Finite Temperature and Ground State Properties of Kagome Antiferromagnets

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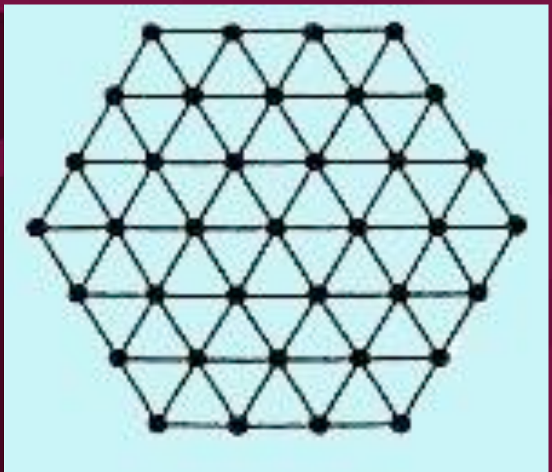
M Rigol, RRPS PRL 98, 207204 + PRB 76, 184403 (2007)
RRPS, D. A. Huse PRB RC (2007)

OUTLINE

- INTRODUCTION
- Numerical Results at T=0 (ED, Series)
- Dimer Expansions: VBC order
- Experiments--Herbertsmithites
- Calculations at finite T (ED,HTE,NLC)
- Discussions and Summary

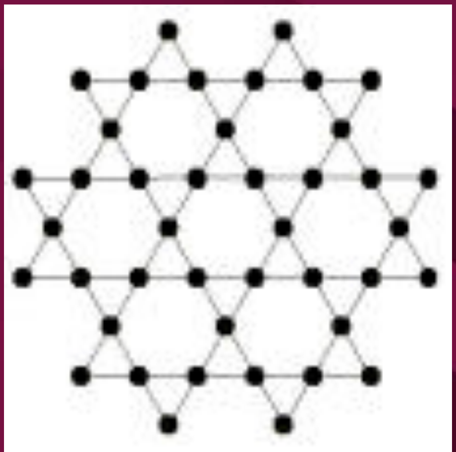
Triangular-Kagome Lattice Magnets

Triangular-Lattice:
Edge sharing triangles



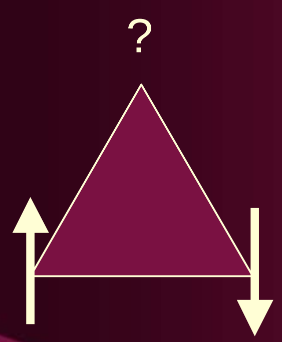
Kagome-Lattice:

Corner sharing triangles



Site-depletion makes Kagome-Lattice more frustrated

Classic example of Frustration
Ising Model



A Triangle: 6 out of 8 states are ground states)

uud udu duu udd dud ddu have same energy

uuu ddd have higher energy

Lattice Models are Exactly Soluble

TLM: T=0 critical point

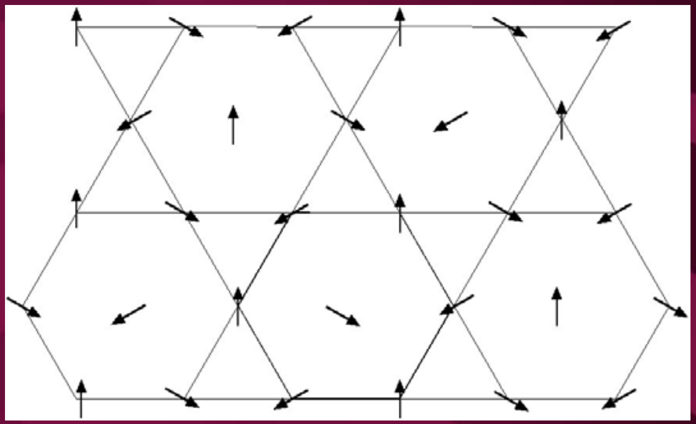
Ground state entropy under 50% of total entropy

KLM: Finite (short) correlation length even at T=0

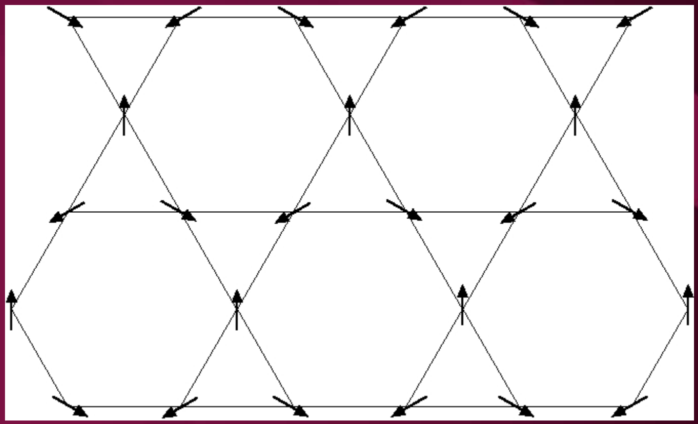
Ground state entropy about 72% of total entropy

Classical Heisenberg Models

- Ground state has 120 degree structure
- TLM: Unique Ground State (apart from symmetry) (Fully Constrained)
- KLM: Finite ground state entropy (see TLM) (Underconstrained)
- **Order by Disorder**



TLM



Q=0

Quantum Heisenberg Model

$$H = \sum_{<ij>} J_{ij} \, \hat{\boldsymbol{S}}_i \cdot \hat{\boldsymbol{S}}_j$$

$$J_{ij} > 0$$

$$[\hat{S}_j^x, \hat{S}_j^y] = i\hbar \, \hat{S}_j^z$$

$$\hat{\boldsymbol{S}}_i^2 = S(S+1)\,\hbar^2$$

Spin is a good quantum number

Most interest in spin-half case

Pair of spins like to form rotaionally invariant singlets –entangled state

Many Open Questions

$$H = \sum_{<ij>} J_{ij} \, \hat{\boldsymbol{S}}_i \cdot \hat{\boldsymbol{S}}_j$$

$$J_{ij} > 0$$

- Is Ground state magnetically ordered? SSB
- Is the ground state a VBC?
- Is there a Quantum Spin-Liquid? RVB
- Is there a spin-gap?
- Is there algebraic spin order?
- Are there fractional-spin excitations? FQHE
- Are there massless Dirac spinons?

Magnetic Long Range Order

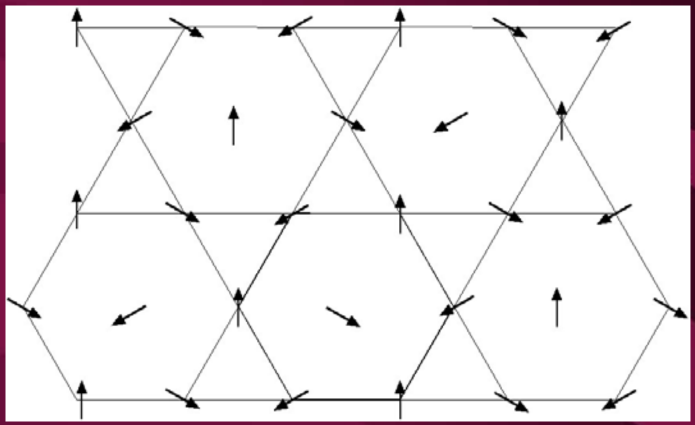
Many Candidates

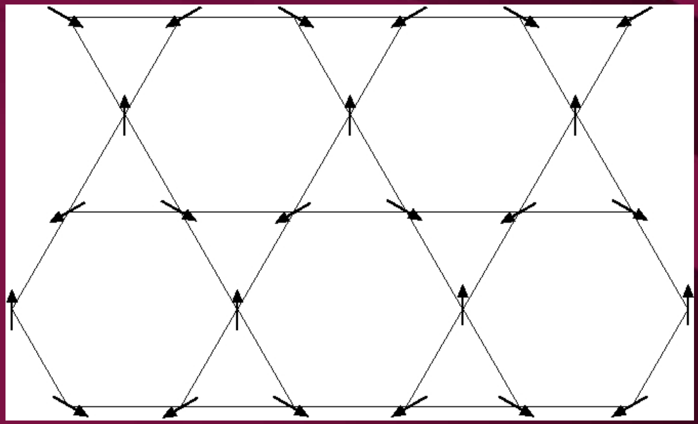
TLM [root(3)by root(3)]

