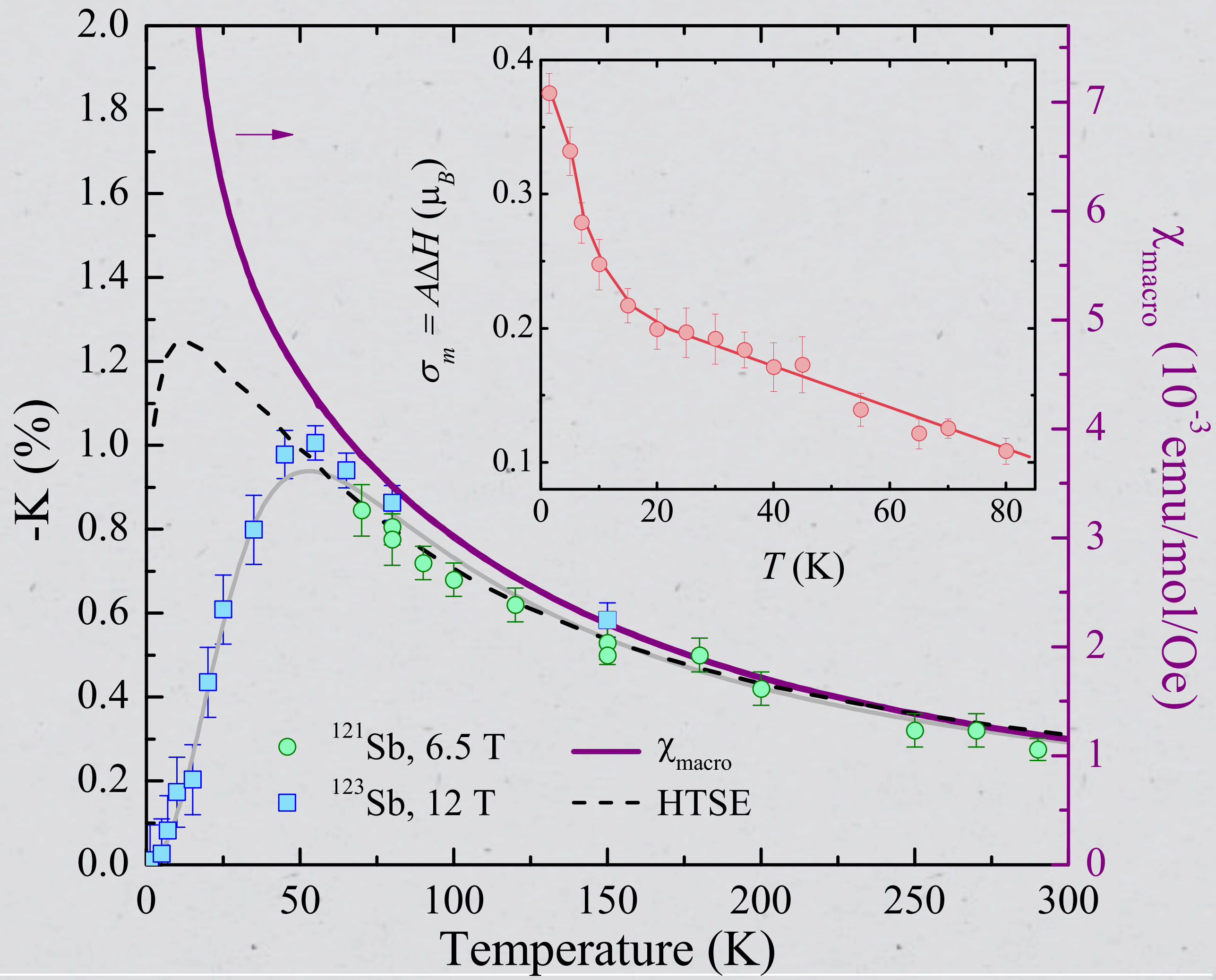
Zhou et al (2011)



