Quantum Frustrated Magnets: From Theory to Experiments

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Collaborators:
See below for each topic.

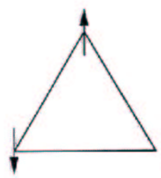
Scope

- **Introduction**
  - Strongly Frustrated Magnets
- **Order by disorder**
  - $J_1 - J_2$ model
  - $LiVOSiO_4$
  - Ising transition and lattice distortion
- **Low-lying singlets**
  - Basic models
  - $Cu_2Te_2O_5Br_2$
- **Magnetization Plateaus**
  - Frustrated ladders
  - $SrCu_2(BO_3)_2$
- **Conclusions**

Strongly Frustrated Magnets

$$H = J \sum_{<i,j>} \vec{S}_i . \vec{S}_j$$

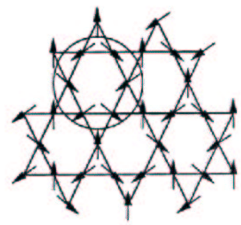
Non bipartite lattice



→ Competition between exchange processes

→ Frustration

Strong frustration (e.g. kagomé)



→ Infinite degeneracy for classical spins

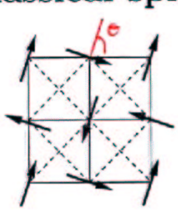
→ Effect of quantum fluctuations?

Order by Disorder

(J. Villain, 1976, for thermal fluctuations)

$$H = J_1 \sum_{<i,j>\ n.n.} \vec{S}_i . \vec{S}_j + J_2 \sum_{<i,j>\ n.n.n.} \vec{S}_i . \vec{S}_j \quad \text{(square lattice,} \quad J_2/J_1 > 1/2)$$

