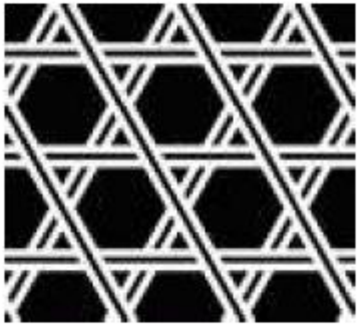
Exotic Antiferromagnets on the kagomé lattice: a quest for a Quantum Spin Liquid

Claire Lhuillier



Université Pierre et Marie Curie
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&CNRS

Physics of New Quantum Phases in Superclean Materials (PSM2010)
Yokohama, Japan (March 9 -12, 2010)

Laura Messio (Ph.D student)

Philippe Sindzingre

Grégoire Misguich IPhT Saclay

J.C. Domenge (Ph. D student *Not shown)*

Outline

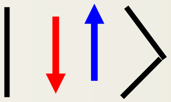
- Spin liquids and exotic antiferromagnetic phases: some definitions (parallel with quantum liquids)
- Spin-1/2 Heisenberg model on the kagome lattice :

  VBC, Critical Spin Liquid or a quantum critical point?
- Herbertsmithite: a quasi perfect Heisenberg model on the kagome lattice?
- Other real compounds on the kagome lattice: Volborthite (*Hiroi et al*.), Cutitmb *(Narumi et al. Europhys. Lett. 2004),* Kapellasite *(A. Wills, B. Fak, 2010)* are not pure n.n. Heisenberg models... but may harbor exotic chiral phases (*Messio 2010*)
- A first order chiral transition at T≠0: the role of Z_2 vortices (*J.-C. Domenge , PRB 77 2008, L. Messio & P. Viot. PRB 78 2008)*

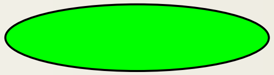
Exchange interaction : W. Heisenberg

$$H_{i,j} = \mathbf{S}_i . \mathbf{S}_j$$

- the “classical ground-state”: $|-,+\rangle$
  is a symmetry breaking state



- the quantum ground-state does not break SU(2):
  $|0\rangle = [\,|+,-\rangle - |-,+\rangle\,] /\sqrt{2}$ called a Valence Bond st.



•The variational energy of the classical state reduces to:

$E_{cl} = \langle +,-|\, H_{i,j}\, |+,-\rangle = \langle +,-|\, S^z_{i,} S^z_j\, |+,-\rangle =- \;- ¼$

whereas:

$E_{qu} = \langle 0|\, S^z_{i,} S^z_j\, |0\rangle \quad + \quad ½\, [\langle 0|\, S^+_{i,} S^-_j\, |0\rangle + [\langle 0|\, S^-_{i,} S^+_j\, |0\rangle\,]$

= the “classical energy” + “energy gain due to quantum fluctuations”

= -1/4 - ½

= <potential energy term> + <kinetic energy term>

What do theoreticians call Quantum Spin Liquids ?

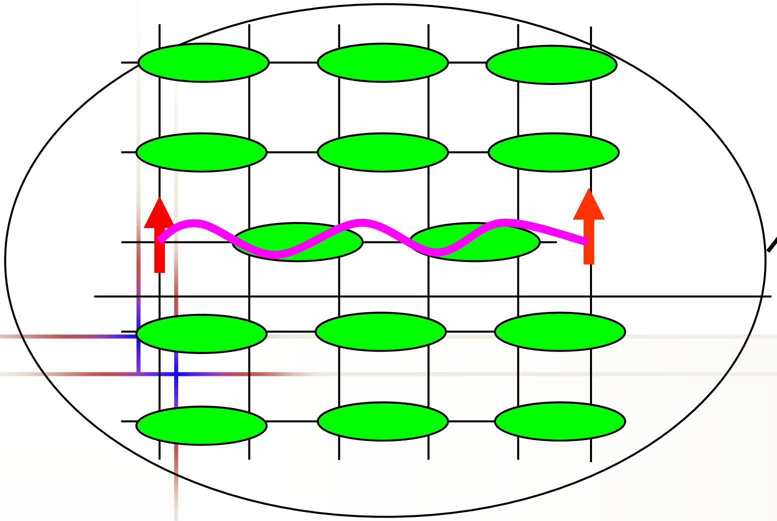
- A magnetic spin system with NO LRO in ANY local order parameter at T=0 and no symmetry breaking.

- Rather rare situation! Most magnets are “solid” like!
  - Colinear or non colinear Néel magnets have on site magnetizations
  - Nematic magnets : 4-spin ring exchange on square lattice -> nematic magnets: *Laeuchli et al, PRL 2005, Shannon, Momoi, Sindzingre 2005*
    on the triangular lattice , Momoi, Shannon ,Sindzingre 2006 -> quadrupolar or octupolar order
  - Valence Bond Crystals (*Shastry Sutherland, Fouet et al. 2003)* have long range order in singlet bonds.
- with gapless or gapful ΔS = 1 wave-like excitations

- Z_2 gapped spin Liquids: with unconfined ΔS = 1 /2 excitations,

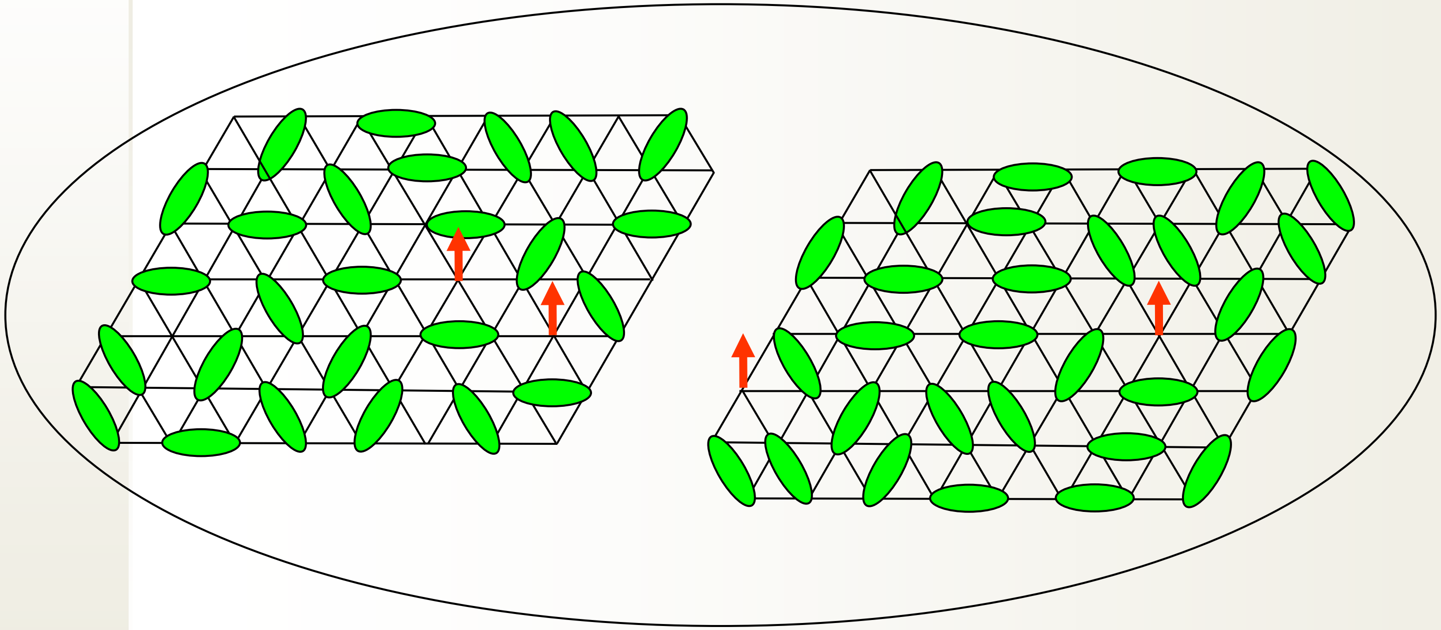
do exist in theoretical toy models, topological g.s. degeneracy, q-bit toy model

Misguich et al 98, 99 , Moessner & Sondhi 98, 99, Balents, Fisher and co-workers.
Experimental realisation? 2-d ^{3}He?