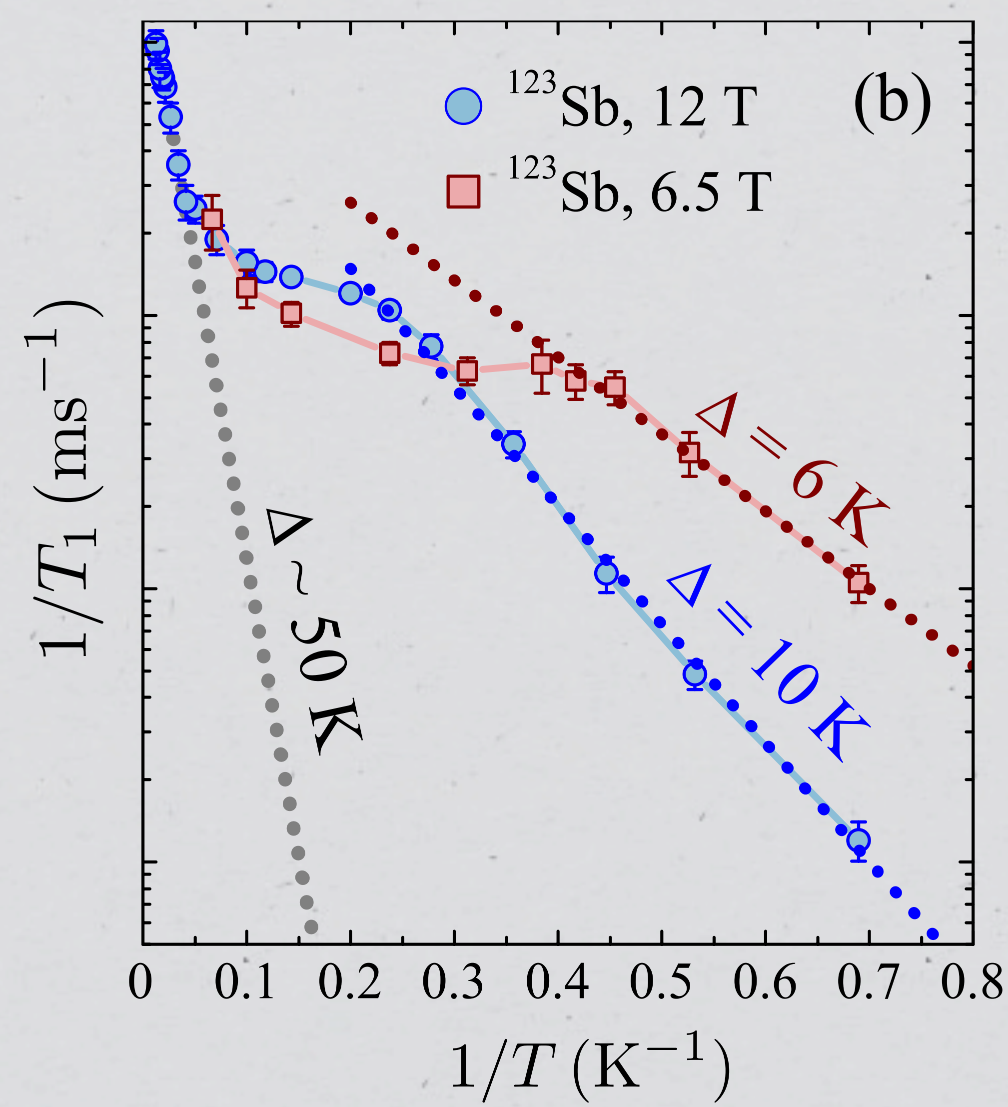
Nakatsuji et al (2012)

NMR (Quilliam et. al. 2012)

- ~singlet correlations at 50K (max) + diverging ‘orphan spins’
- field dep (distribution) of gaps at lower energy
- extreme line broadening at low T