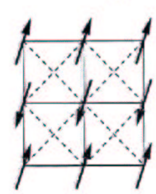
Classical spins



$E_{\text{classical}}$ independent of Θ

→ Classical degeneracy

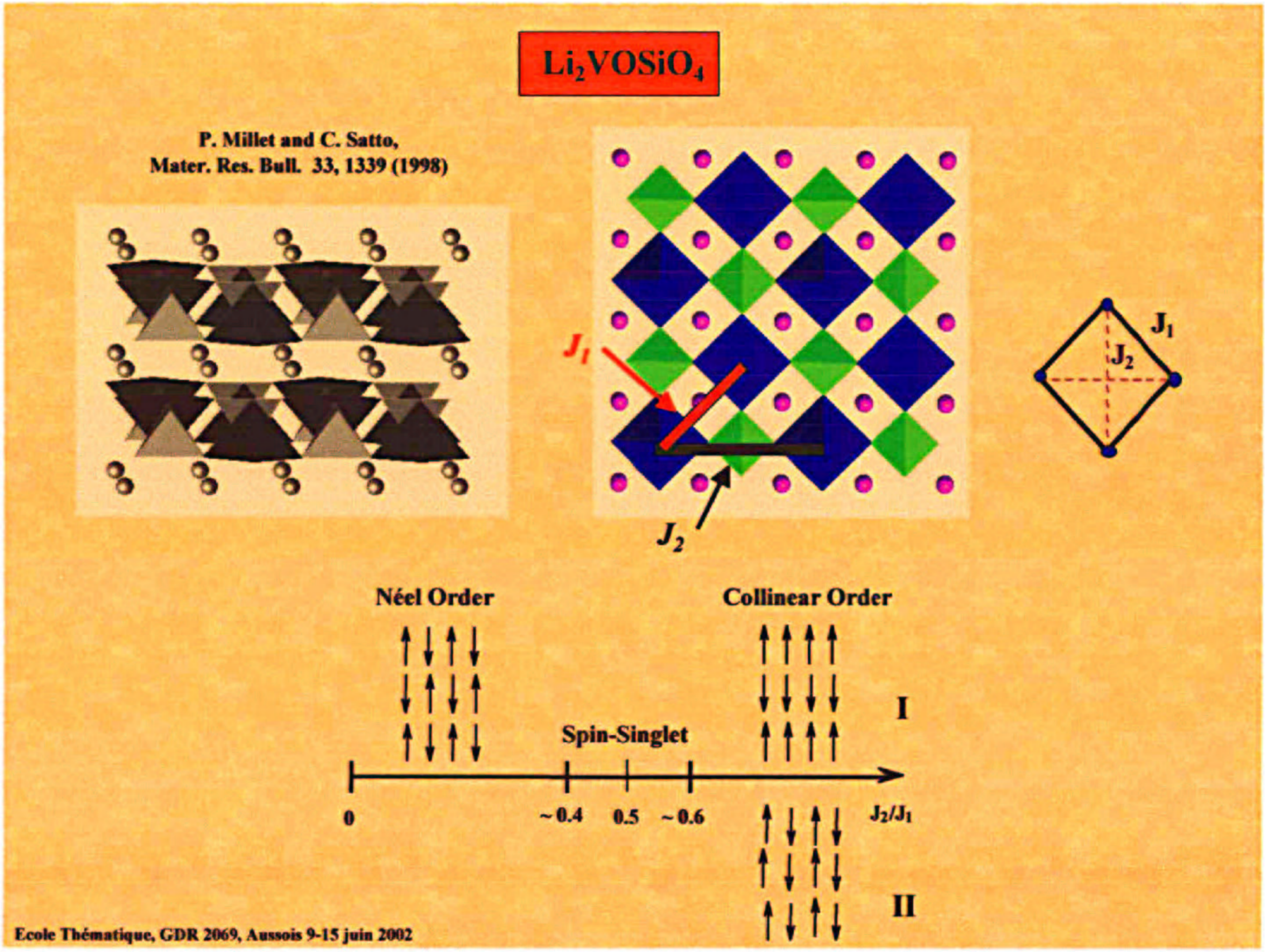
S=1/2



Henley; Chandra and Douçot.

$E \simeq E_{\text{classical}} + \frac{1}{2} \sum_q \omega_q(\theta)$
minimal for $\theta = 0$ or π

→ Helical (collinear) order



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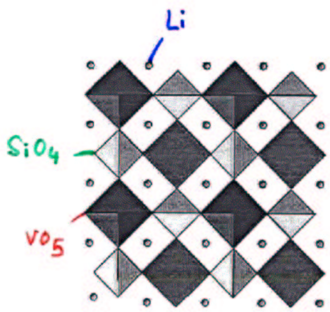
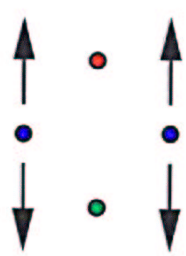
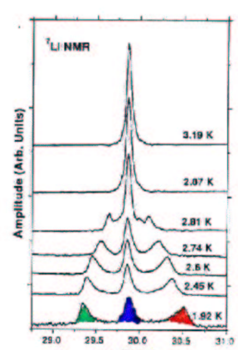
Li_2VOSiO_4