Q=0

Doubled Unit Cell along Y

Answer appears to be NO



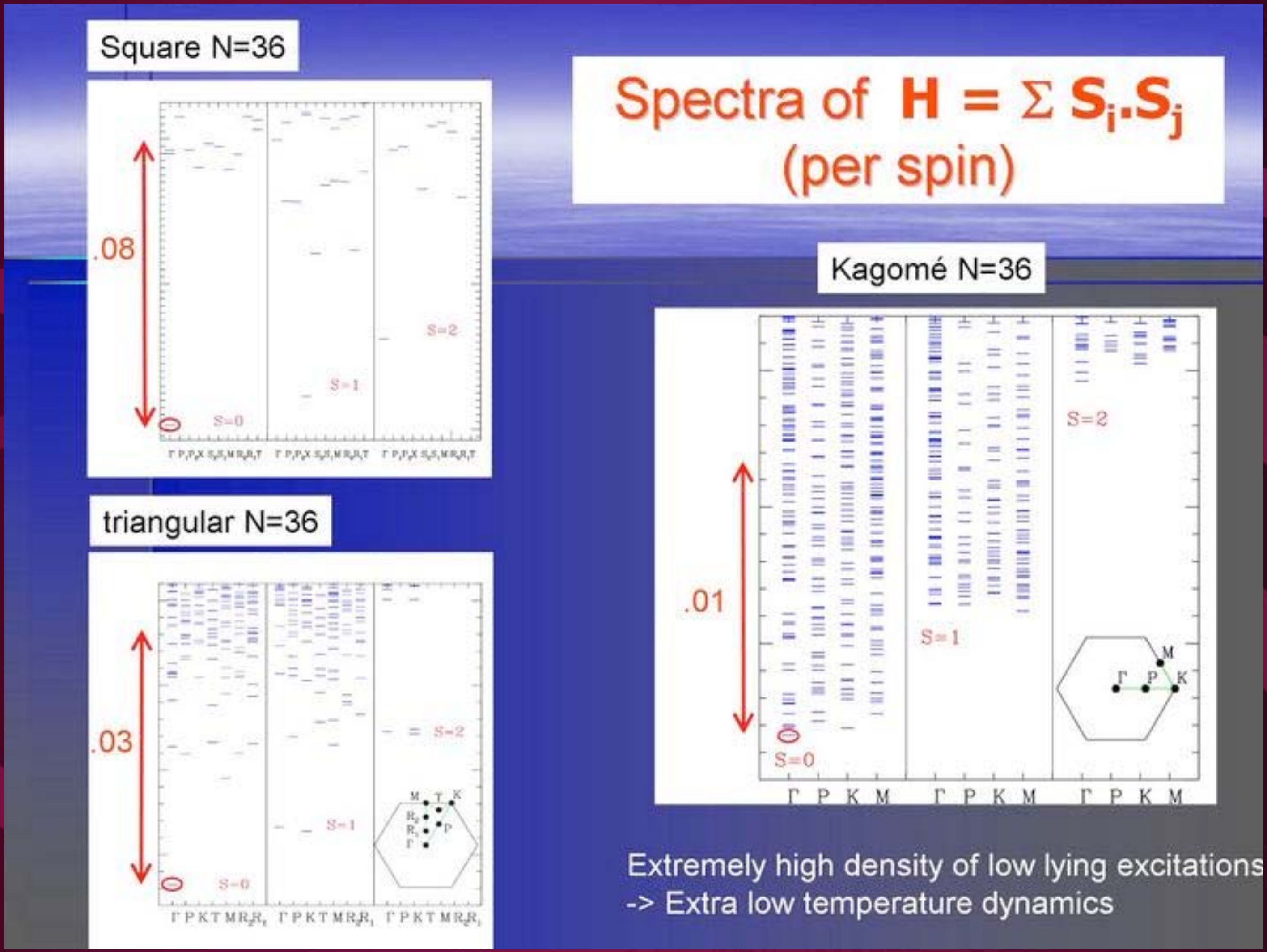


- Spectra from exact diagonalization
- Series expansions
- Other numerics

Exact Diagonalization
French Group, Elser+Zeng, …

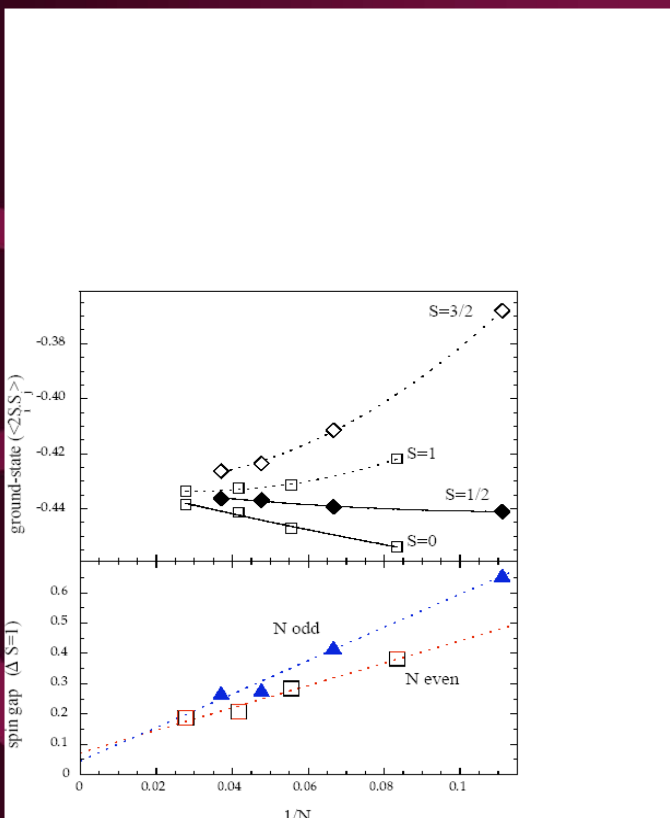
- Clusters upto size 36 PBC (one choice)
- Lots of low lying singlets
- # of singlets below triplet goes as a**(N)?

Low Lying Spectra (Lhuillier)



Spin-gap is zero or small?

Extrapolation from Exact Diagonalization



GS Energy =-0.43 -- -0.44

Triplet Gap <0.05

Maybe 0!

Momentum Dependence
36-site PBC

- Four q points- 0(Gamma), K, Q, M
- Minimum triplet gap at q=M (French Group)
- Zeng+Elser Spin and Dimer Correlations (Huse+RRPS)
- Largest eigenvalues of correlation matrices

q	spin-spin (3X3)	dimer-dimer(6X6)
0	0.49998	0.4013
Q	0.35856	0.3375
M	0.43806	0.6736

Is there a VBC?: SU(N) Large N: Many Possibilities Here Too
Large N: Max-Perfect Hexagons

Marston

Zeng

Nikolic

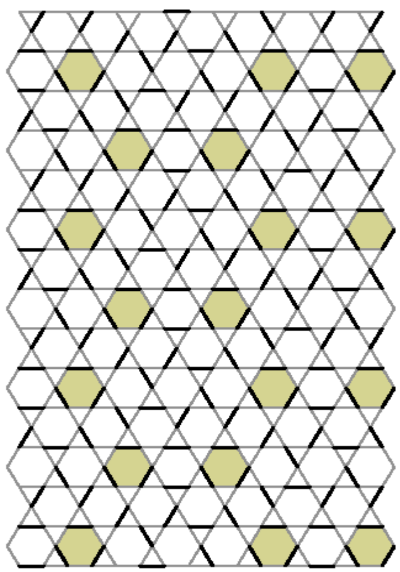
Senthil

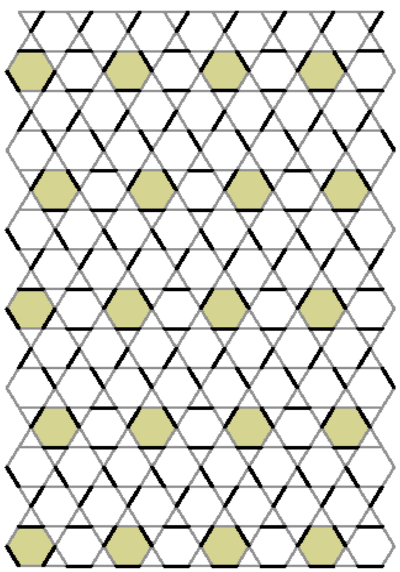
36-site unit cell

Honeycomb

Stripes

Both have 36-site unit cells (Need different PBC)