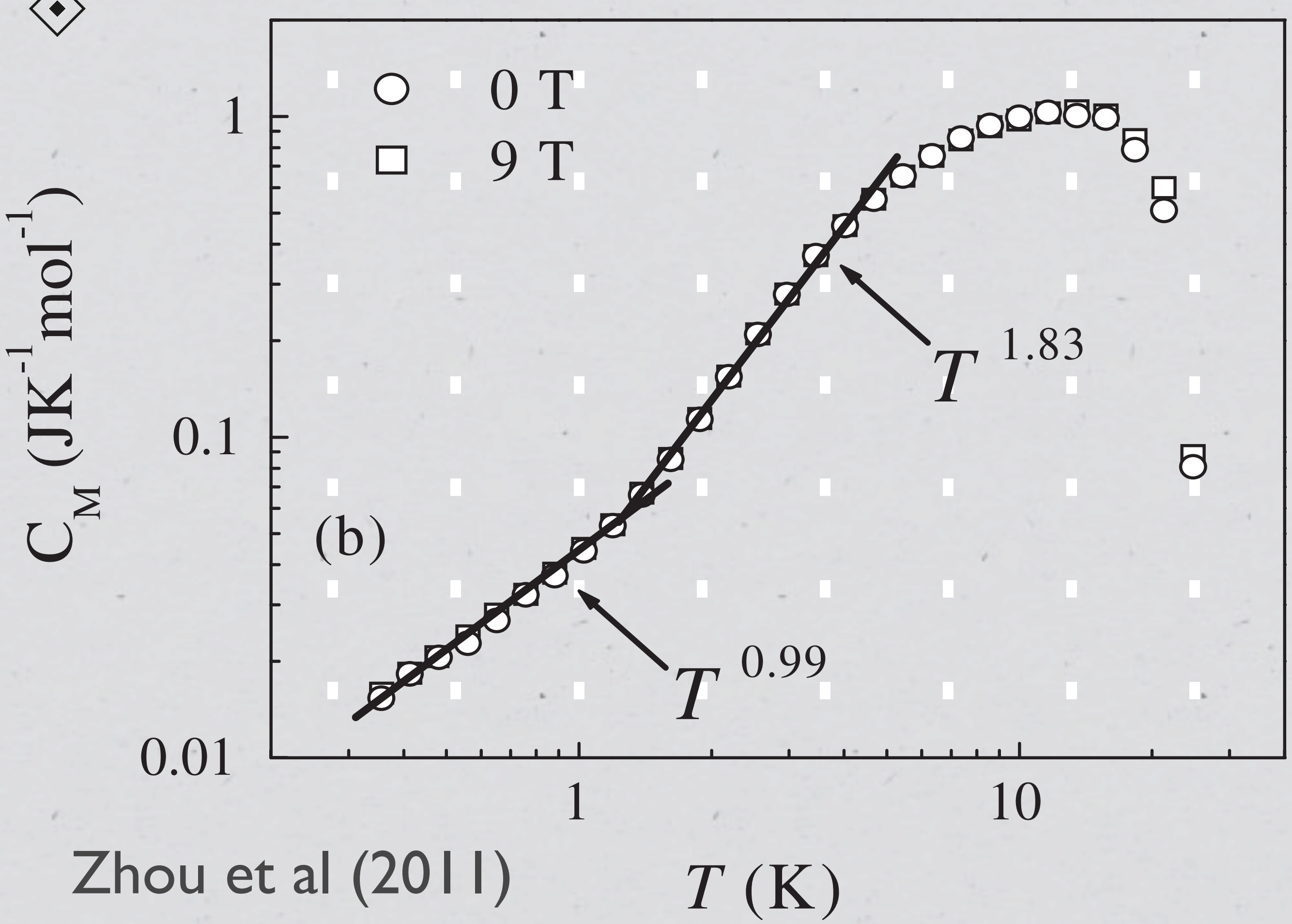


NMR lineshift, K ~ χ_{int}

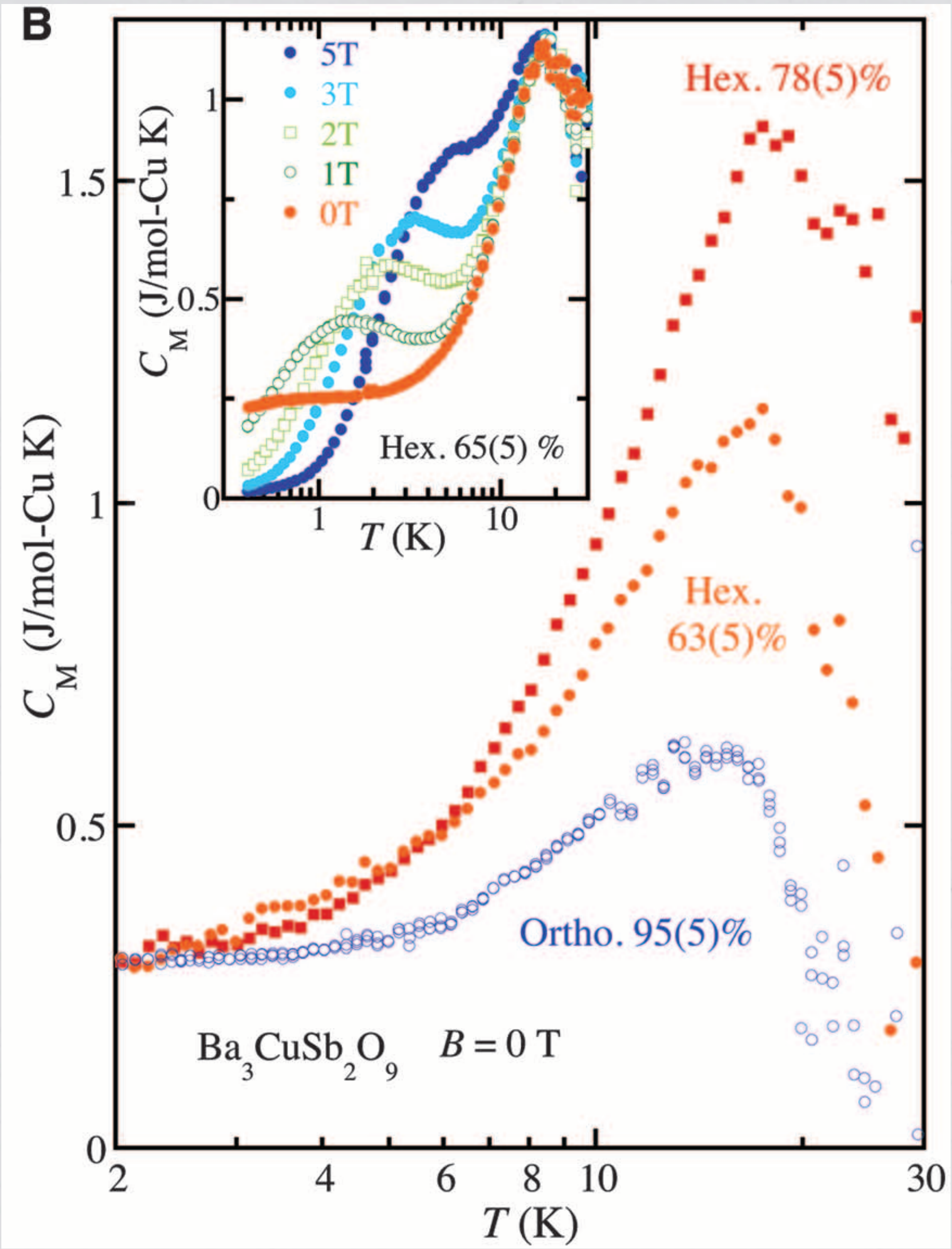


magnetic relaxation rate

Specific Heat



Zhou et al (2011)



- broad max around 10-30 K ≲ J/2