$V^{4+} \rightarrow S = 1/2$ $\theta_{cw} = J_1 + J_2 \simeq 8.2$ K



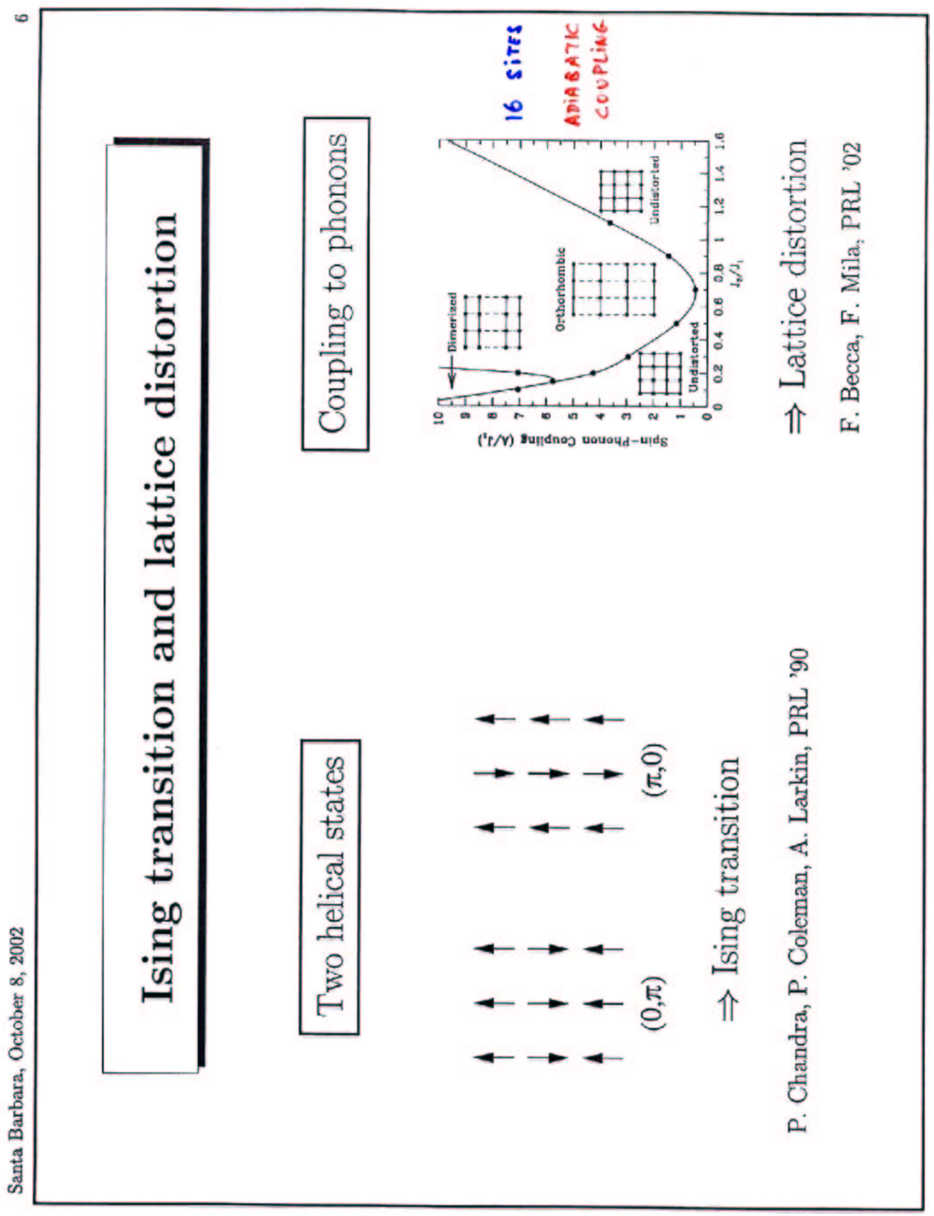




COLLINEAR ORDER BELOW $T_c \simeq 2.83$ K $\Rightarrow$ $J_2/J_1 > 1/2$

R. Melzi, P. Carretta, A. Lascialfari, M. Mambrini, M. Troyer, P. Millet, F. Mila, Phys. Rev. Lett. 85, 1318 (2000)

See also R. Melzi et al, PRB 64, 024409 (2001)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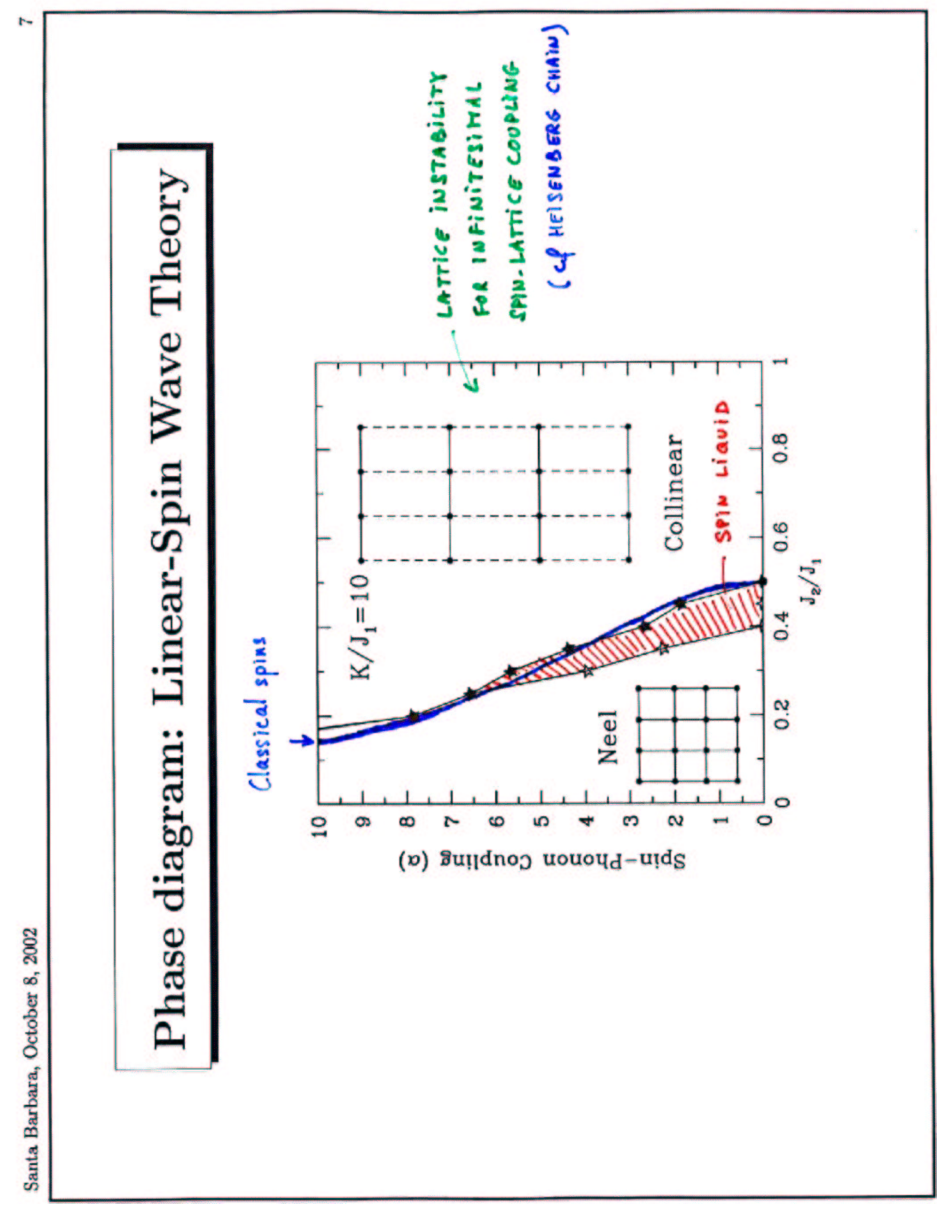
Ising transition and lattice distortion
Two helical states
(0,π)
(π,0)
⇒ Ising transition
P. Chandra, P. Coleman, A. Larkin, PRL '90