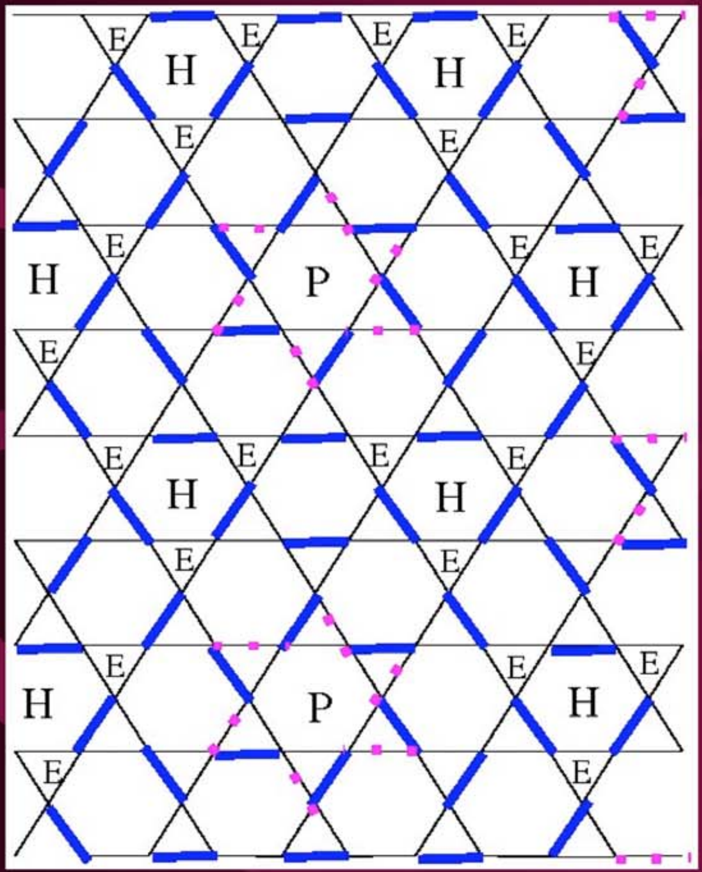




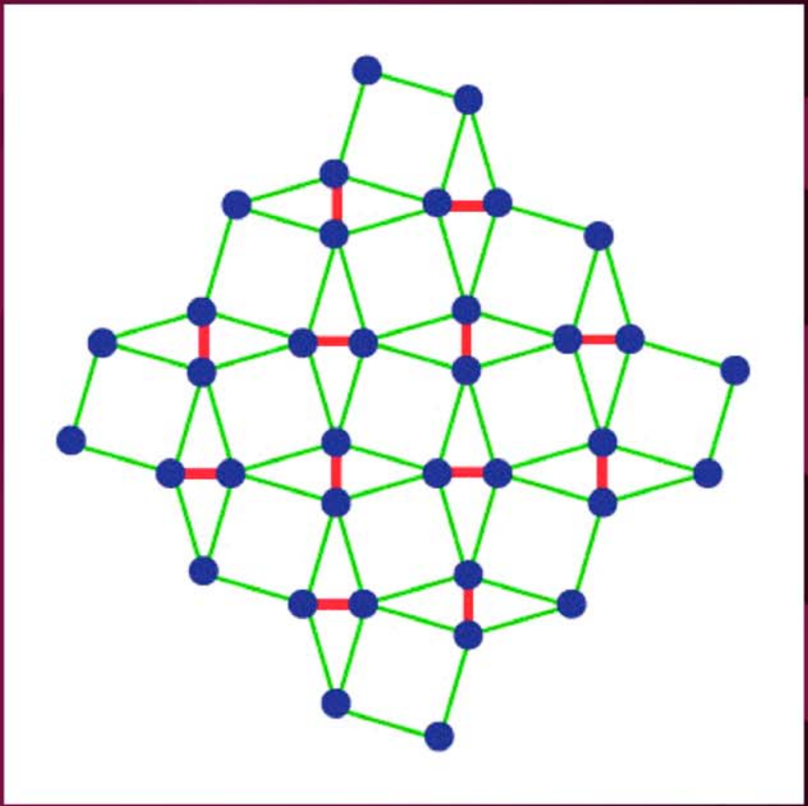
Dimer Expansion for spin-half

Empty Triangles are Key

The rest are in local ground state



Kagome Lattice

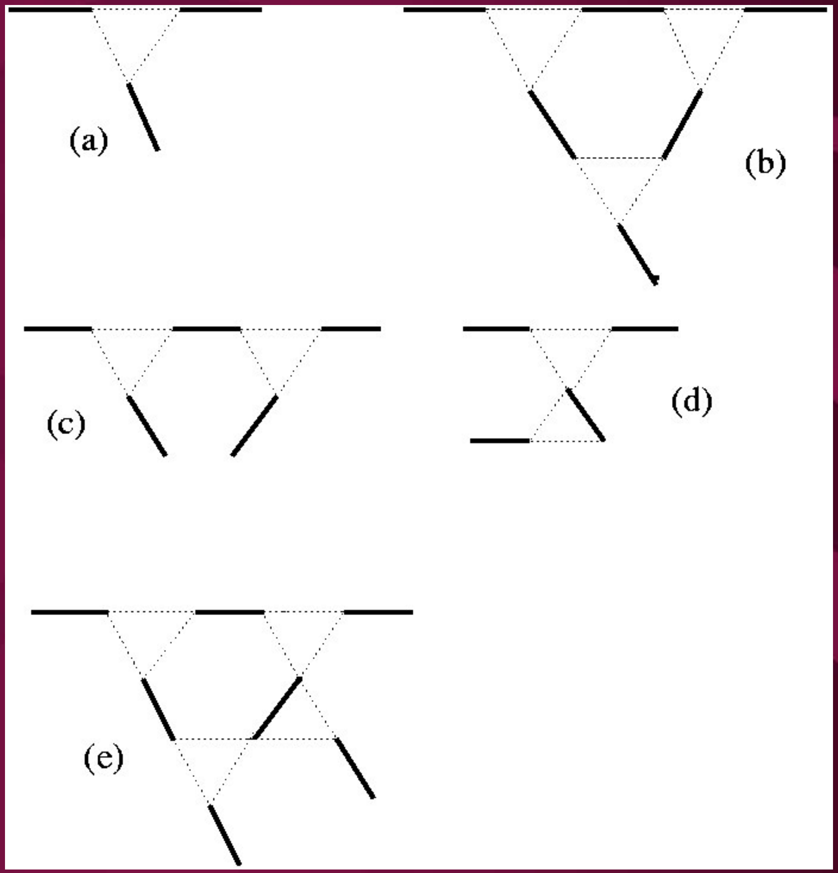


Shastry-Sutherland Lattice

Series Expansion around arbitrary Dimer Configuration

Graphs defined by triangles

All graphs to 5th order



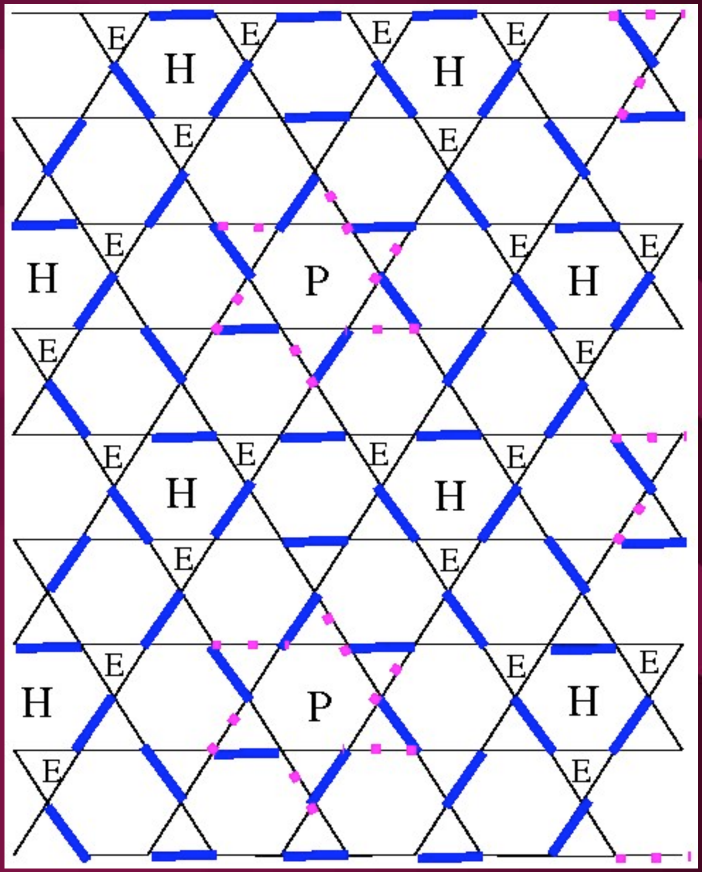
Degeneracy Lifts in 3rd/4th Order But Not Completely

3rd Order: Bind 3Es into H

4th Order: Honeycomb over Stripe

Leftover: Pinwheels

24*2^(N/36) Low energy states



Series show excellent Convergence

Order	Honeycomb	Stripe VBC	36-site PBC
0	-0.375	-0.375	-0.375
1	-0.375	-0.375	-0.375
2	-0.421875	-0.421875	-0.421875
3	-0.42578125	-0.42578125	-0.42578125
4	-0.431559245	-0.43101671	-0.43400065
5	-0.432088216	-0.43153212	-0.43624539

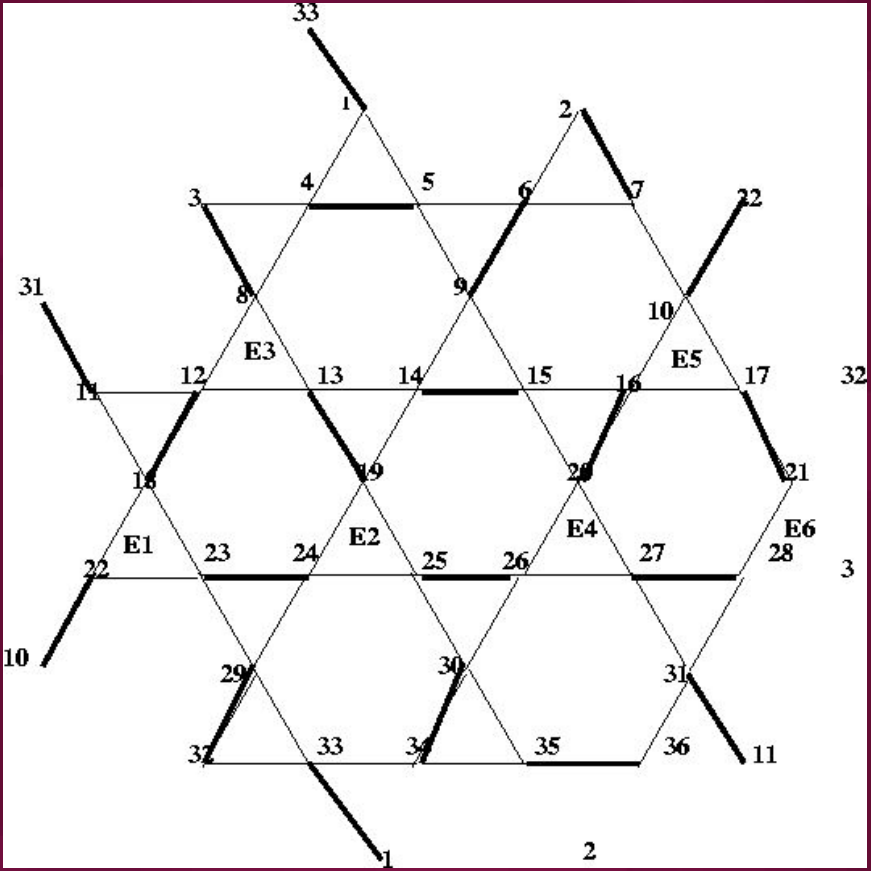
Ground State Energy per site

Estimated H-VBC energy: -0.433(1)

36-site PBC: Energy=-0.43837653

Variational state of Ran et al (Hastings)-0.429

36-site PBC wraps around
New graphs start contributing in 4^{th} order
Closed Loops of 4 triangles



Dimer Order Parameter

Order	0th	2nd	3rd	4th	5th	6th
Strong (within hexagon)						
	-.75	-.5625	-.516	-.437	-.428	-.423
Weak (within hexagon)						
	0	-.1875	-.258			

Resonance within hexagons maybe restored

Both strong and weak approximately -0.4!