confined spinons in the
V-B crystal

unconfined spinons in
the R.V.B. Spin Liquids



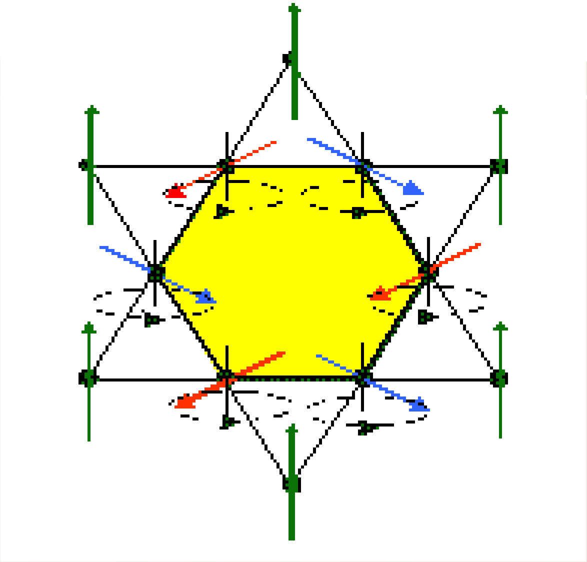
Classical & Quantum Heisenberg Hamiltonian on the kagomé lattice

$$H = \sum S_i . S_j$$

$$= \frac{1}{2} \sum_{\alpha} S_{\alpha}^{2} + \mathrm{Cst}$$

An infinite number of soft modes, an infinite T=0 degeneracy

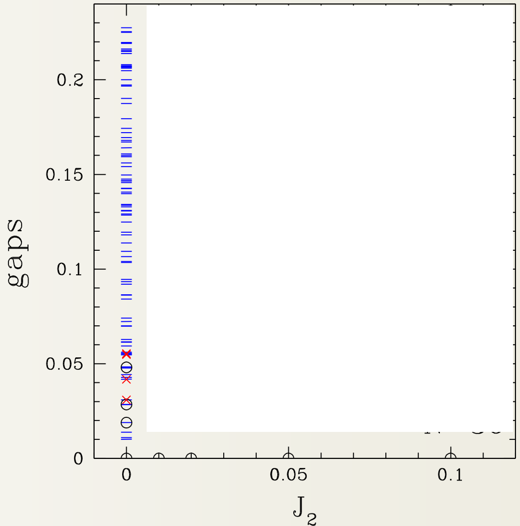
J. Chalker, et al 92, Huse & Rutemberg 92 , Reimers & Berlinsky 93

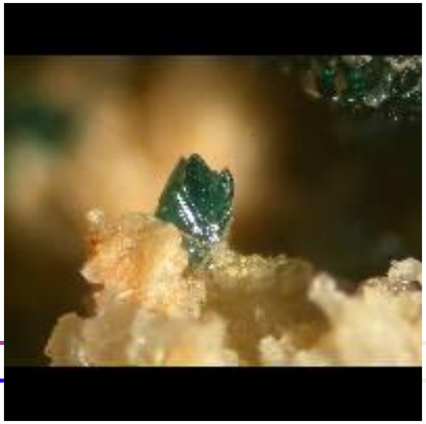


Quantum spectrum of excitations of N=36 spin-1/2 molecule: good ingredients for a spin liquid behavior, no local order parameter, discretization of energy ~10 $^{-3}$ …

Lecheminant & al. 97, Waldtmann & al. 99

Is it a large enough size to extrapolate to an infinite lattice?





$ZnCu_3(OH)_6Cl_2$

Shores& Nocera 2005
Bert & Mendels group Orsay 2007
Y. S. Lee group MIT 2007
Imai et al. Mac Master Univ. 2007-2008
S.H. Lee group

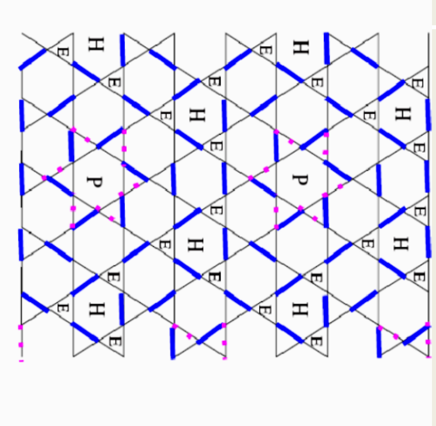
- Curie-Weiss temperature Θ_{cw}= -300 K
- No magnetic order down to 50 mK
- Dynamical features down to 50 mK
- No observable gap down to 0.1 meV
- No SG transition

A Spin Liquid phase down to $T \leq J/4000$

Role of impurities ? Dzyaloshinskii Moriya interactions?

Quantum Spin Liquid on the kagome lattice?
controversies amongst theoreticians

- Heisenberg model on the kagomé lattice :
  - *A Valence Bond Crystal* ?
    RRP Singh & D. Huse 2007,
    A small gap and a very large unit cell



  - *An algebraic spin liquid: Ran, Hermele et al. 2007-2008,*
    An extended gapless phase with fermionic spin-1/2 excitations
  - *A vortex spin liquid: S. Ryu, Motrunich, Alicea & MPA Fisher 2007 (XY model)*

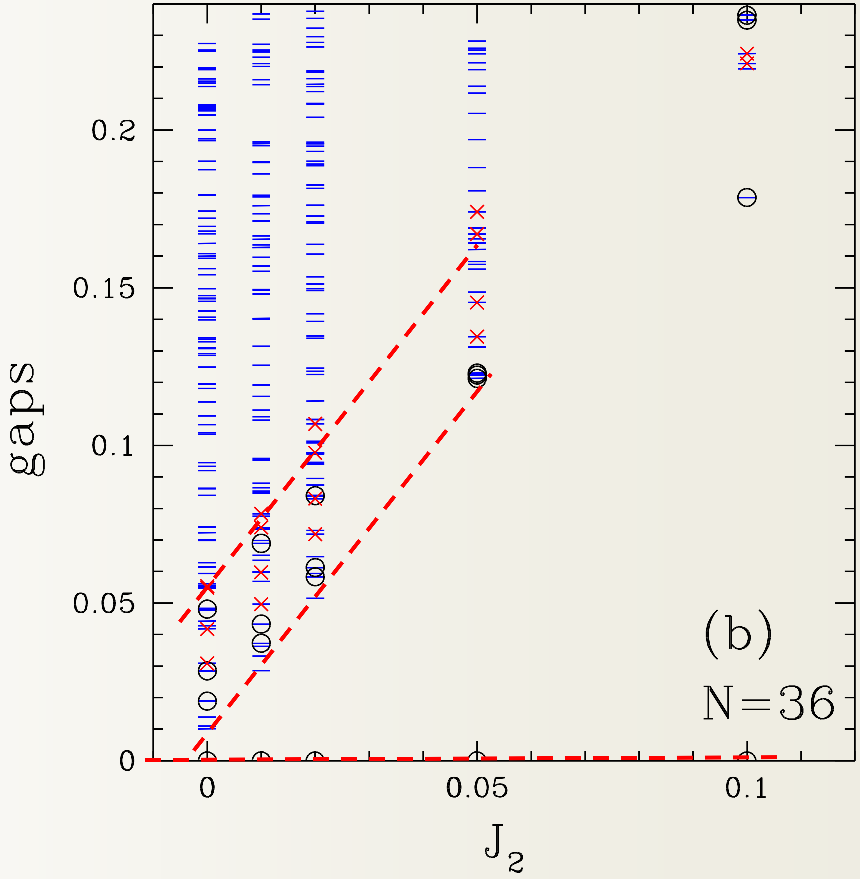
The Heisenberg model on the kagomé lattice: a Spin Liquid near a Quantum Critical Point?

P. Sindzingre & C.L: EPL 88 2009, arXiv:0907.4164/v2

- Instability of a putative VBC or of Hermele S.L.

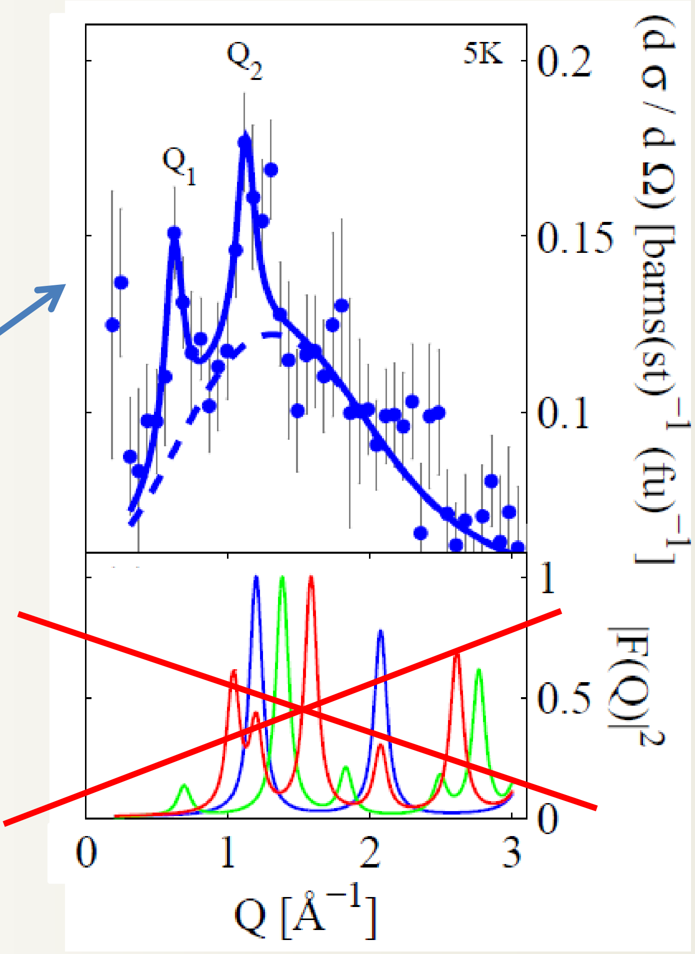
- No intrinsic low energy scale $3\ 10^{-3}$ for N=36 …

Could it be the signature of a QCP?