Coupling to phonons
16 SITES
ADIABATIC COUPLING
Dimerized
Orthorhombic
Undistorted
Undistorted
Spin-Phonon Coupling (A/J1)
J2/J1
⇒ Lattice distortion
F. Becca, F. Mila, PRL '02
Santa Barbara, October 8, 2002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Phase diagram: Linear-Spin Wave Theory
Classical spins
K/J1=10
Neel
Collinear
SPIN LIQUID
LATTICE INSTABILITY FOR INFINITESIMAL SPIN-LATTICE COUPLING
(cf HEISENBERG CHAIN)
Spin-Phonon Coupling (α)
J2/J1
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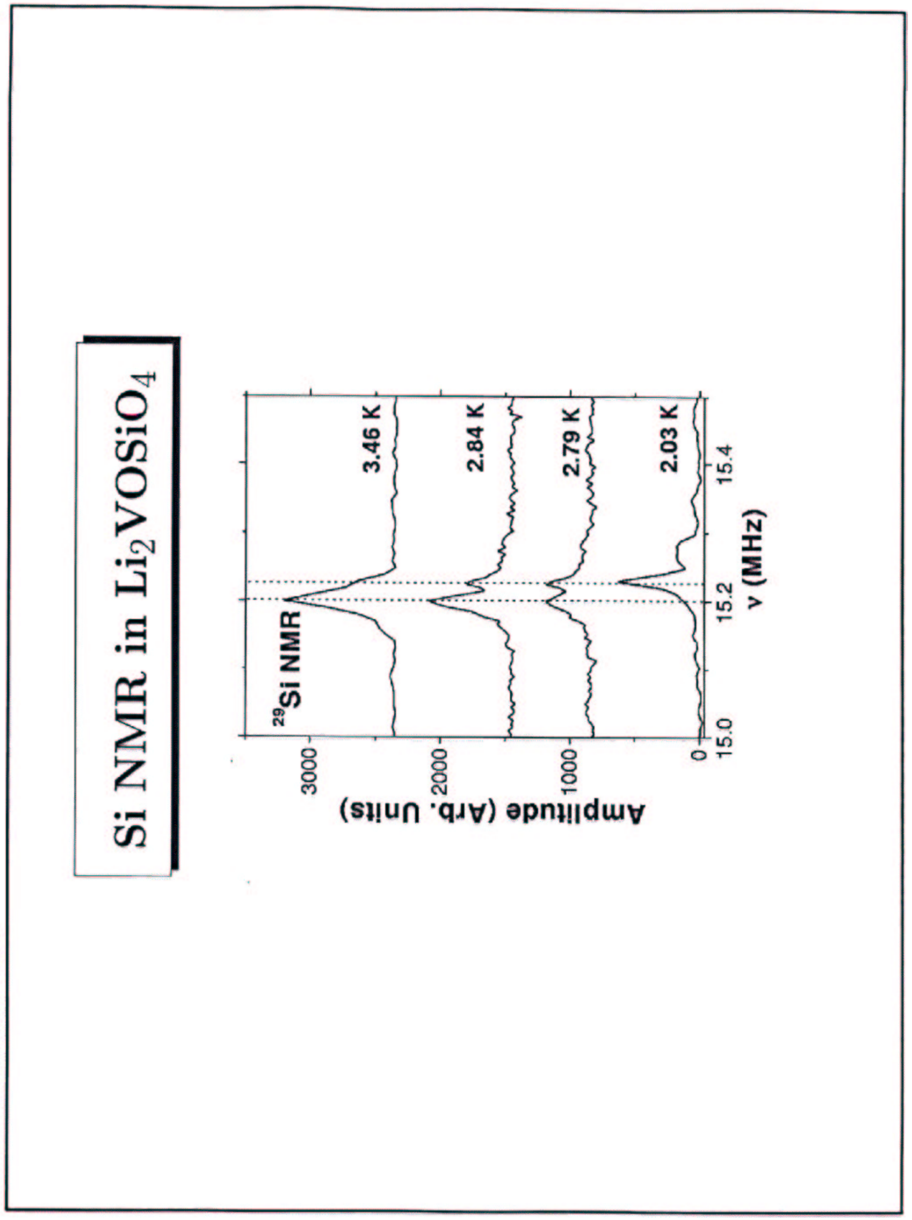