- more d.o.f. in hex phase
- local moments can be frozen in field

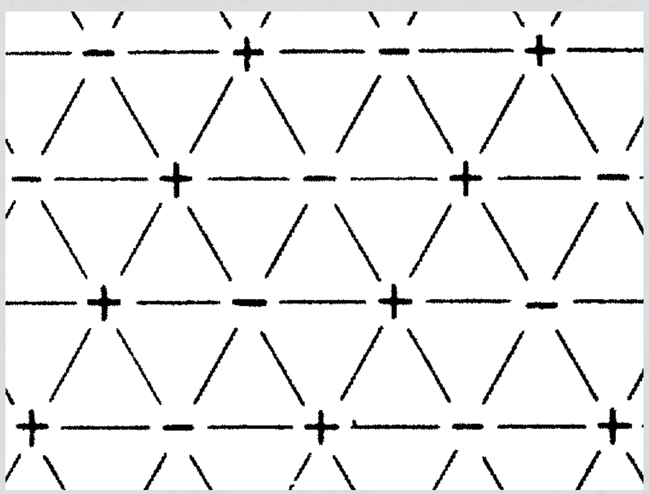
Nakatsuji et al (2012)

‘Dumbbell’ Order

- electric dipoles ‘dumbbells’ ~ Ising pseudospin; Hamiltonian:

$$H = \sum_{<ij>} J_{afm} s_i s_j - \sum_{<<ik>>} J_{fm} s_i s_k$$

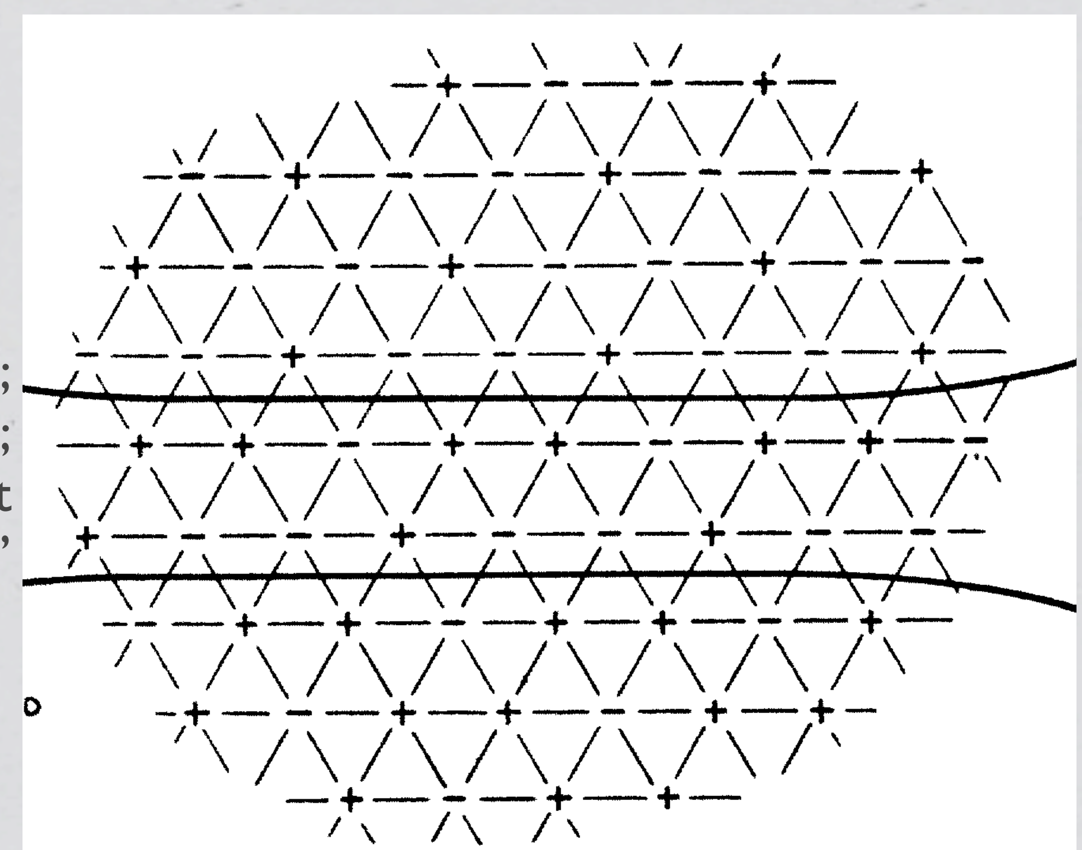
stripes



honeycomb

entropically favoured:

zero E domain walls; many domains; honeycomb-like, but rather ‘disordered’

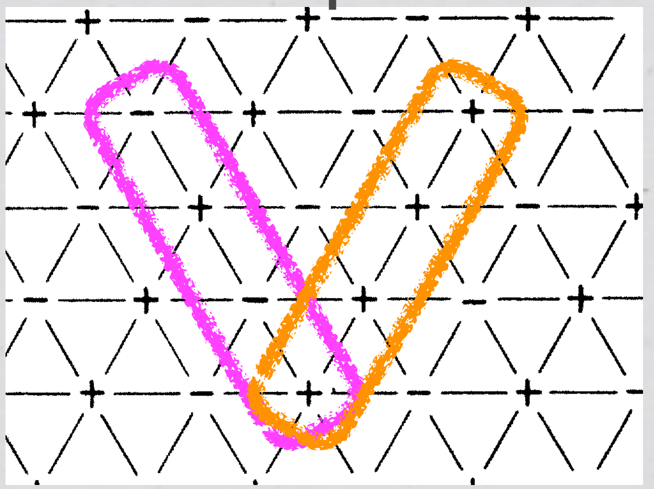


- agrees with “3 sub-lattice, ferrielectric short-range order” reported by Nakatsuji (2012)

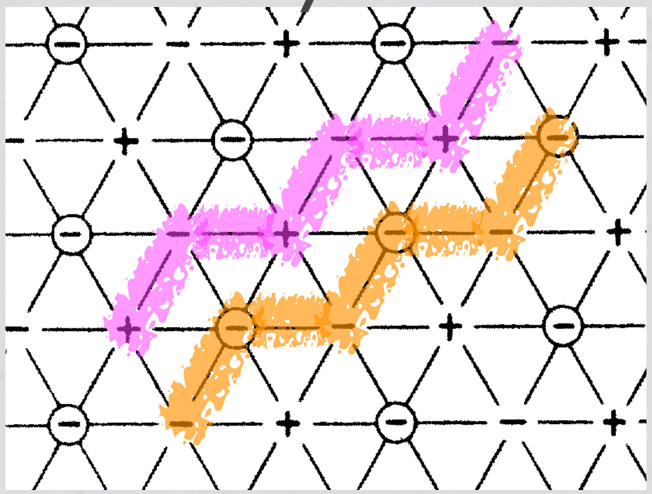
‘Dumbbell’ Defects

- off-stoichiometry of Cu/Sb translates to a vacancy in the effective Ising lattice
- ~1/9 extra/fewer Cu means 1 vacancy per hexagon; should consider defect correlation effects

stripes



honeycomb



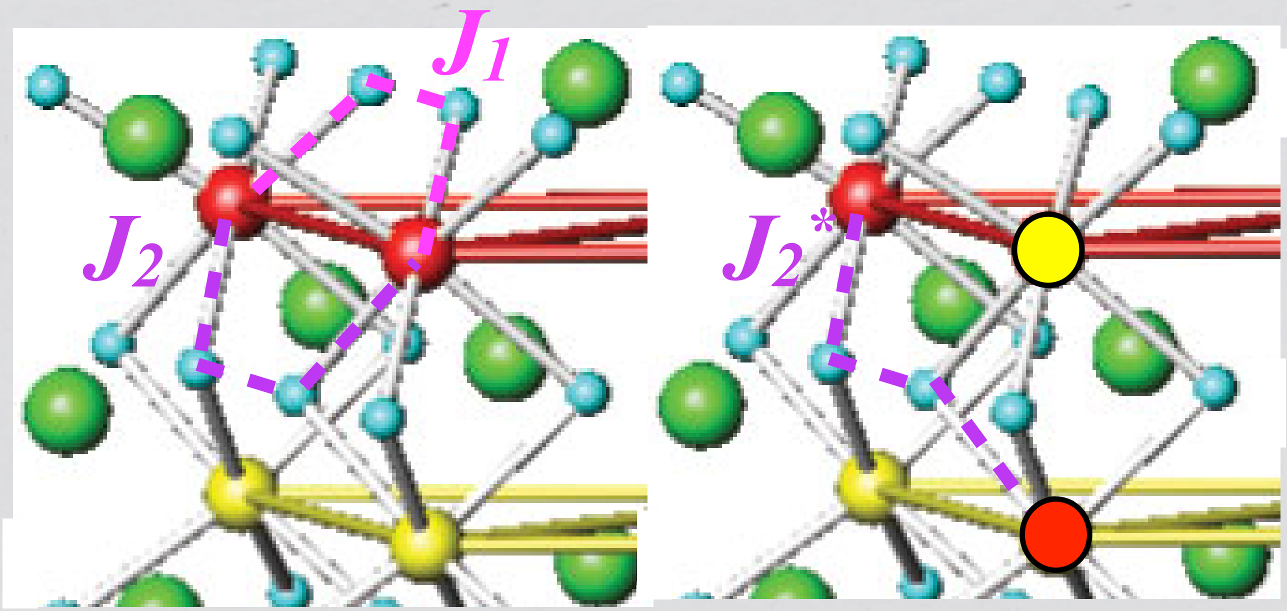
- single defects in honeycomb prefer maj. spin sites, ∴net polarization
- in stripes, net zero polarization