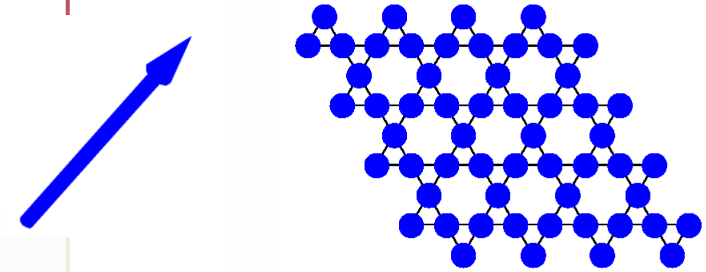


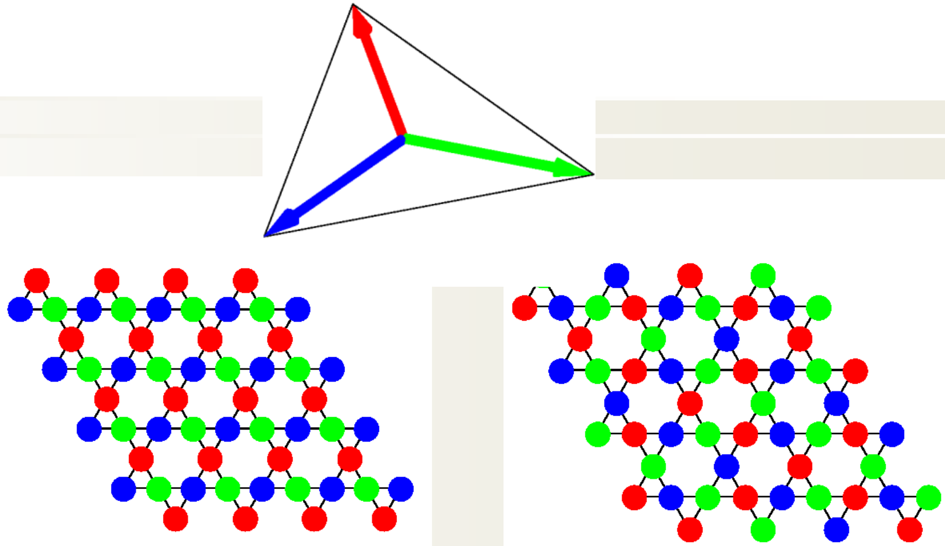
Other “real compounds” on the kagome lattice: Volborthite (*Hiroi et al*.),
Cutitmb *(Narumi et al. Eur.. Lett. 2004),*
Kapellasite *(A. Wills, B. Fak, 2010)*
are not pure n.n. Heisenberg models...

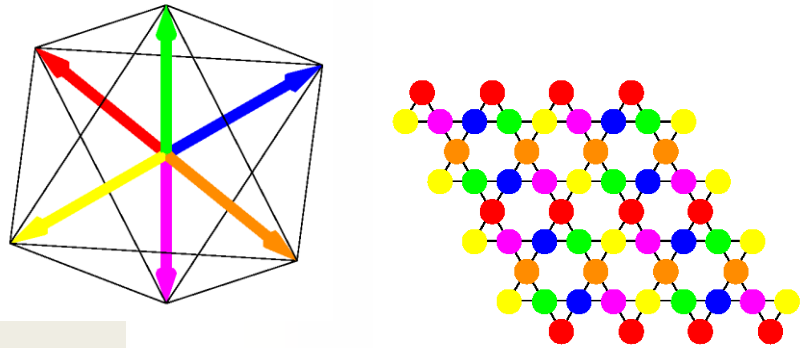
- Experimental indications of non coplanar SRO in Vollborthite
  - –Z. Hiroi’s group J. of Phys. Soc. Jpn 78, 2009,
  - –G. Nilsen & al. (EPFL 2010)
    arxiv:1001.2462

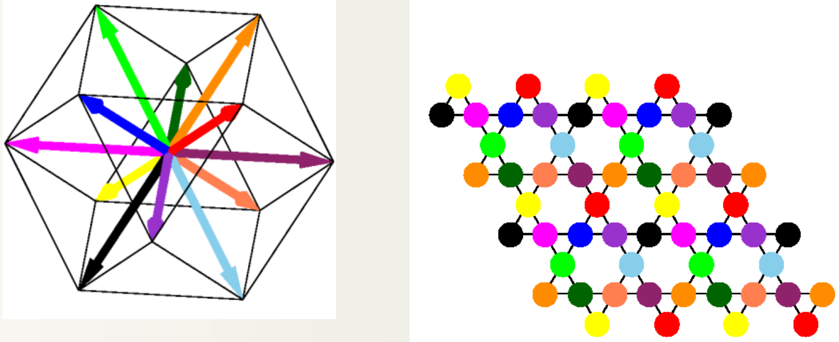


The classical & quantum short range orders on the kagome lattice (PSG analysis, *Messio 2010*)







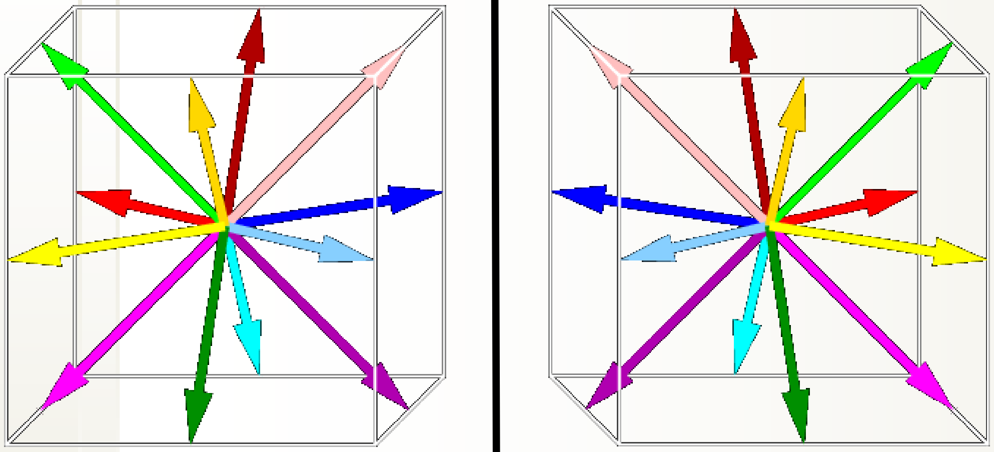


+ spirals …

Chiral sym. breaking in the 12-sublattice cuboc. phase and chiral phase transition

Domenge, Messio & al. PRB 77, 78 2008.

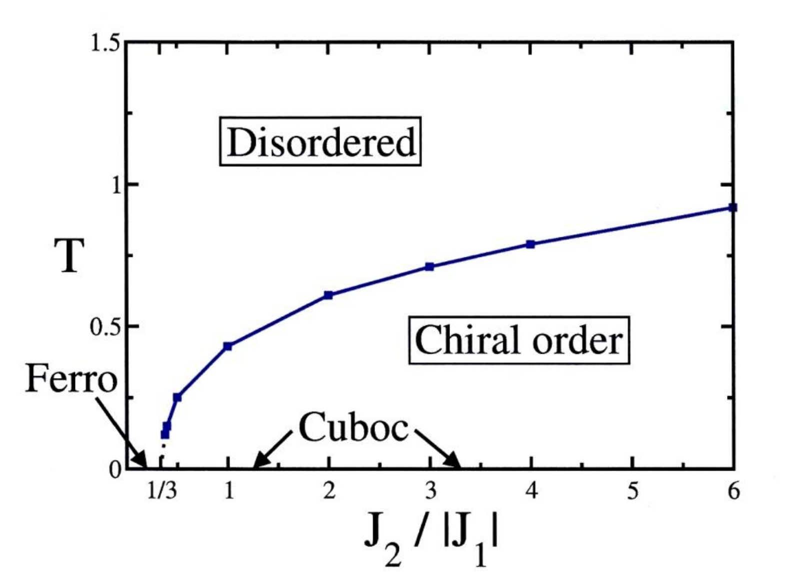
M.C. simulation – class. spins



Scalar chirality

σ = +1

σ = -1



Similar physics in MSE model

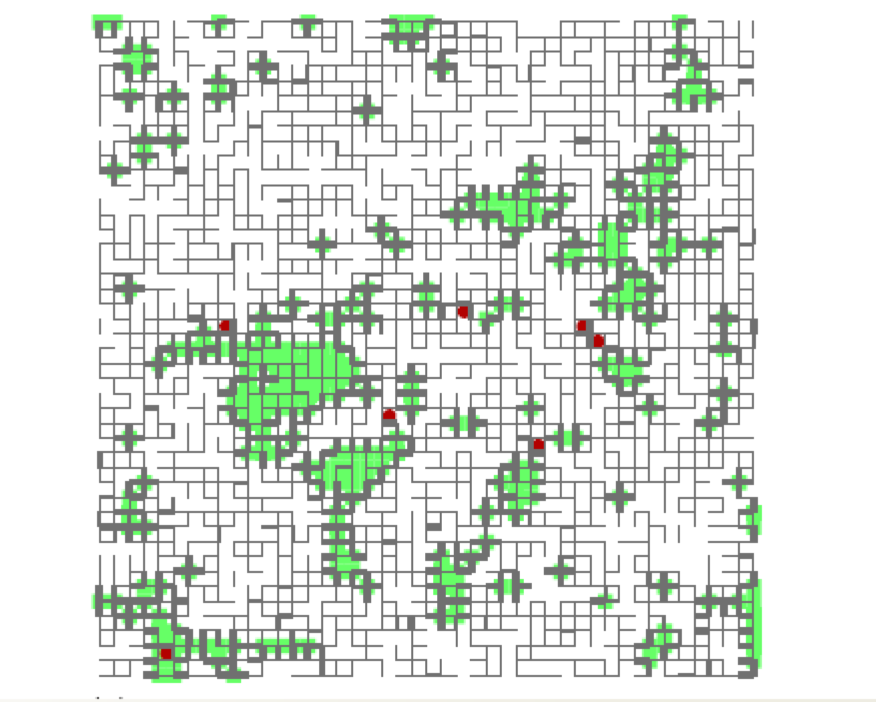
Momoi et al PRL 97

Weak universality (Suzuki 1984) ?