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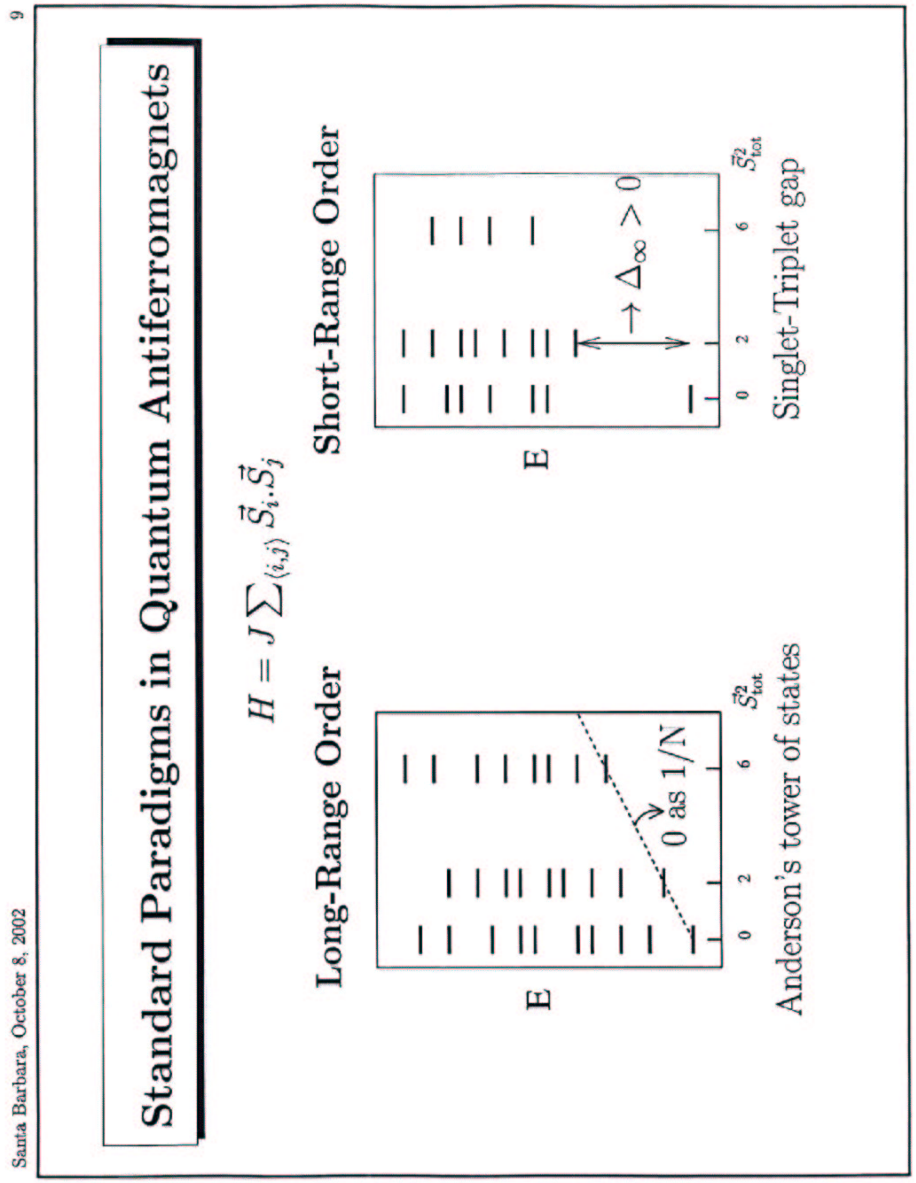
Si NMR in Li_2VOSiO_4
^{29}Si NMR
3.46 K
2.84 K
2.79 K
2.03 K
Amplitude (Arb. Units)
3000
2000
1000
0
15.0
15.2
15.4
ν (MHz)
Santa Barbara, October 8, 2002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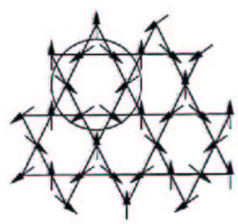
Standard Paradigms in Quantum Antiferromagnets
$H = J \sum_{\langle i,j \rangle} \vec{S}_i.\vec{S}_j$
Long-Range Order
Short-Range Order
E
$\vec{S}^2_{tot}$
0
2
6
0 as 1/N
$\Delta_\infty > 0$
Anderson's tower of states
Singlet-Triplet gap
Santa Barbara, October 8, 2002