- minimum energy ‘defect line’ in stripes when:

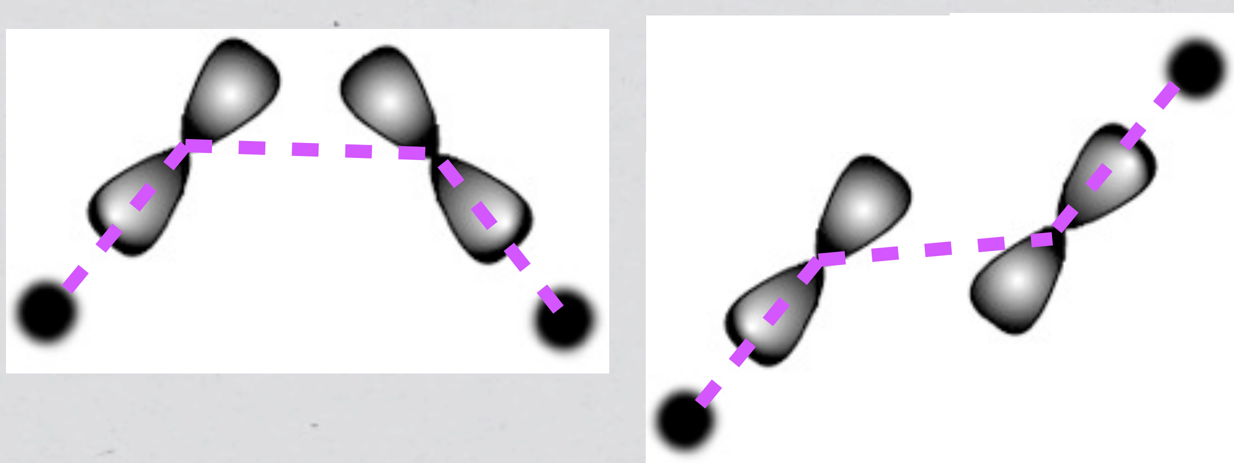
$$J_{afm} < nJ_{fm}$$

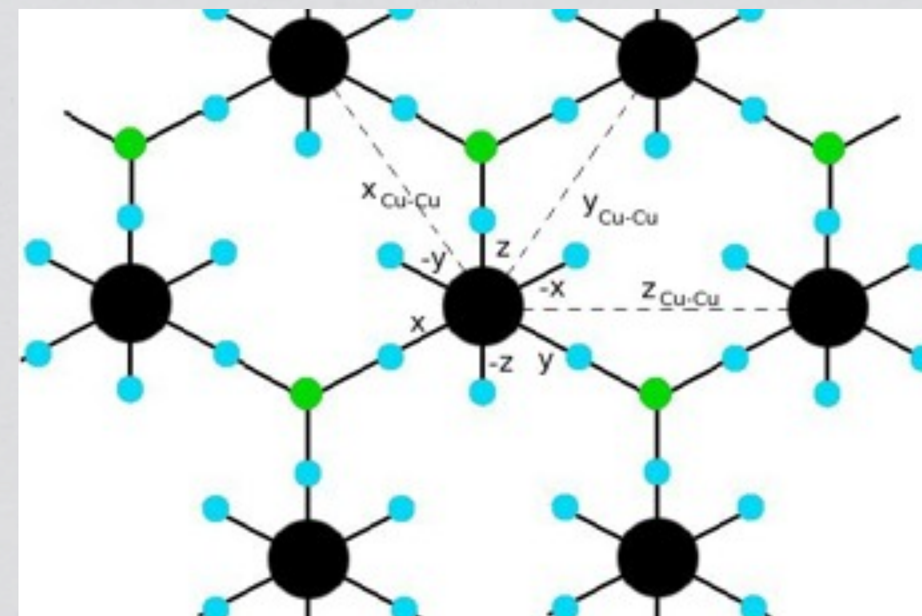
- with enough defects, expect a transition to (JT distorted) stripe phase