First order phase transition mechanism

Snapshot of a spin chirality configuration near the phase transition: *Z_2 vortices (brown points) nucleate in the domain walls of chirality (white/green boundaries) and modify the domain wall energy*

Messio et al. PRB 78 2008



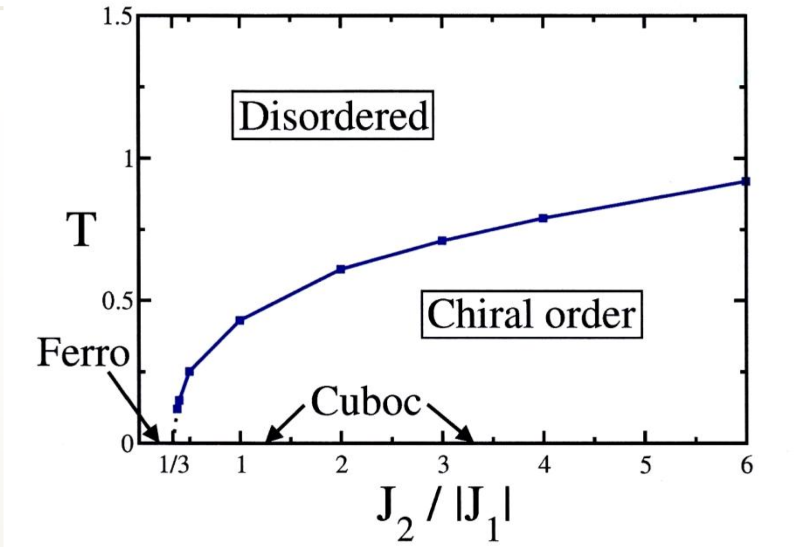
Summary

- Spin liquids and exotic antiferromagnetic phases: some definitions
- Spin-1/2 Heisenberg model on the kagome lattice :

  VBC, Critical Spin Liquid or a system near a Quantum Critical Point? (Sindzingre &C.L. 2009)
- Herbertsmithite: a quasi perfect Heisenberg model on the kagome lattice
- Other real compounds on the kagome lattice: Volborthite (*Hiroi et al.*), Cutitmb *(Narumi et al. Europhys. Lett. 2004),* Kapellasite *(A. Wills, B. Fak, 2010)* are not pure n.n. Heisenberg models... but may harbor exotic chiral phases (*Messio 2010*)
- A weakly first order chiral phase transition at T≠0: the role of Z_2 vortices (*J.-C. Domenge , PRB 77 2008, L. Messio & P. Viot. PRB 78 2008)*

T≠0 Phase diagram of the F-AF model

Domenge, Messio et al PRB 2008



The chiral phase transition:

- ✓ weakly first order at small $J_2/|J_1|$ due to Z_2 vortices
- ✓ Going towards criticality when $J_2/|J_1|$ increases

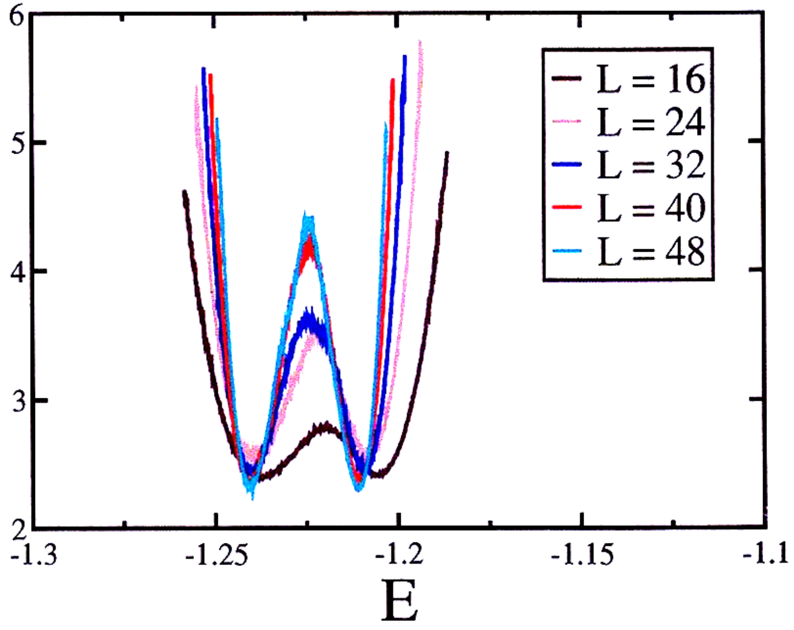
May be not too rare in frustrated magnets

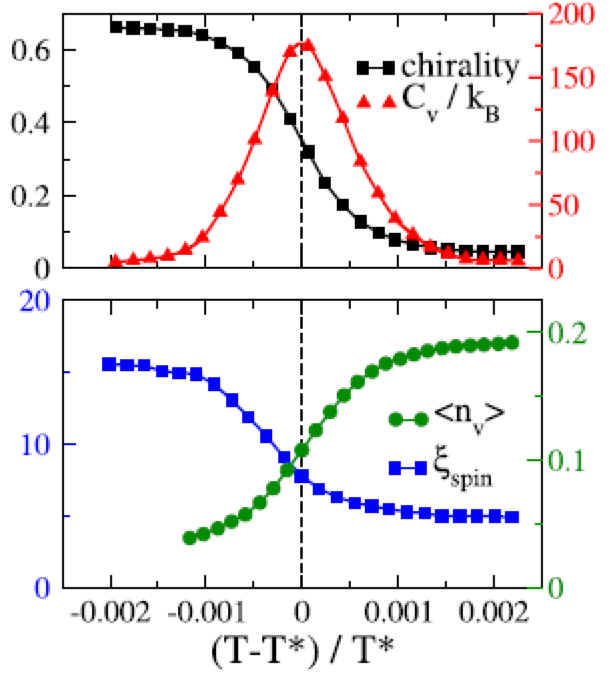
Cyclic 4-spin exchange model *Momoi et al PRL 97*

Unfortunately $Cu_3(titmb)_2(OCOCH_3)_6.H_2O$ undergoes a ferromagnetic transition at 0.05 Kelvin. (3d effect) Y. Karaki (2008)

As γ- Cu2 (OH)3 Cl (*Kageyama et al*. 2001)