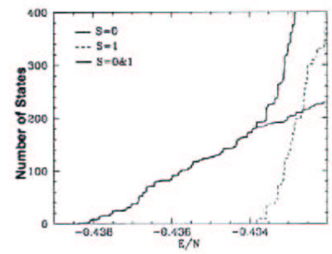
S=1/2 Kagomé antiferromagnet

Classical



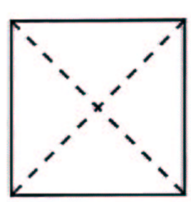
Infinitely degenerate ground state

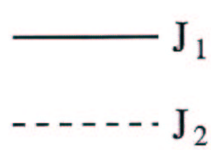
Quantum (S=1/2)

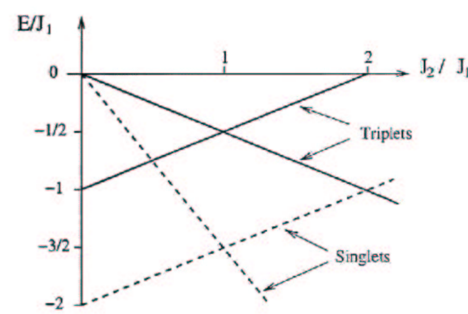


- $\Delta_\infty > 0$
- Proliferation of low-lying singlets(Lecheminant et al, Phys. Rev. B **56**, 2521, 1997).
- RVB interpretation (F. Mila, PRL'98; M. Mambrini and F. Mila, EPJB'00).

Weakly coupled tetrahedra

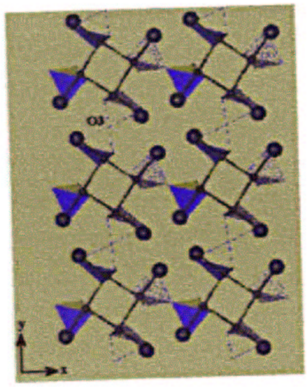


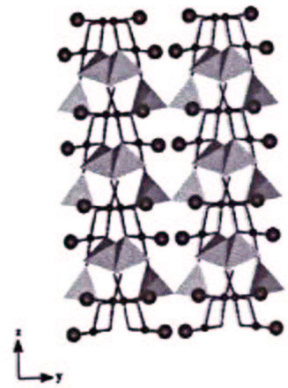




2^N low-lying singlets + Gap to triplet excitations

$Cu_2Te_2O_5X_2$ (X=Cl,Br) : Structure

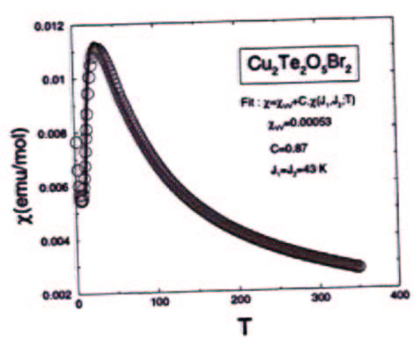


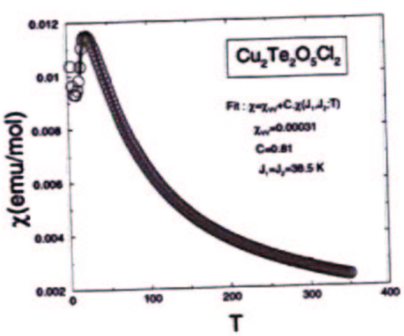


Collaborators:

P. Millet (Toulouse), M. Johnsson (Stockholm), K. Törnroos (Bergen), P. Lemmens (Aachen), C. Geibel (Dresden), K. Becker (Dresden), W. Brenig (Braunschweig), C. Gros, R. Valenti (Saarbrücken)