Mean energy per bond = -0.217

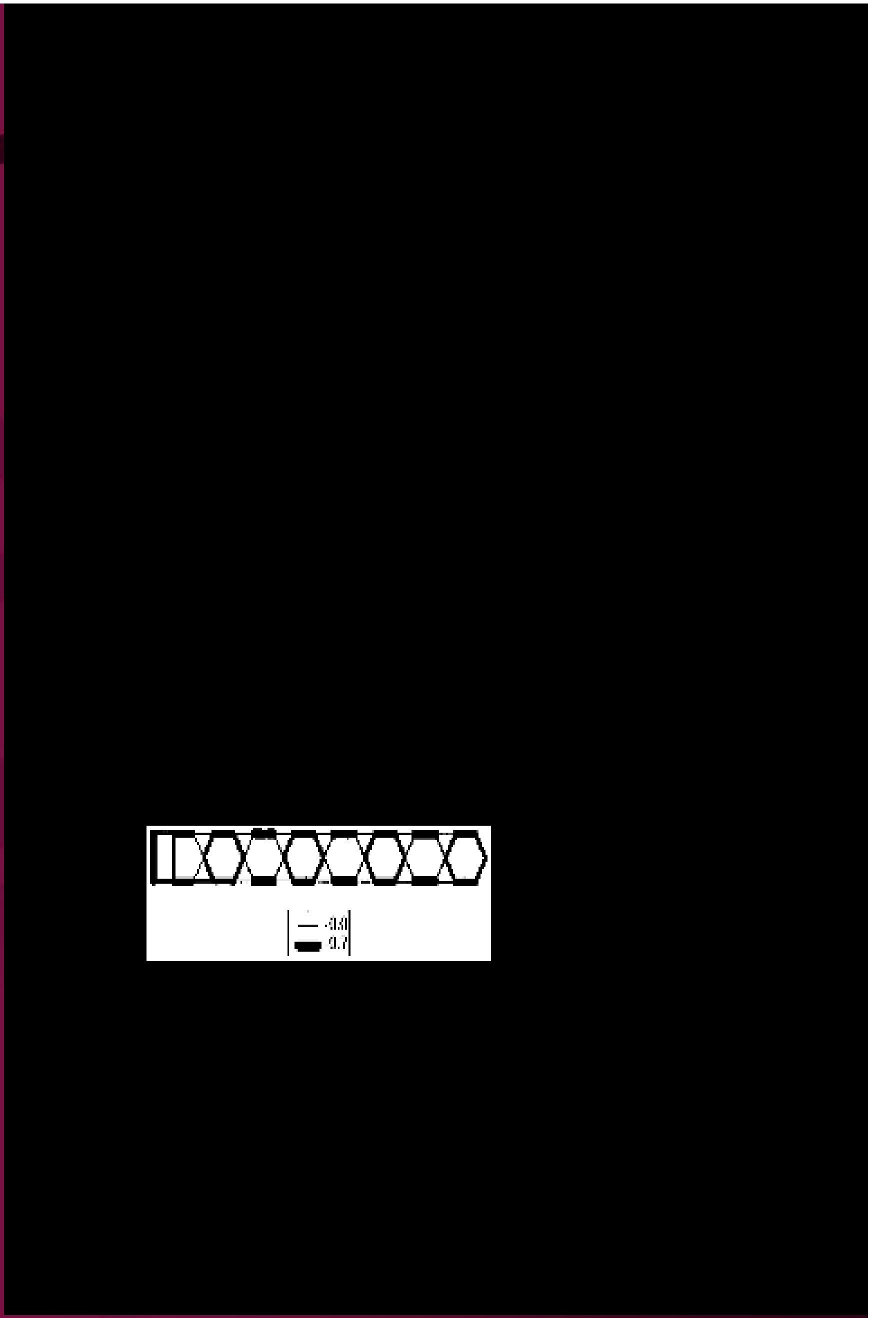
Kagome Stripes

Azaria et al PRL 81, 1694 (1998)

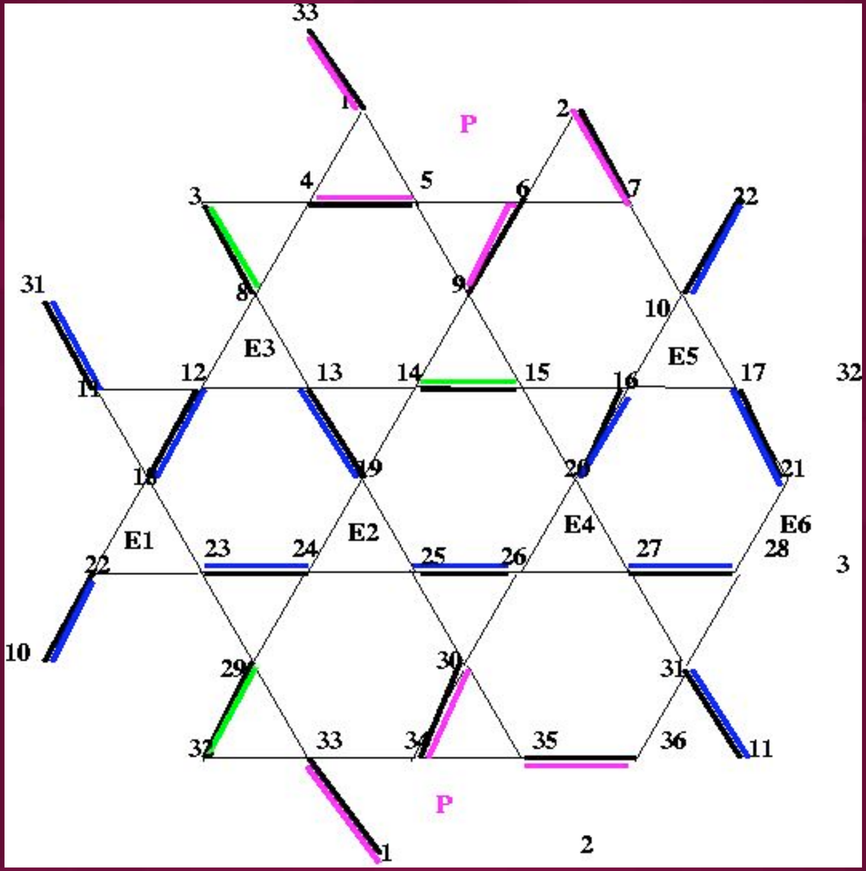
Gapless Singlet Modes

S. R. White and RRPS PRL 85, 3330 (2000)

Stripe VBC (gap 0.01)



What about the spin spectra?
18 by 18 matrix



Spin Spectra

- Up to Second Order Perturbation Theory:
  --Partly like bits of chains
  --Partly like Shastry Sutherland Model
  --Only some of the triplets can hop

- Lowest lying triplet localized to Hexagons
  E=1-0.5 -0.875 +0.890625-0.5… (poor convergence)
  Higher order analysis (future work)

ED (PBC36) has many more states at low energies

Misguich

+Sindzingre

Symmetry of

Low Lying States

Mambrini+Mila

Dimer subspace

Has continuous

spectra

n	E	k	R_3	R_2	σ	Deg.
1	-15.7815551190	0	1	1	1	1
5	-15.7714422841	B	$e^{\pm 2i\pi/3}$			4
7	-15.7705526907	0	$e^{\pm 2i\pi/3}$	1		2
8	-15.7677646622	0	1	1	1	1
14	-15.7626378391	C			1	6
15	-15.7530636858	0	1	-1	1	1
18	-15.7530438440	A		1	1	3
24	-15.7506986611	C			-1	6
25	-15.7397638762	0	1	1	-1	1
27	-15.7387667284	0	$e^{\pm 2i\pi/3}$	1		2
30	-15.7373154352	A		-1	1	3
32	-15.7338387327	0	$e^{\pm 2i\pi/3}$	-1		2
34	-15.7334329978	B	1		1	2
37	-15.7332490188	A		-1	-1	3
38	-15.7269119422	0	1	-1	-1	1
40	-15.7267147209	B	1		-1	2
43	-15.7261524204	A		1	-1	3
49	-15.7260314565	C			1	6
52	-15.7254780685	A		1	1	3
58	-15.7221590458	C			-1	6
60	-15.7202552097	B	1		1	2
64	-15.7199632440	B	$e^{\pm 2i\pi/3}$			4
70	-15.7186955013	C			1	6
76	-15.7116604180	C			-1	6
79	-15.7115521063	A		1	-1	3
83	-15.7092916444	B	$e^{\pm 2i\pi/3}$			4
85	-15.7074451283	B	1		-1	2
88	-15.6974529791	A		1	1	3
94	-15.6953790715	C			-1	6
95	-15.6950982554	0	1	1	-1	1
101	-15.6894717552	C			1	6
103	-15.6881000899	B	1		1	2
105	-15.6870862487	0	$e^{\pm 2i\pi/3}$	1		2
111	-15.6780830086	C			-1	6
117	-15.6775636462	C			1	6
120	-15.6749606790	A		1	1	3
126	-15.6721274935	C			1	6
129	-15.6678064885	A		-1	-1	3
133	-15.6635057830	B	$e^{\pm 2i\pi/3}$			4
136	-15.6620584319	A		1	-1	3
139	-15.6567299552	A		-1	1	3
140	-15.6530663535	0	1	1	1	1
141	-15.6524533863	0	1	-1	1	1
143	-15.6508267100	0	$e^{\pm 2i\pi/3}$	-1		2
145	-15.6506529276	B	1		-1	2

States with R3 not unity are unrelated to Honeycomb-VBC

BUT

Energy of PBC is much larger than separation (0.005 vs 0.001)

Dimer order has not yet set in at this scale

Overall picture from these studies

- VBC Order very weak (Delta E order .001)
- Only sets in at very low T--large L
- Intermediate L,T: Dimer Liquid (RVB?)
- Small spin-gap to nearly localized triplets
- Lots of singlets at the gap-edge (chi)
- Sensitivity to Perturbations (Further Neighbor Js, Spatial anisotropy, DM, Impurity, …..)