A very weak first order phase transition



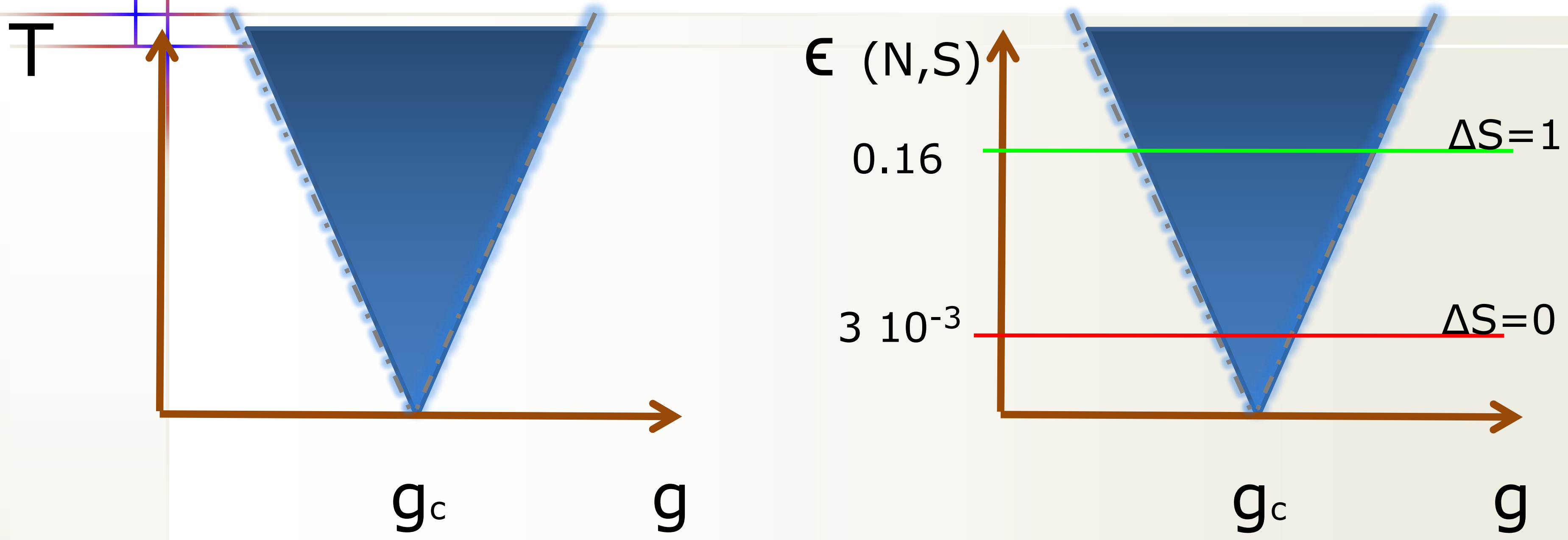


Free energy histogram

$J_2 / |J_1| = 0.38$

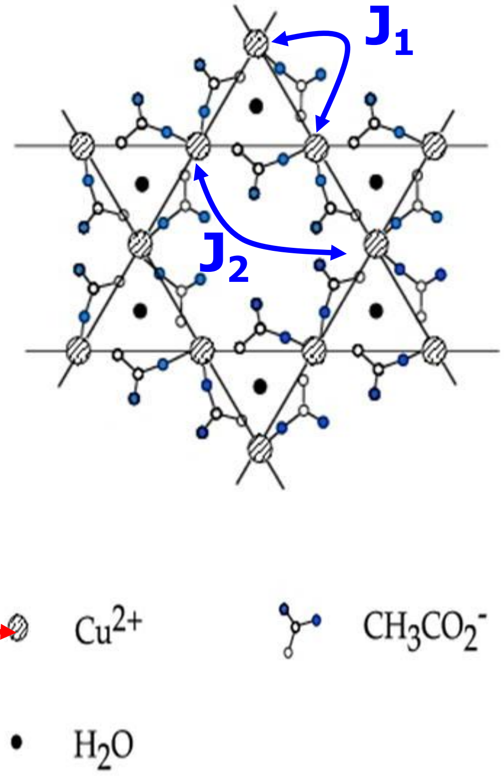
$<n_v>$ = density of Z_2 point defects of the continuous spin texture (green colour above)

QCP and Quantum critical regime



On a finite sample due to total spin quantization there is an infrared cut off to magnetic excitations that can be probed:
in the KAH pb this low energy cut off is 0.16

$Cu_3(titmb)_2(OCOCH_3)_6.H_2O$
AF Heisenberg magnet on kagomé lattice?

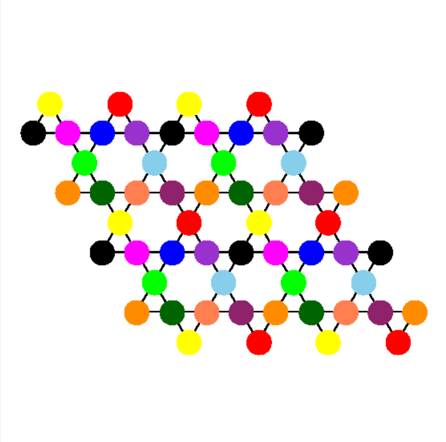


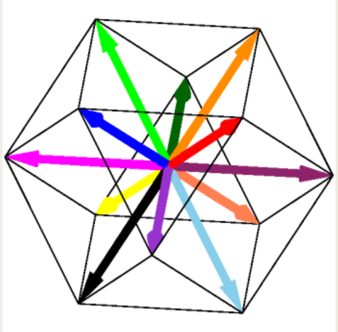
S=1/2

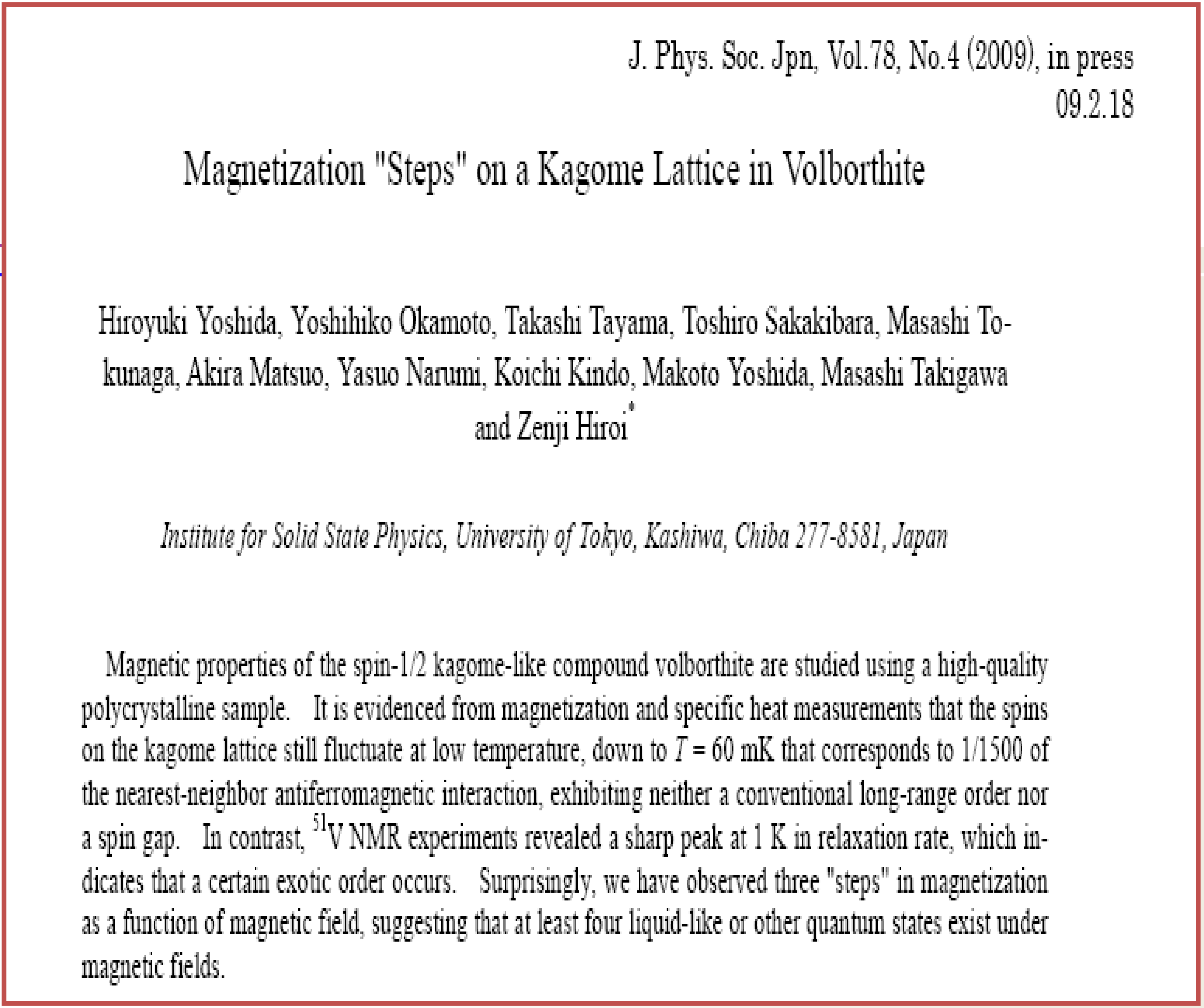
Liu *et al., Inorg. Chem.* (1999)

Honda *et al., J. Phys. Condens. Matter* (2002)
Narumi *et al., Europhys. Lett.* (2004)

$J_1 \sim -19K$ $J_2 \sim 6K$







J. Phys. Soc. Jpn, Vol.78, No.4 (2009), in press
09.2.18

Magnetization "Steps" on a Kagome Lattice in Volborthite

Hiroyuki Yoshida, Yoshihiko Okamoto, Takashi Tayama, Toshiro Sakakibara, Masashi Tokunaga, Akira Matsuo, Yasuo Narumi, Koichi Kindo, Makoto Yoshida, Masashi Takigawa and Zenji Hiroi*

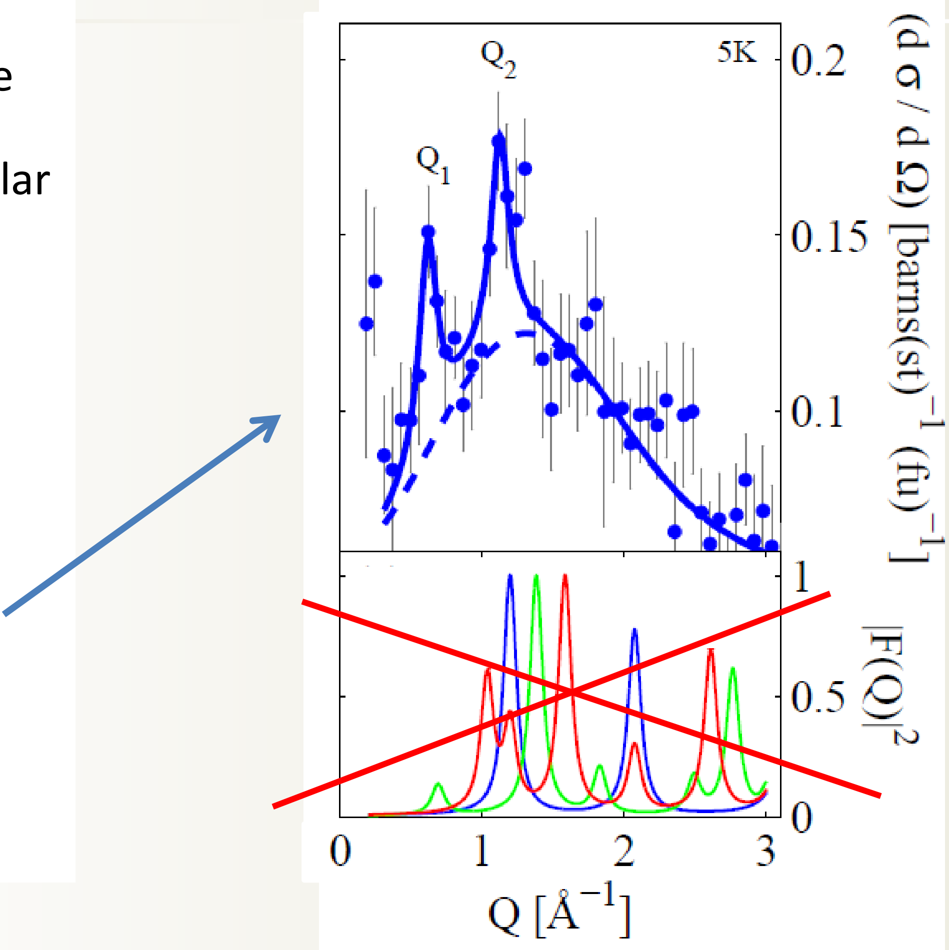
Institute for Solid State Physics, University of Tokyo, Kashiwa, Chiba 277-8581, Japan