$Cu_2Te_2O_5X_2$ (X=Cl,Br): Magnetic susceptibility

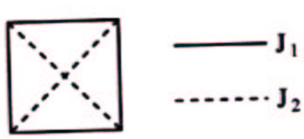




Fit: Isolated plaquettes

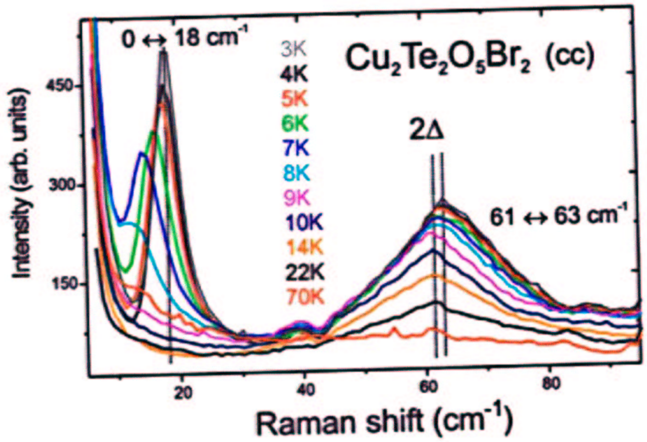
$\rightarrow J_1 = J_2$

Weakly coupled tetrahedra



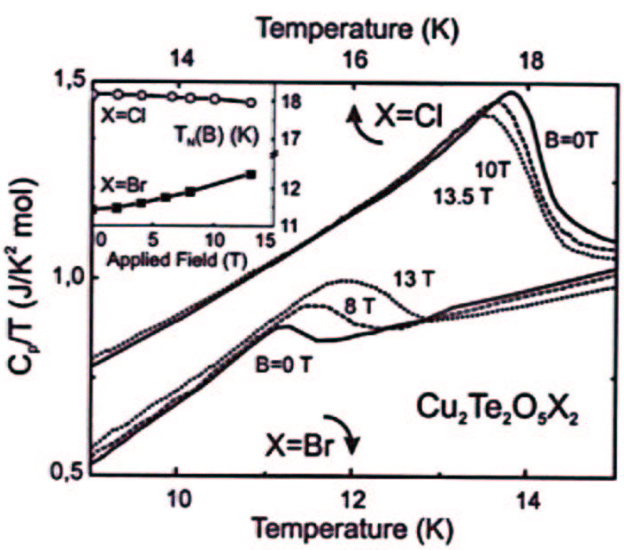
M. Johnsson et al. (Chem. Mat. '00)

$Cu_2Te_2O_5Br_2$: Raman data



P. Lemmens, K.-Y. Choi, E. E. Kaul, C. Geibel, K. Becker, W. Brenig, R. Valenti, C. Gros, M. Johnsson, P. Millet, F. Mila, Phys. Rev. Lett. **87**, 227201 (2001)

$Cu_2Te_2O_5X_2$ (X=Cl,Br): Low-T phase transition



T_c increases with magnetic field!

Ground-state of a tetrahedron

$$H = J(\vec{S}_1.\vec{S}_2 + \vec{S}_1.\vec{S}_3 + \vec{S}_1.\vec{S}_4 + \vec{S}_2.\vec{S}_3 + \vec{S}_2.\vec{S}_4 + \vec{S}_3.\vec{S}_4) = \frac{J}{2}(\vec{S}_{\text{tot}}^2 - 3)$$

Ground-state: $S_{\text{tot}} = 0$

$$\frac{1}{2} \otimes \frac{1}{2} \otimes \frac{1}{2} \otimes \frac{1}{2} = (0 \oplus 1) \otimes (0 \oplus 1) = 0 \otimes 0 \oplus 1 \otimes 1 \oplus 0 \otimes 1 \oplus 1 \otimes 0$$

$$\rightarrow 0 \oplus 0 \oplus 1 \oplus 2 \oplus 1 \oplus 1$$

$\rightarrow$ 2 singlets

1 non-magnetic degree of freedom

$\vec{\tau}$: chirality (Pseudospin $\frac{1}{2}$)

$Cu_2Te_2O_5Br_2$: weakly coupled plaquettes

Phase transition associated with **chirality** pseudo-spin.

$T > T_c$ $J_2 = J_1 \Rightarrow$ tetrahedra

- 2-magnon states at $2J_1$
- 2^{N_t} Singlets $\rightarrow$ quasi-elastic peak.