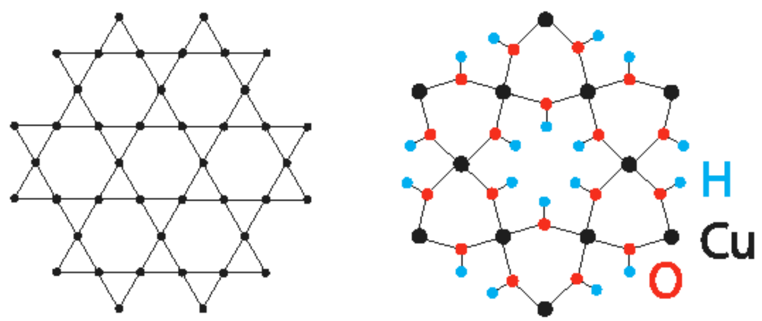
Experimental Status

- New material: Herbertsmithite
  ZnCu_3(OH)_6Cl_2

Cu atoms carry spin-half

Kagome-layers of Cu

Separated by layers of Zn



Some experimental properties

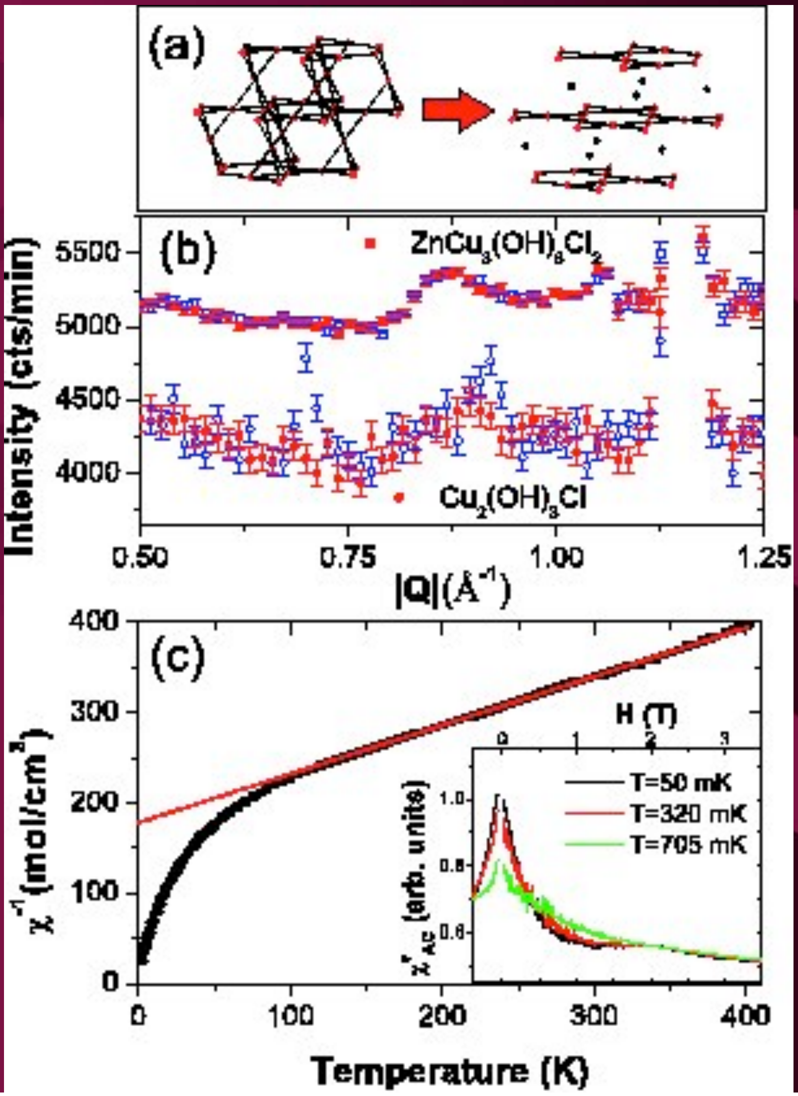
- Curie-Weiss T=300K
- No LRO down to 50mK

BUT

- Susceptibility turns up at low T!

Helton et al PRL

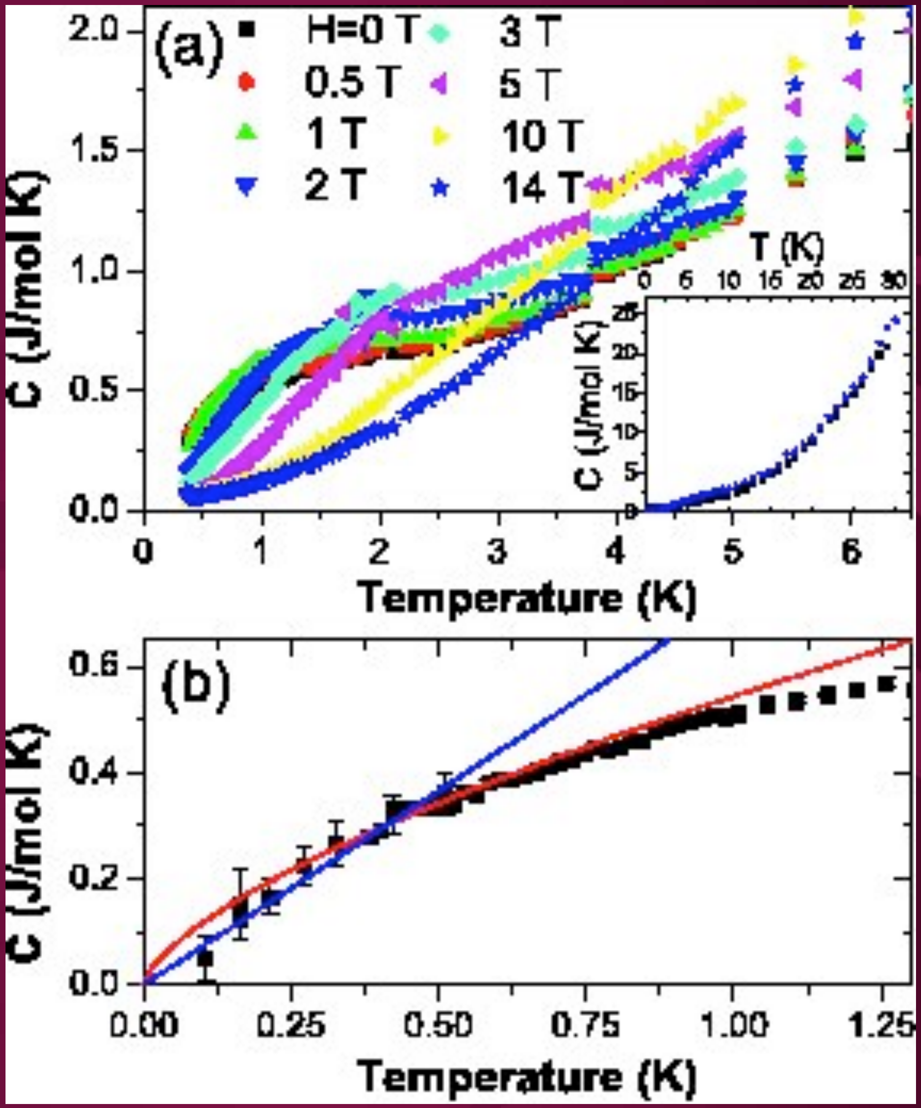
Ofer et al cond-mat



Specifc heat sublinear at low-T

Highly sensitive

to magnetic field!