Magnetic properties of the spin-1/2 kagome-like compound volborthite are studied using a high-quality polycrystalline sample. It is evidenced from magnetization and specific heat measurements that the spins on the kagome lattice still fluctuate at low temperature, down to $T = 60$ mK that corresponds to 1/1500 of the nearest-neighbor antiferromagnetic interaction, exhibiting neither a conventional long-range order nor a spin gap. In contrast, ^{51}V NMR experiments revealed a sharp peak at 1 K in relaxation rate, which indicates that a certain exotic order occurs. Surprisingly, we have observed three "steps" in magnetization as a function of magnetic field, suggesting that at least four liquid-like or other quantum states exist under magnetic fields.

Quantum behavior

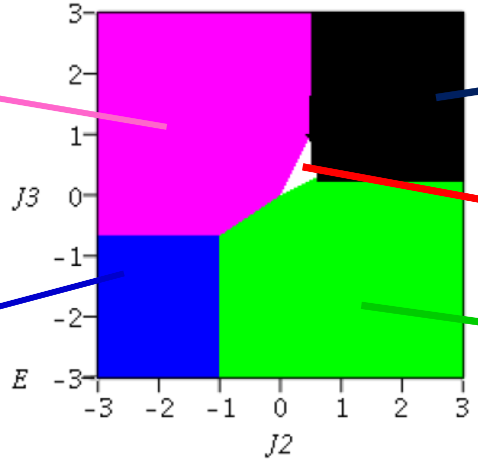
(work in progress L. Messio)

- Chiral spin liquids can exist
  - The J_1-J_2-J_3 models with competitive interactions on the kagome lattice
  - the MSE model on the triangular lattice
- Experimental indications of non coplanar SRO in Vollborthite
  - Z. Hiroi's group J. of Phys. Soc. Jpn 78, 2009,
  - G. Nilsen & al. (EPFL 2010) arxiv:1001.2462
- & possibly Kapellasite:
  - B. Fåk & A. Wills (ILL 2010)



Phase diagram of the classical J_1-J_2-J_3 model

J1= 1 (AF)



3-sublat.
Q=0 order

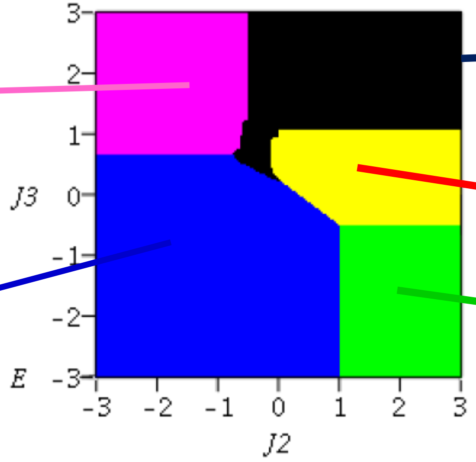
Spiral order

12-sublat. cuboctaedron order

ferromagnet

3-sublat. √3 √3 order

J1= -1 (F)