Cu-O-O-Cu Superexchange



parallel   anti-parallel

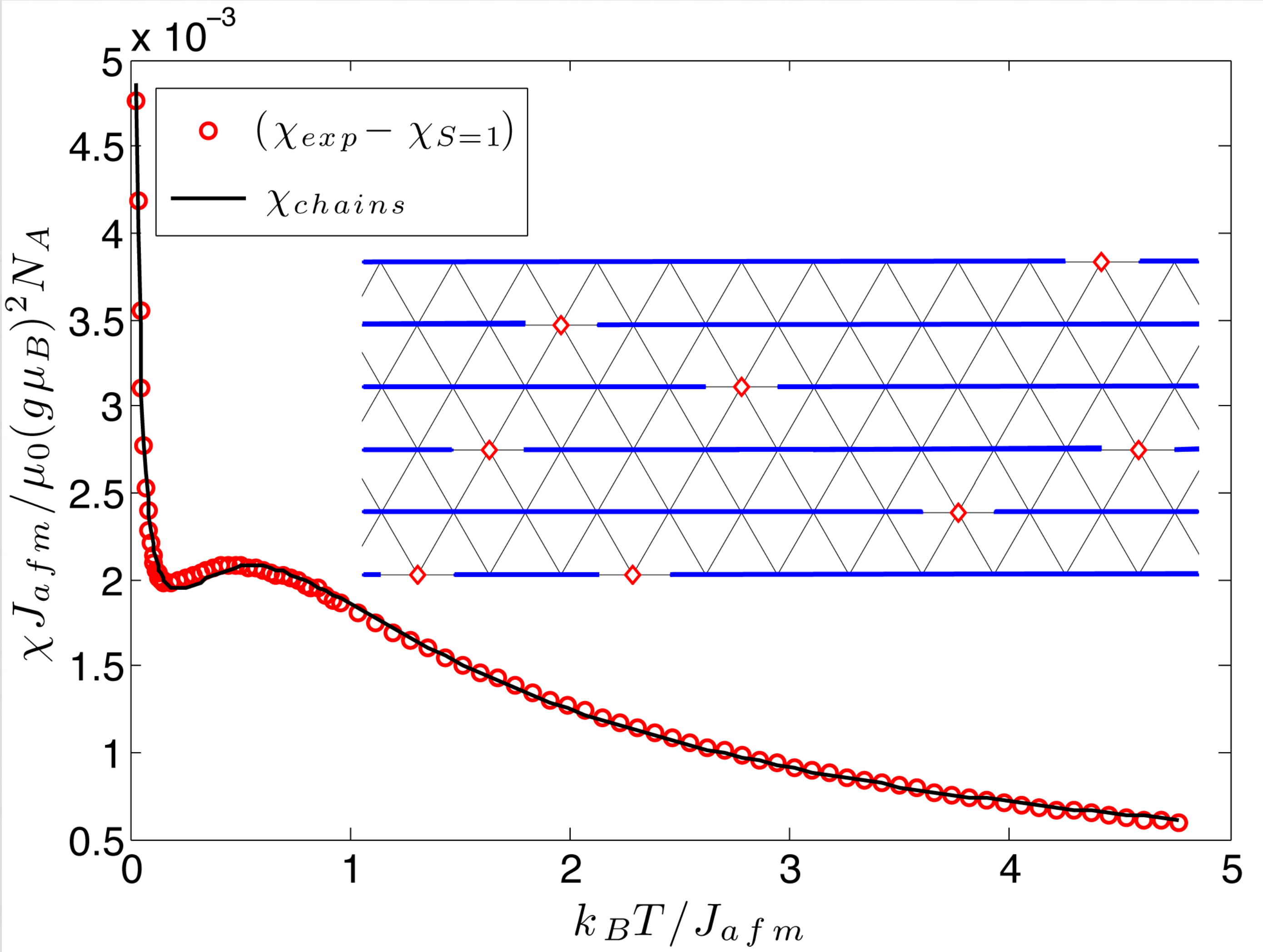




- Cu^{2+}/Sb^{5+} dumbbell
- O^{2-}
- Sb^{5+}: Sb(1) sites shown, Sb (2) sites sit directly under Cu sites

overlap is of *same order* and J_2^* (as opposed to arguments in Nakatsuji et. al.)

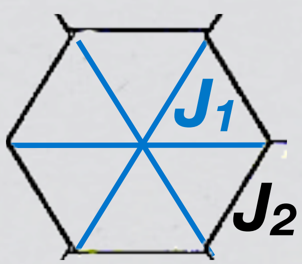
Spin chains in stripe order



arXiv:1202.5655 [cond-mat]

- previously motivated by a triangular lattice of Cu & defects
- MF favours a uniform JT distortion leading to decoupled chains
- remarkable agreement in χ with ~20% s=$^1/_2$ 'orphan spins'
- BUT, JT distortion is not always present: what about hex phase?