Rigol and RRPS

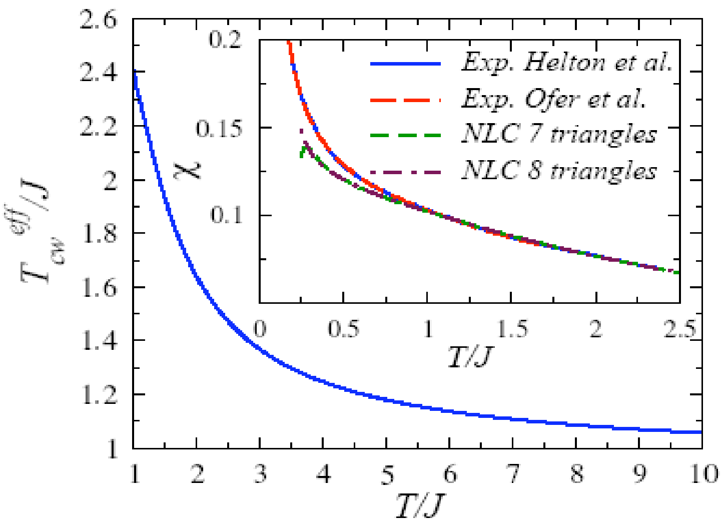
Good Fit with

J=170K, g=2.19

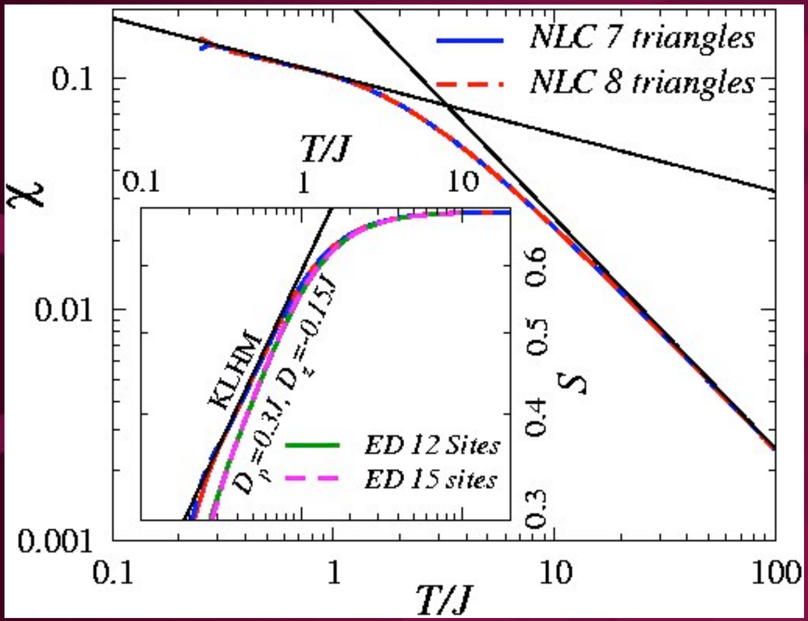
CW is not asymptotic

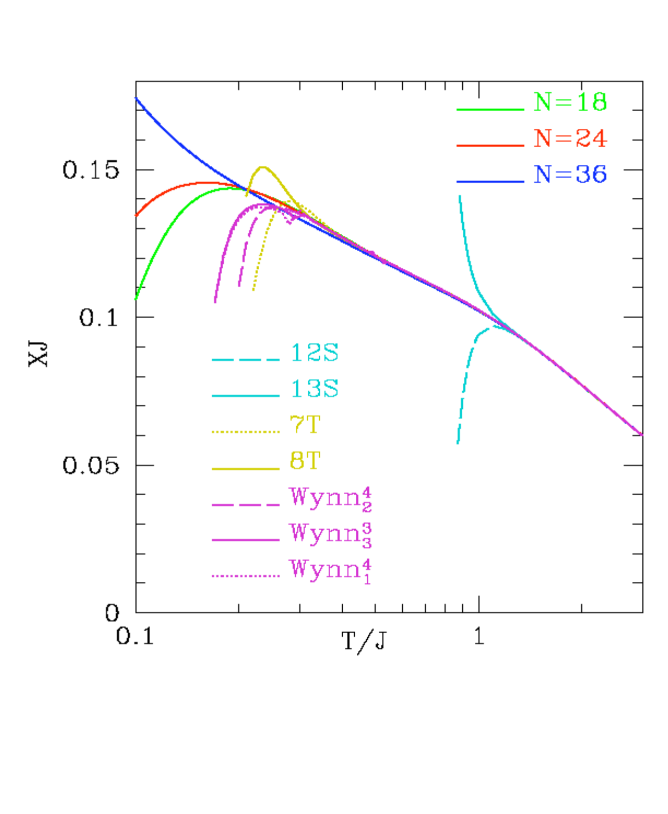
PRL 2007

Sharp upturn at low T not consistent with Kagome-HAFM



Where does the Kagome susceptibility peak? (below T=0.1!)



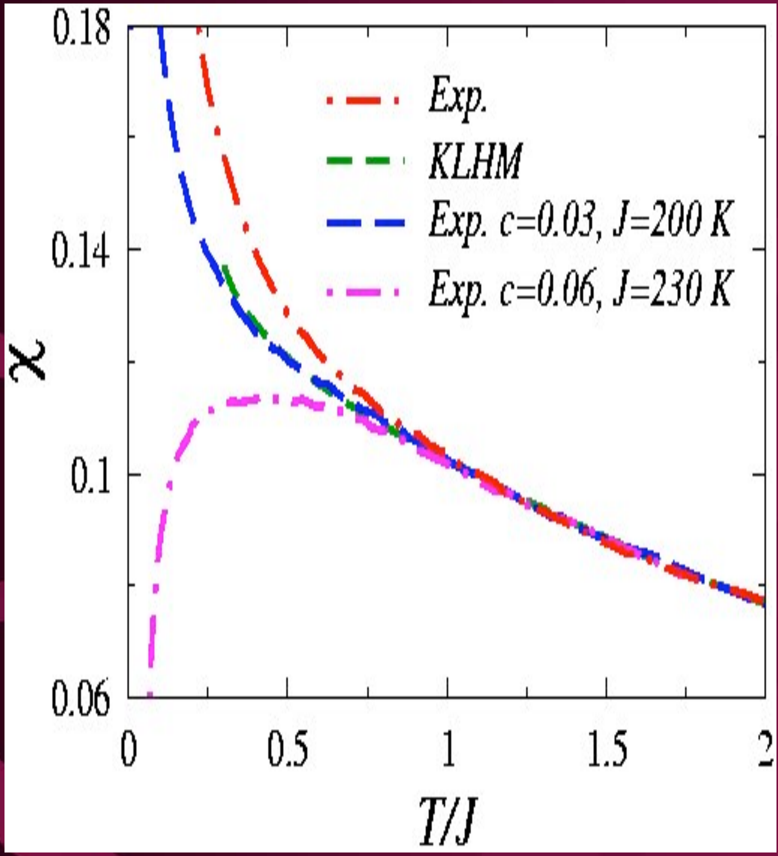


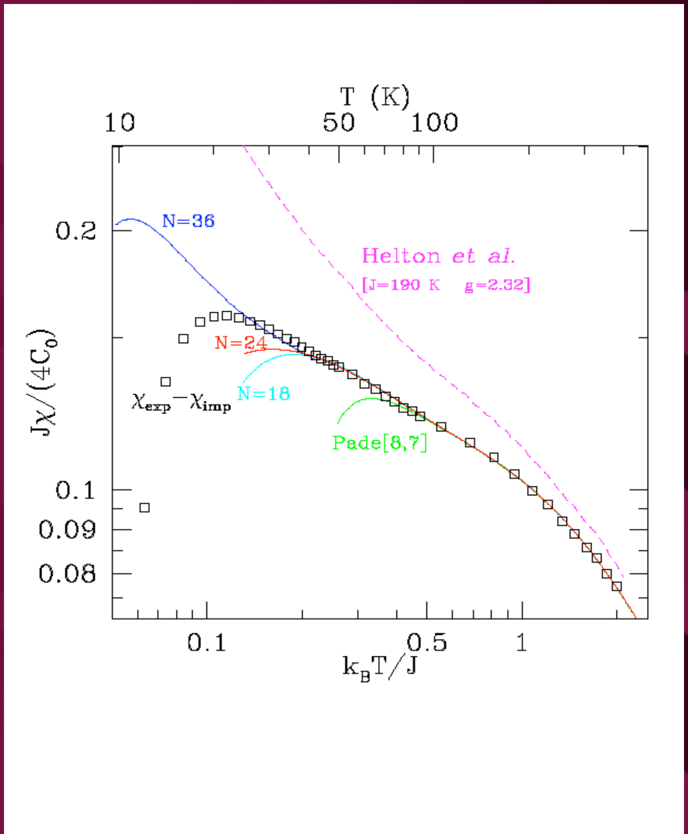
Rigol+RRPS

Misguich+ Sindzingre

Crossover to Reduced # of Localized Triplets?

Is the upturn due to impurity?





Misguich+sindzingre

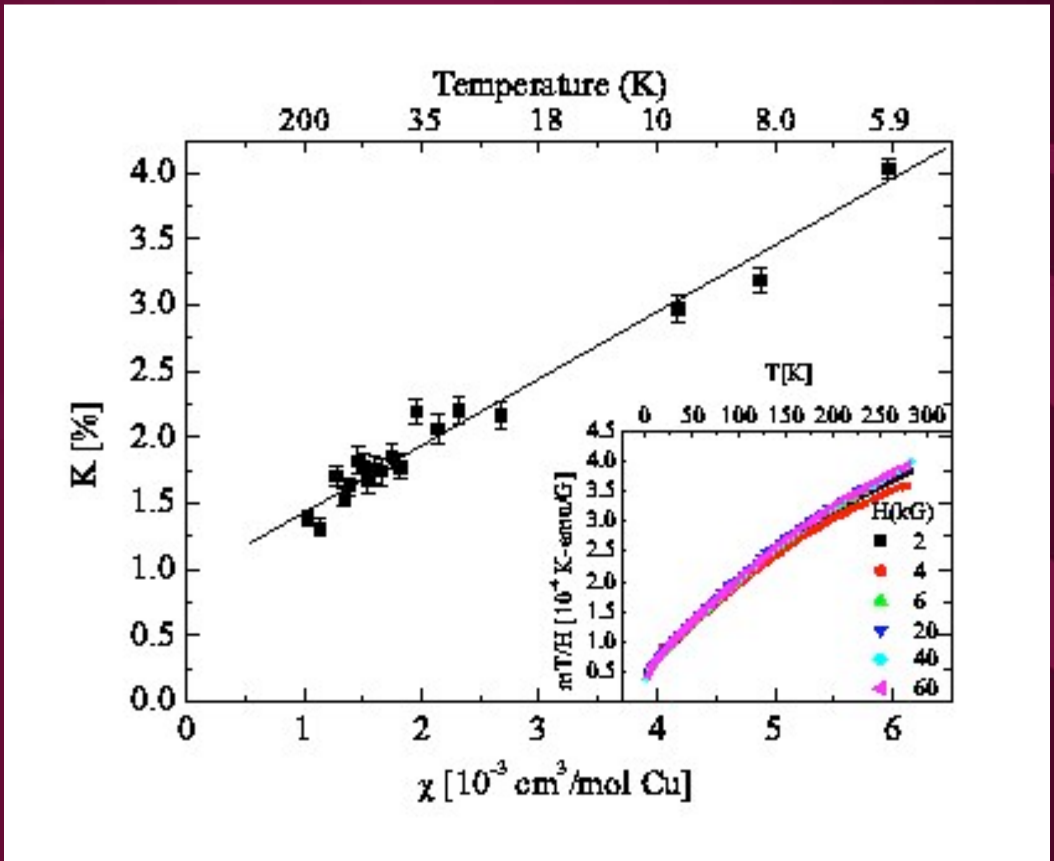
Rigol+RRPS

FM CW constant 6.5K

c=0.04 Agrees to 0.3 J

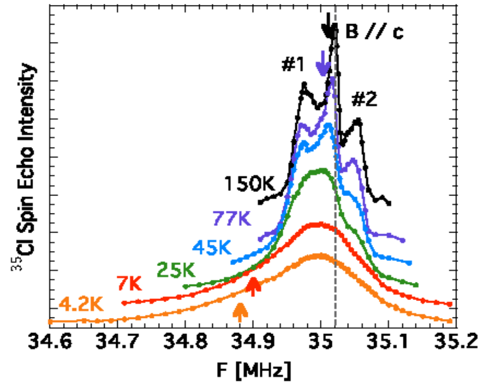
Agrees to 0.1 J

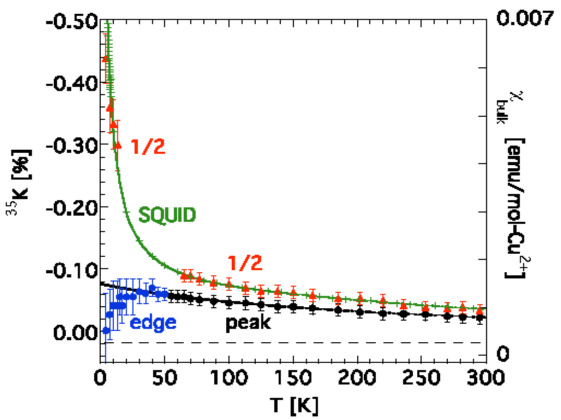
muSR tracks bulk susceptibility suggests it is intrinsic! (Ofer et al)