3-sublat.
Q=0 order

Spiral order

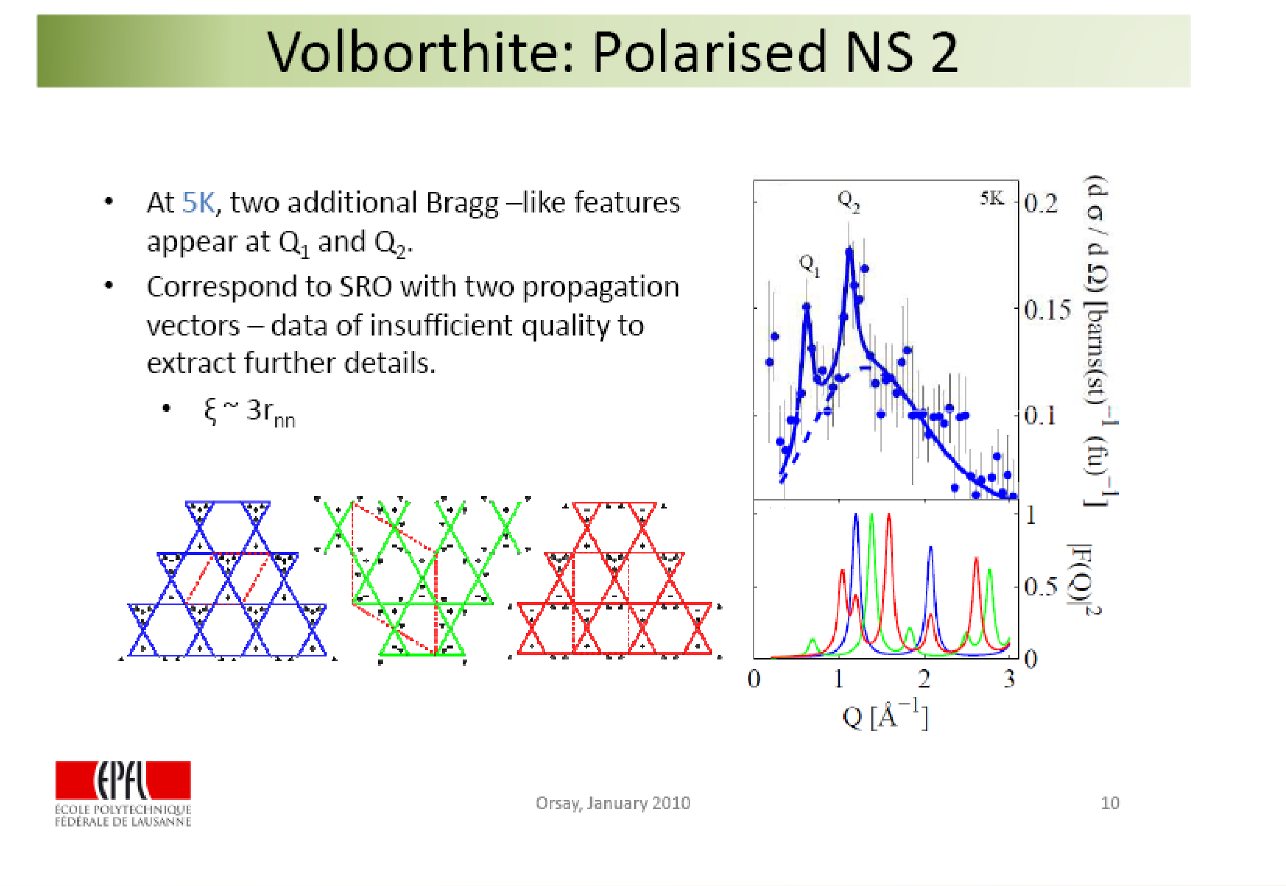
12-sublat. cuboctaedron order

ferromagnet

3-sublat. √3 √3 order

Polarized Neutron experiment

Goran Nilsen 2010



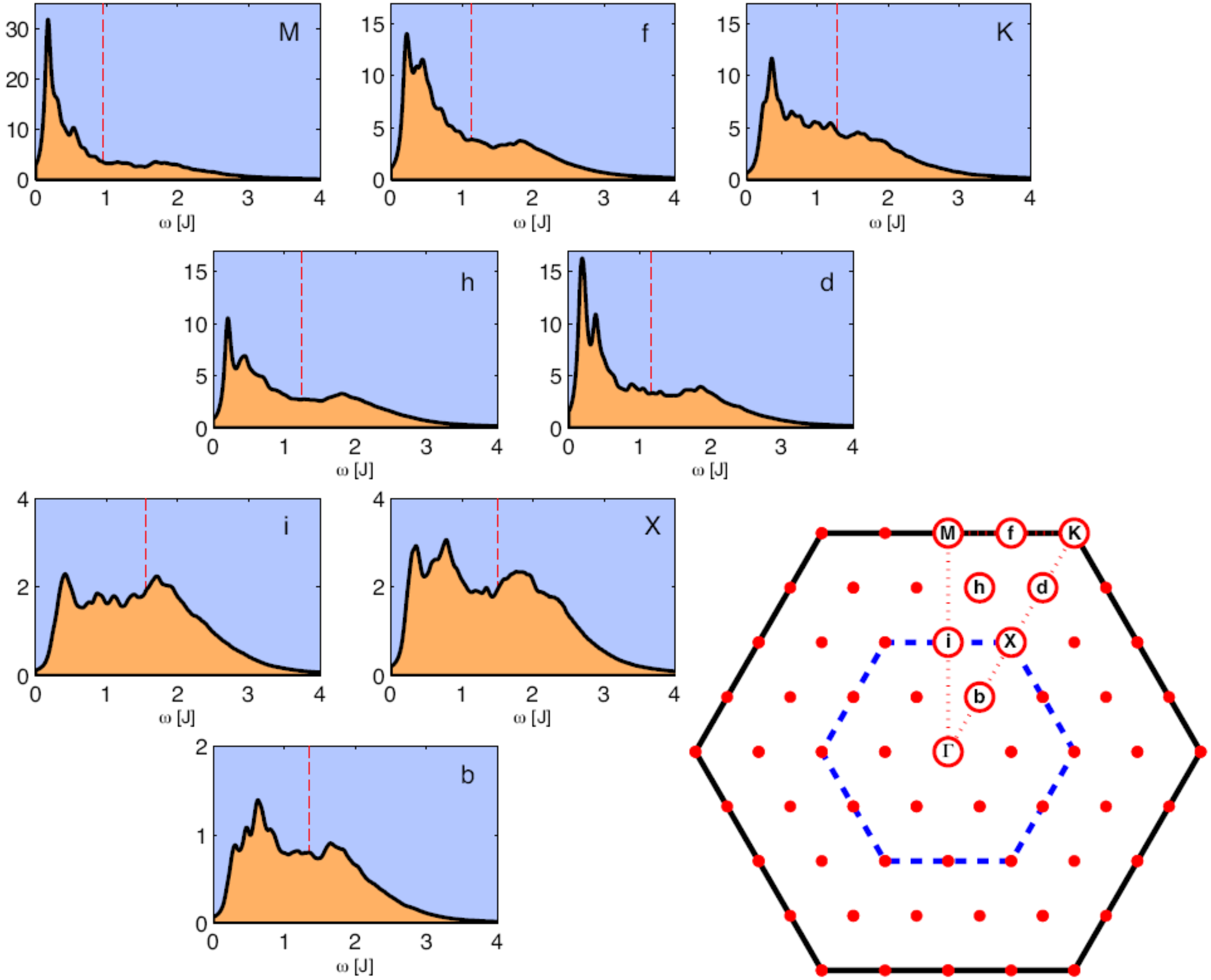
Volborthite: Polarised NS 2

- At 5K, two additional Bragg –like features appear at Q_1 and Q_2.
- Correspond to SRO with two propagation vectors – data of insufficient quality to extract further details.
  - $\xi \sim 3r_{nn}$

Orsay, January 2010

10

Dynamical Structure Factor
Andreas Laeuchli 2007



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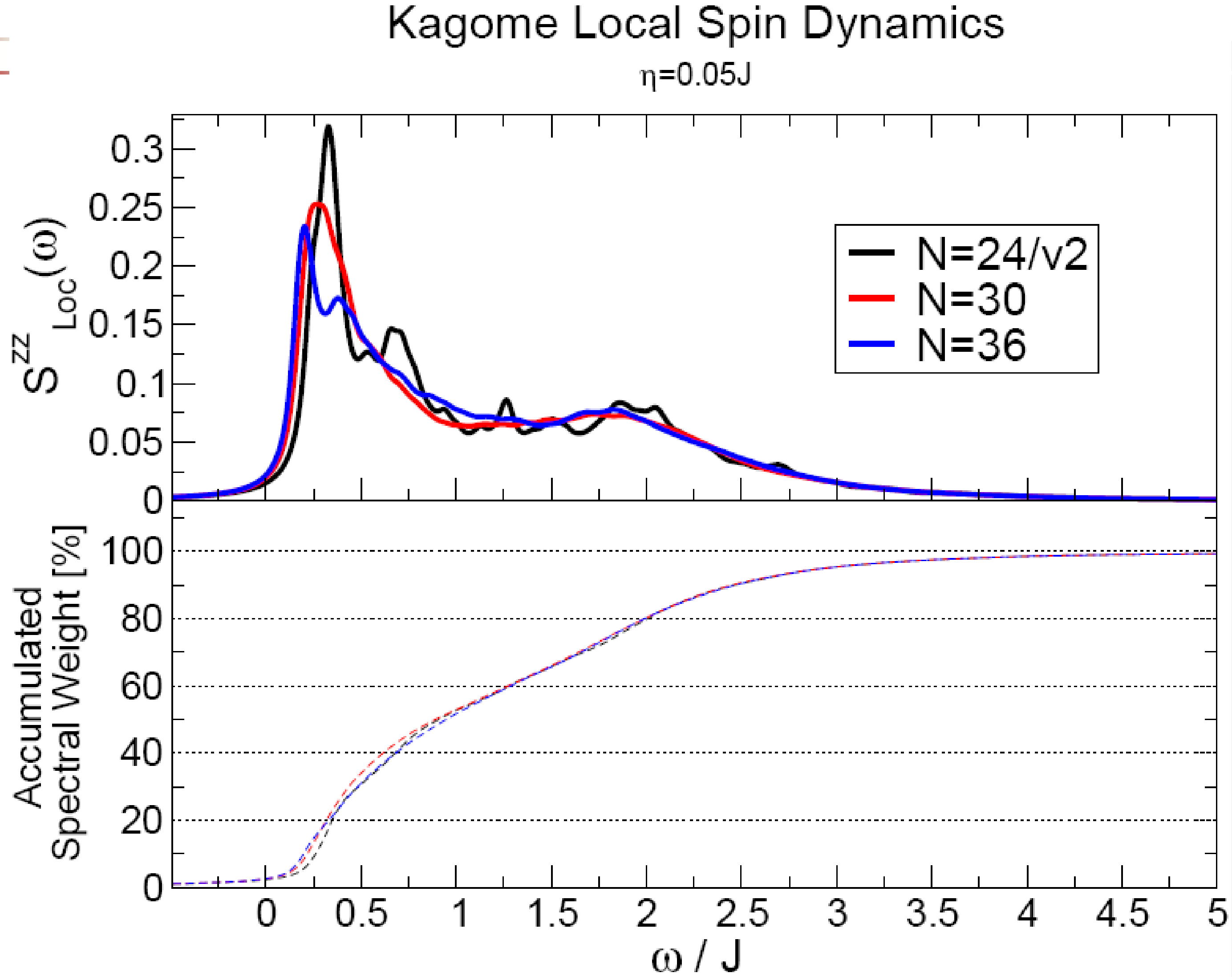
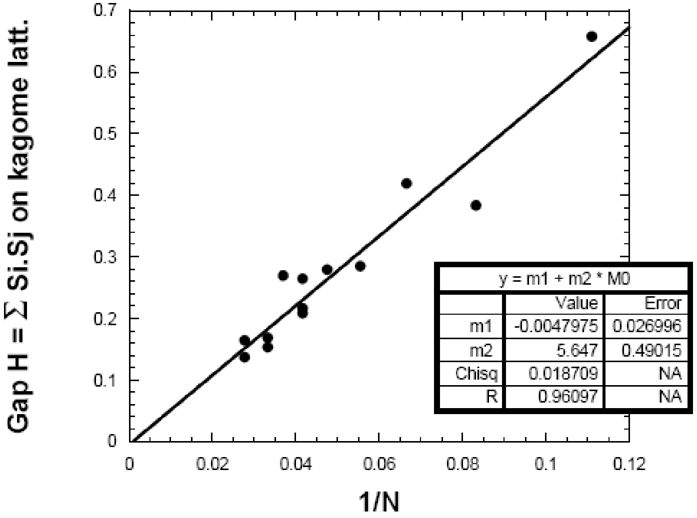
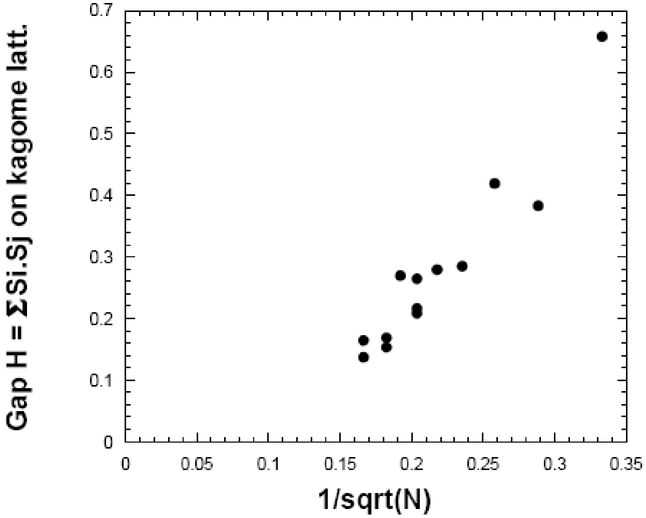


All data (from N=9 to N=36) versus 1/N
P. Sindzingre et C.L.



y = m1 + m2 * M0		
	Value	Error
m1	-0.0047975	0.026996
m2	5.647	0.49015
Chisq	0.018709	NA
R	0.96097	NA

All data from N=9 to N=36 versus 1/sqrt(N)
P. Sindzingre et C.L.



Small digression around ^{3}He

[6] K. Ishida, M. Morishita, K. Yawata, and H. Fukuyama, Phys. Rev. Lett. **79**, 3451 (1997).

[7] R. Masutomi, Y. Karaki, and H. Ishimoto, Phys. Rev. Lett. **92**, 025301 (2004).