Disordered Honeycomb



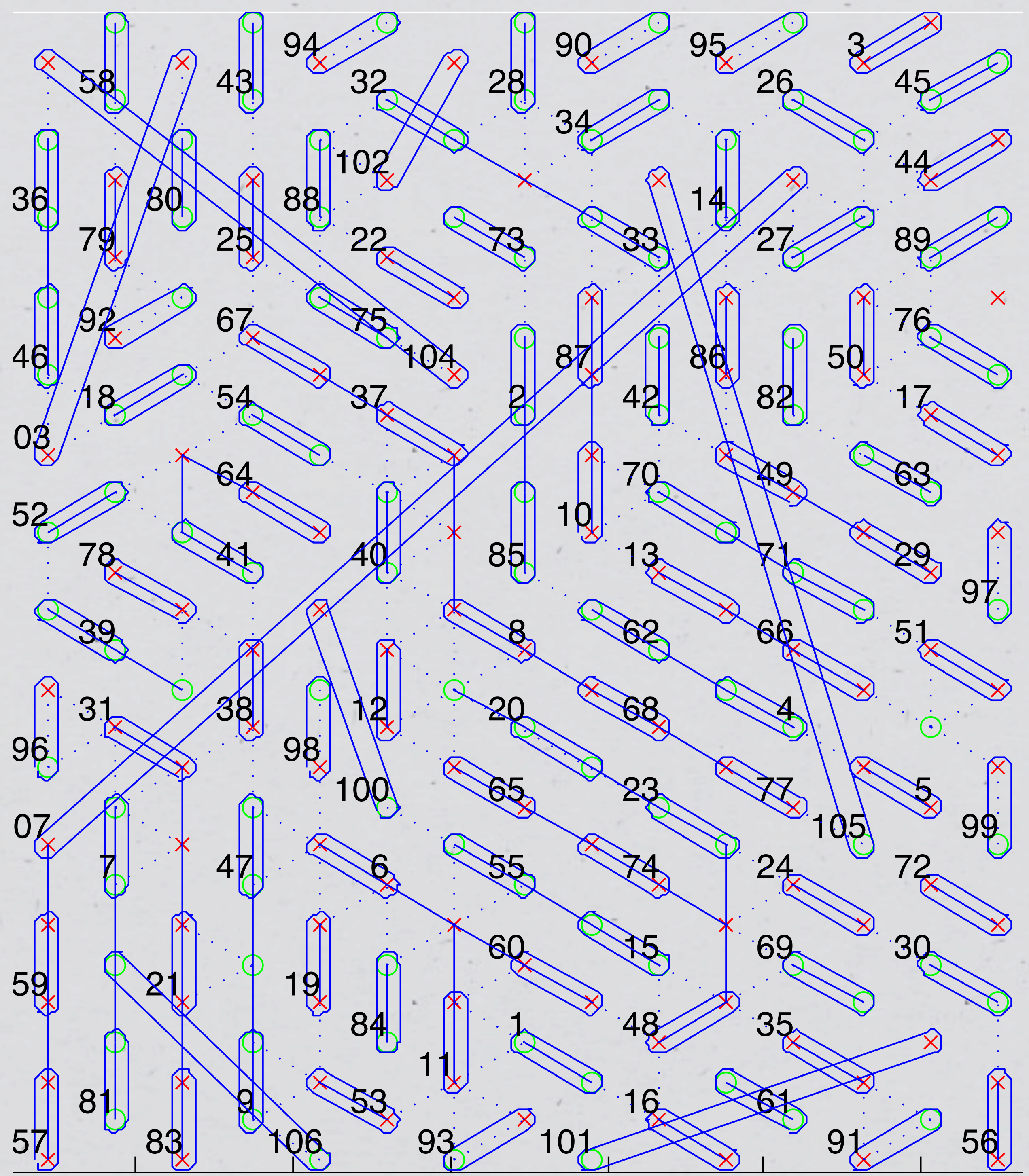
- with $J_1 \sim J_2/2$ *when orbitals are aligned*

- relative alignment of e_g orbitals far more important

- aligned orbitals naturally decouple rows; relieve frustration

- even in disordered phase, expect (zigzag) spin chains

example output of (unoptimized) singlet pairing ➤



$Ba_3CuSb_2O_9$ Summary

- No magnetic order.
- typical afm exchange of ~50-60 K
- broad peak in χ_{int}, C_m: singlets, spin chains?
- ~15-20% ‘orphan spins’ (intrinsic?)
- dumbbell order of in- and out-of-plane Cu on a triangular lattice ~ disordered honeycomb
- JT distorted in off-stoichiometry samples; remnant JT at low T in hex phase