Neutrons show 6% antisite disorder (Bert et al arXiv:0710.0451)

Cl NMR (Imai's group)





Sign of T-dependent inhomogeneity

Dzyloshinski-Moria Interactions?

J|A|C|S
COMMUNICATIONS

Published on Web 09/09/2005

A Structurally Perfect $S = {}^{1}/_{2}$ Kagomé Antiferromagnet

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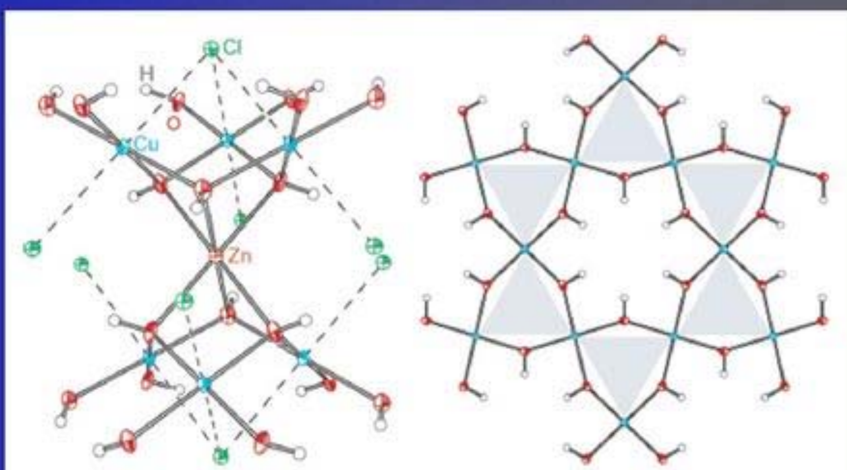


Figure 1. Crystal structure of Zn−paratacamite (**1**), $Zn_{0.33}Cu_{3.67}(OH)_6Cl_2$.

Kagome maybe perfect

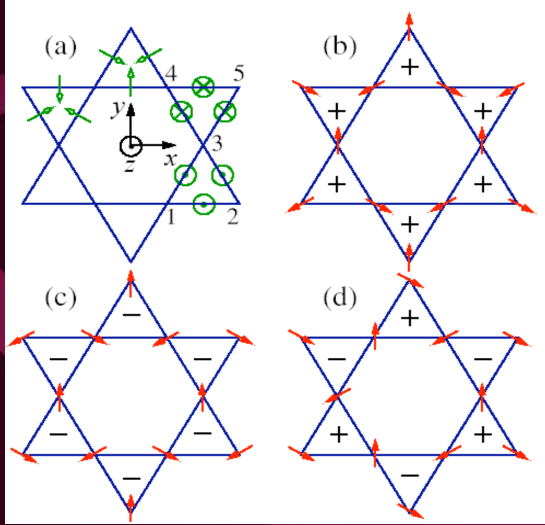
But overall structure is quite distorted

Two independent DM parameters allowed by Symmetry

Dz and Dp

Dzyloshinski-Moria Interactions
Cross Product between spins

Both Dz and Dp are of order 10% of J in structurally related Fe-based spin-5/2 material

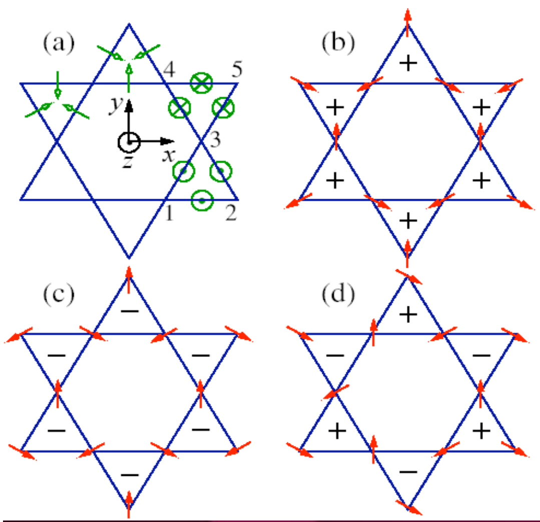


Dz can order the system!

Planar + preferred helicity

Selects a **unique Classical Ground state** in the 120-degree subspace

Dzyloshinski-Moria Interactions
Cross Product between spins



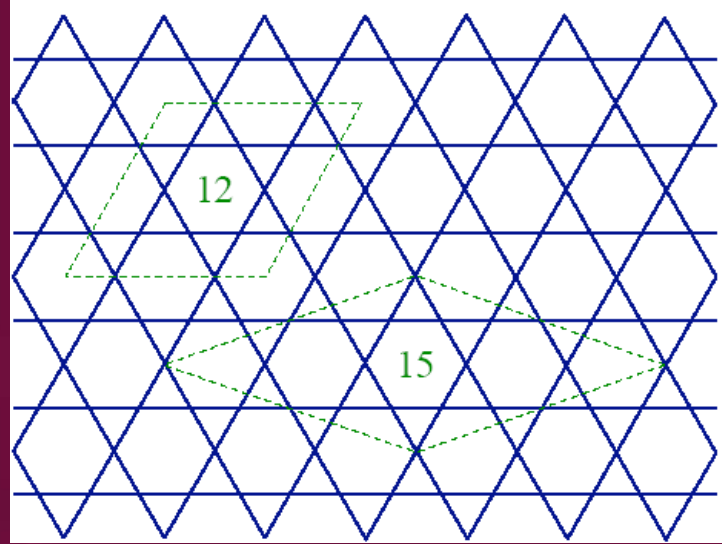
Dp rotates from bond to bond

No spin symmetry left

Cannot be satisfied in 120-degree subspace

Classically a small Dp leads to canting—like a FM Ising anisotropy!