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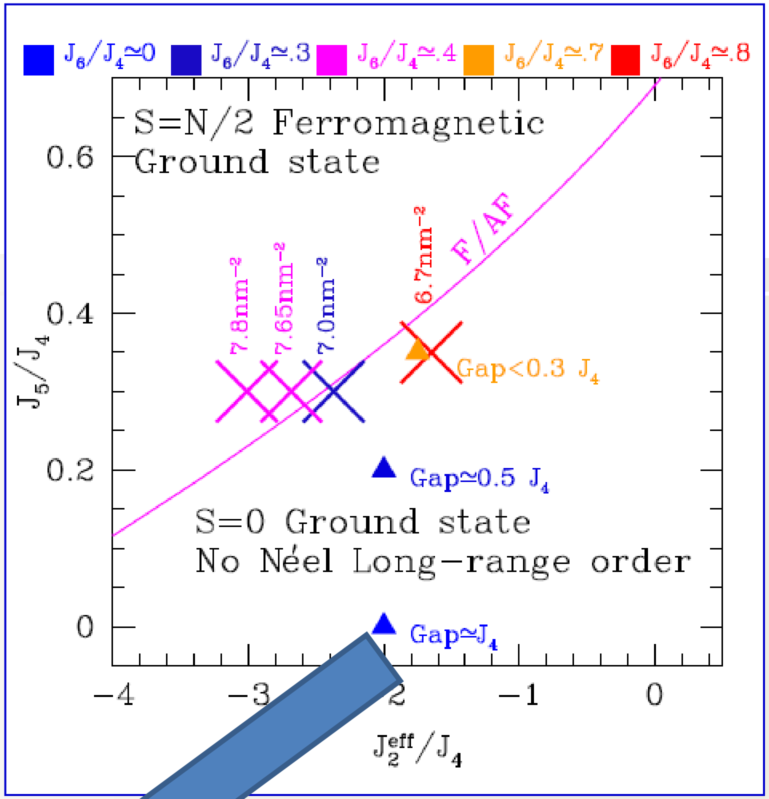
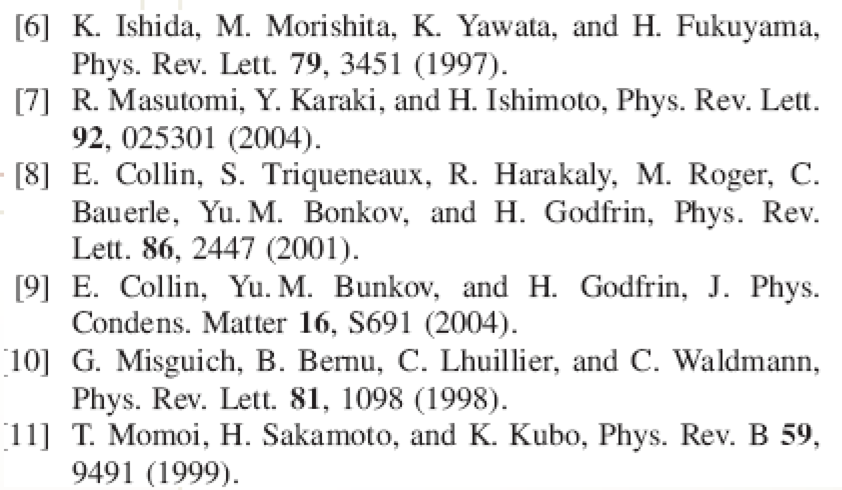
[10] G. Misguich, B. Bernu, C. Lhuillier, and C. Waldmann, Phys. Rev. Lett. **81**, 1098 (1998).

[11] T. Momoi, H. Sakamoto, and K. Kubo, Phys. Rev. B **59**, 9491 (1999).



Gapped Spin Liquid

Small digression around ^{3}He



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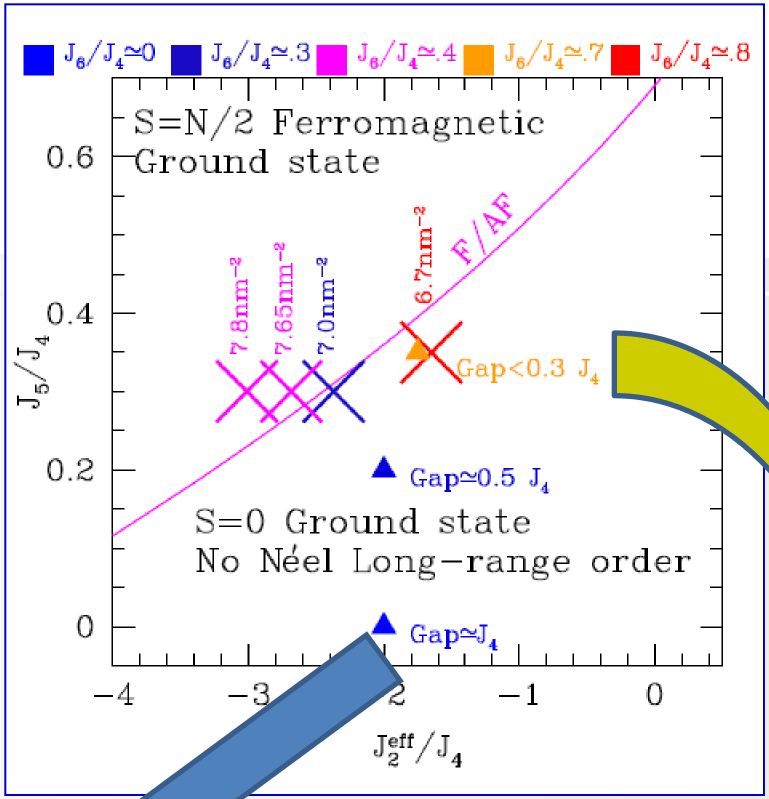
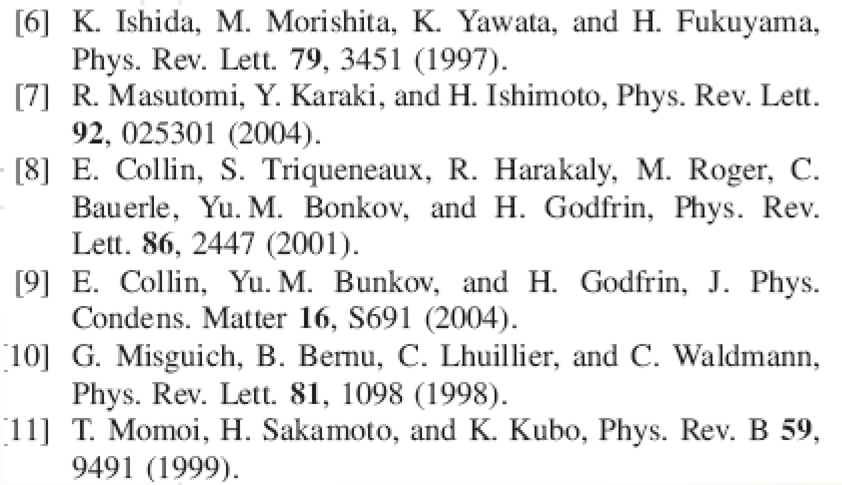


Nematic quadrupolar order near the ferromagnetic phase: *Momoi, Sindzingre, Shannon PRL 2006*

Gapped Spin Liquid

Small digression around ^{3}He



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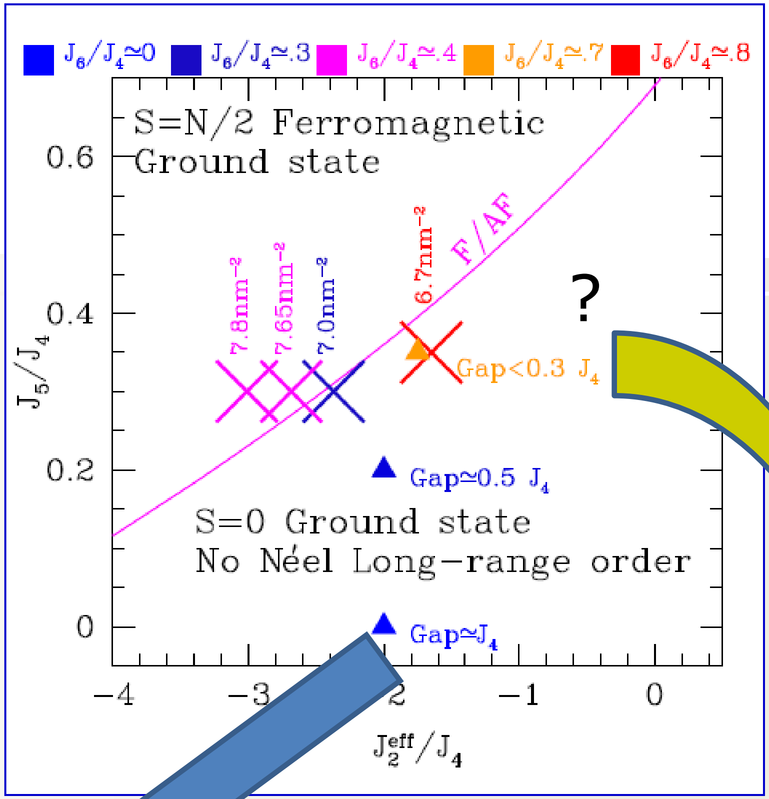

Recent measurement of the m=1/2 plateau:
Nema et al. PRL 2009

Confirm the multi-spin exchange model
But would justify revisiting the values of the coupling constants

Nematic quadrupolar order near the ferromagnetic phase :
Momoi, Sindzingre, Shannon PRL 2006

Gapped Spin Liquid