Rigol+RRPS

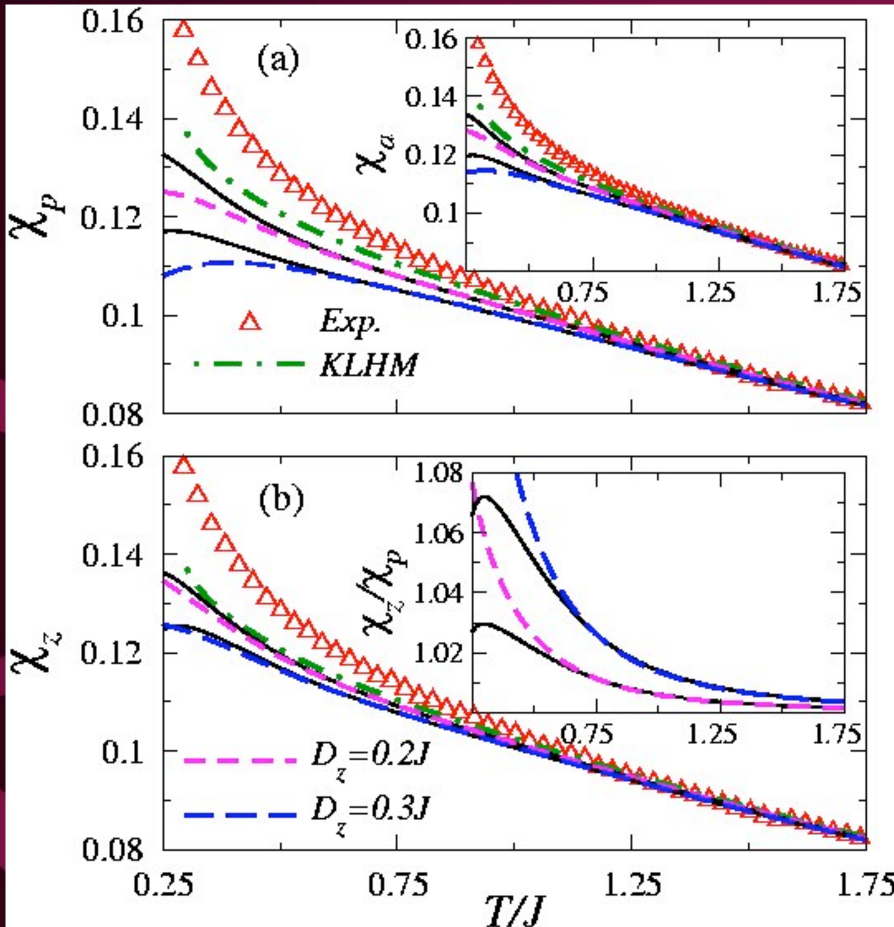


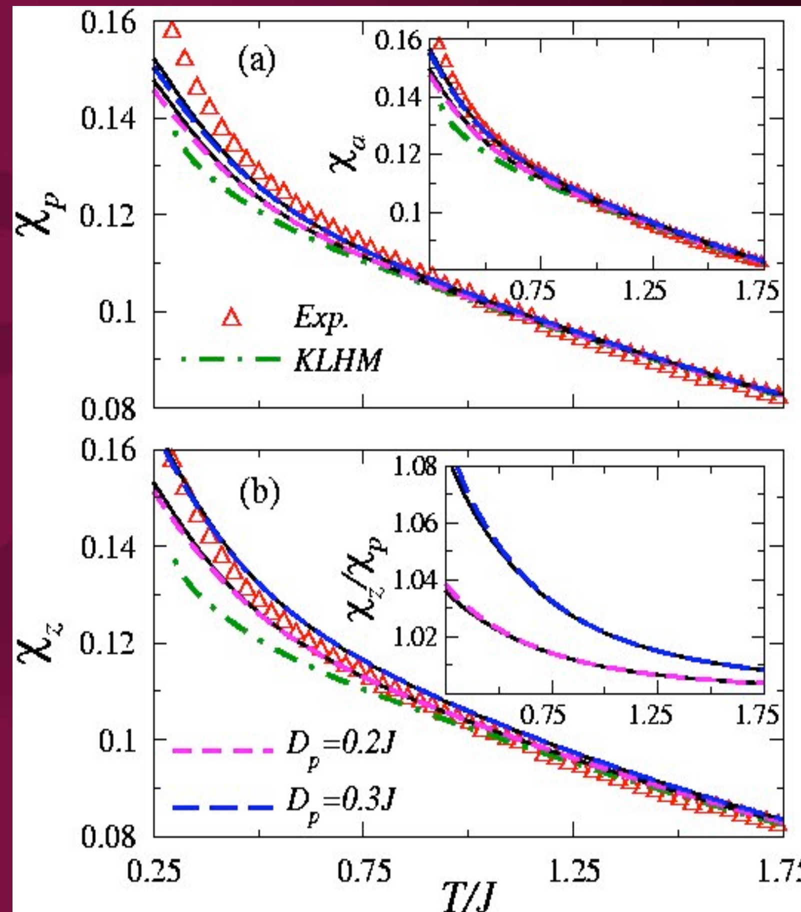
Lack of conservation laws makes numerics harder

Clusters for finite-size studies with

Periodic Boundary Conditions

Susceptibility with Dp and Dz

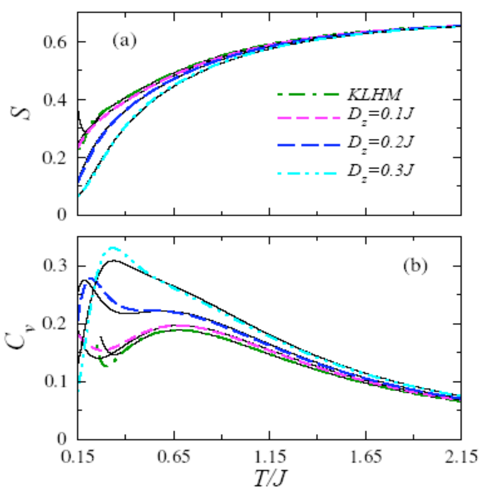


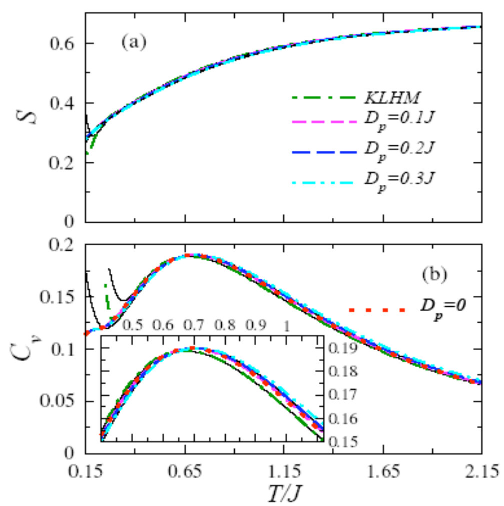


Dz lowers susceptibility

Dp increases susceptibility

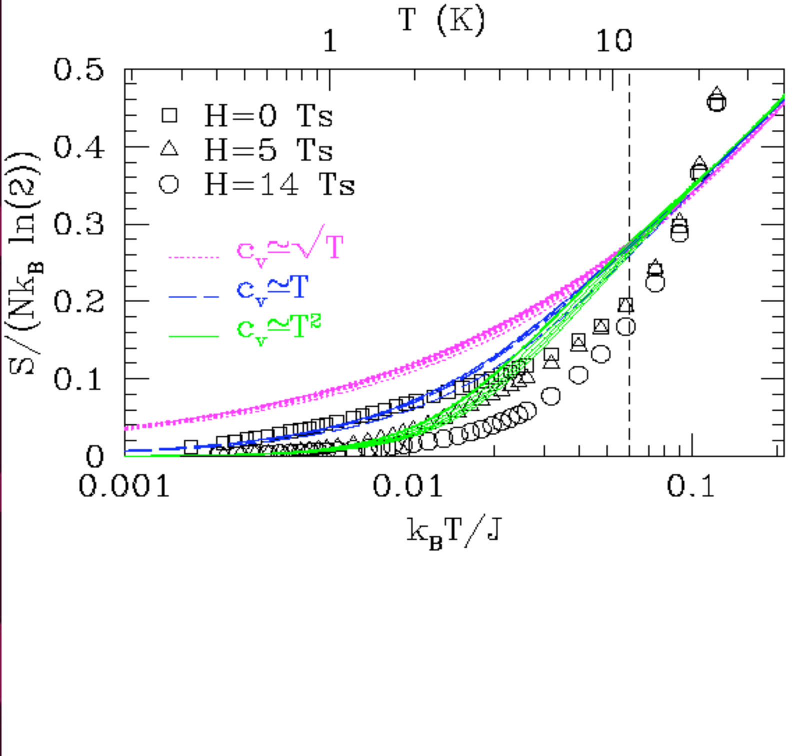
Both lead to anisotropy enhancing z-susceptibility

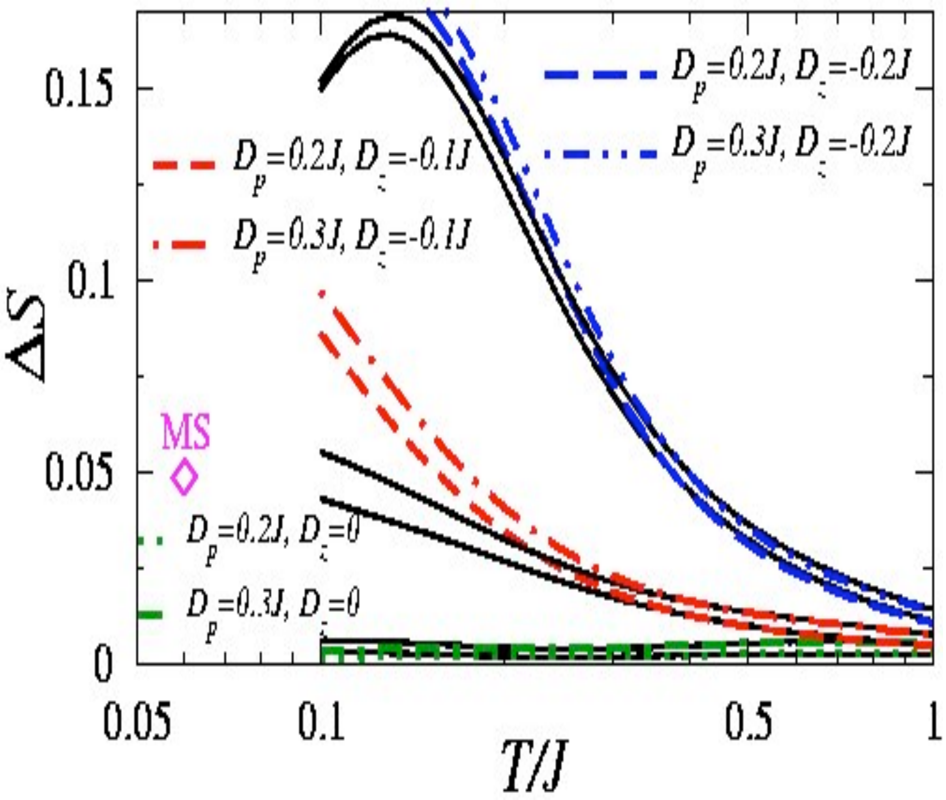




Entropy and Specific Heat
Dz: Entropy drops rapidly
Dp: No discernible change
What does Dp do to states?

Entropy and experiments: Assuming no frozen entropy, impurity (glassiness)





Misguich and Sinzindgre

High-T expansions

Lowering of entropy due to DM Interactions

DM Interactions
Finite-T studies: conclusions

- D_z: Reduces entropy, reduces isotropic susceptibility—Leads to long-range XY order

  -----cant be the answer by itself
- D_p No change in entropy, increases susceptibility suddenly, makes it highly anisotropic—could be the answer

  induced FM Ising anisotropy
- Must have D_p greater than D_z (Expt)
- With impurities much smaller Dz maybe enough

  If Dz >gap chi: 1/Dz (> 10/J at low T)
- Anisotropy measurements

Summary and Conclusions

- Kagome Lattice may have a VBC ground state (Debate Not Over)
- Perfect Hexagons more robust than fragile VBC
- Very small energy scales between different phases
  What are the implications at finite T?
- Small triplet gap--lots of states near the edge--chi does not peak down to quite low Temperatures
- DM interactions are allowed- Will be there only magnitudes can vary
- Dz and Dp are quite different—the latter is more intriguing and relevant to materials
- If there is an exotic state (such as Dirac spin-liquid) how can it be established?

The End