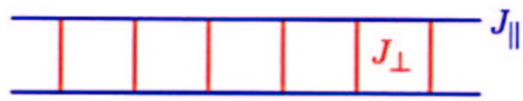
$T < T_c$ $J_2 \neq J_1$ ("Jahn-Teller" distortion - similar to Yamashita and Ueda, PRL 2000)

- Splitting of $\tau = \frac{1}{2}$ and $\tau = -\frac{1}{2}$ (τ = chirality)
- Singlets at finite energy.

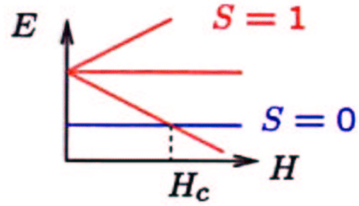
Open issues

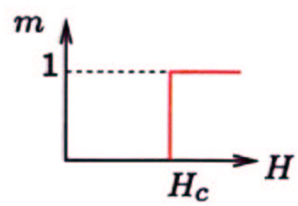
- Residual magnetism (T_c depends on magnetic field,...)
- Pyramid-like shape of 2-magnon states

Ladders in a magnetic field

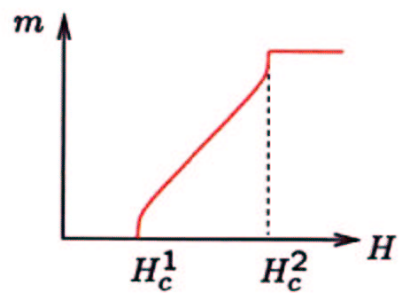


$J_\parallel = 0$





$J_\parallel \neq 0$



Strong-coupling theory: $J_\perp \gg J_\parallel$

F. Mila, EPJ B '98; G. Chaboussant et al, EPJ B '98.

Close to H_c

Two states/rung: $S = 0 \leftrightarrow \sigma^z = |\uparrow\rangle$ $S_z = 1 \leftrightarrow \sigma^z = |\downarrow\rangle$

$$\mathcal{H}_{\text{eff}} = J_{\text{eff}}^{xy} \sum_i (\sigma_i^x \sigma_{i+1}^x + \sigma_i^y \sigma_{i+1}^y) + J_{\text{eff}}^z \sum_i \sigma_i^z \sigma_{i+1}^z - H^{\text{eff}} \sum_i \sigma_i^z$$

$$J_{\text{eff}}^{xy} = J_\parallel \quad J_{\text{eff}}^z = \frac{J_\parallel}{2} \quad H^{\text{eff}} = H - H_c - \frac{J_\parallel}{2}$$

Jordan-Wigner transformation

$$\mathcal{H}_{\text{eff}} = t \sum_i (c_i^\dagger c_{i+1} + \text{h.c.}) + V \sum_i n_i n_{i+1} - \mu \sum_i n_i$$

$$t = \frac{J_\parallel}{2} \quad V = \frac{J_\parallel}{2} \quad \mu = H - H_c$$

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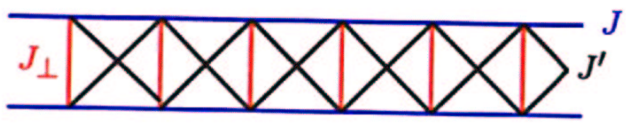
Frustrated Ladders



$J_{\text{eff}}^{xy} = J - J'$ $t = \frac{J-J'}{2}$

$J_{\text{eff}}^{z} = \frac{J+J'}{2}$ $V = \frac{J+J'}{2}$

$H^{\text{eff}} = H - H_c - \frac{J+J'}{2}$ $\mu = H - H_c$

Metal-Insulator transition for $V/t = 2$

⇒ Magnetization plateau for $J' > \frac{J}{3}$

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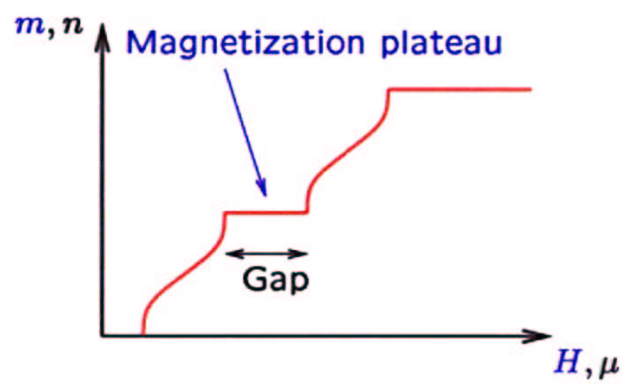
Magnetization Plateau

D. Cabra et al, PRL '97

K. Totsuka, PRB '98

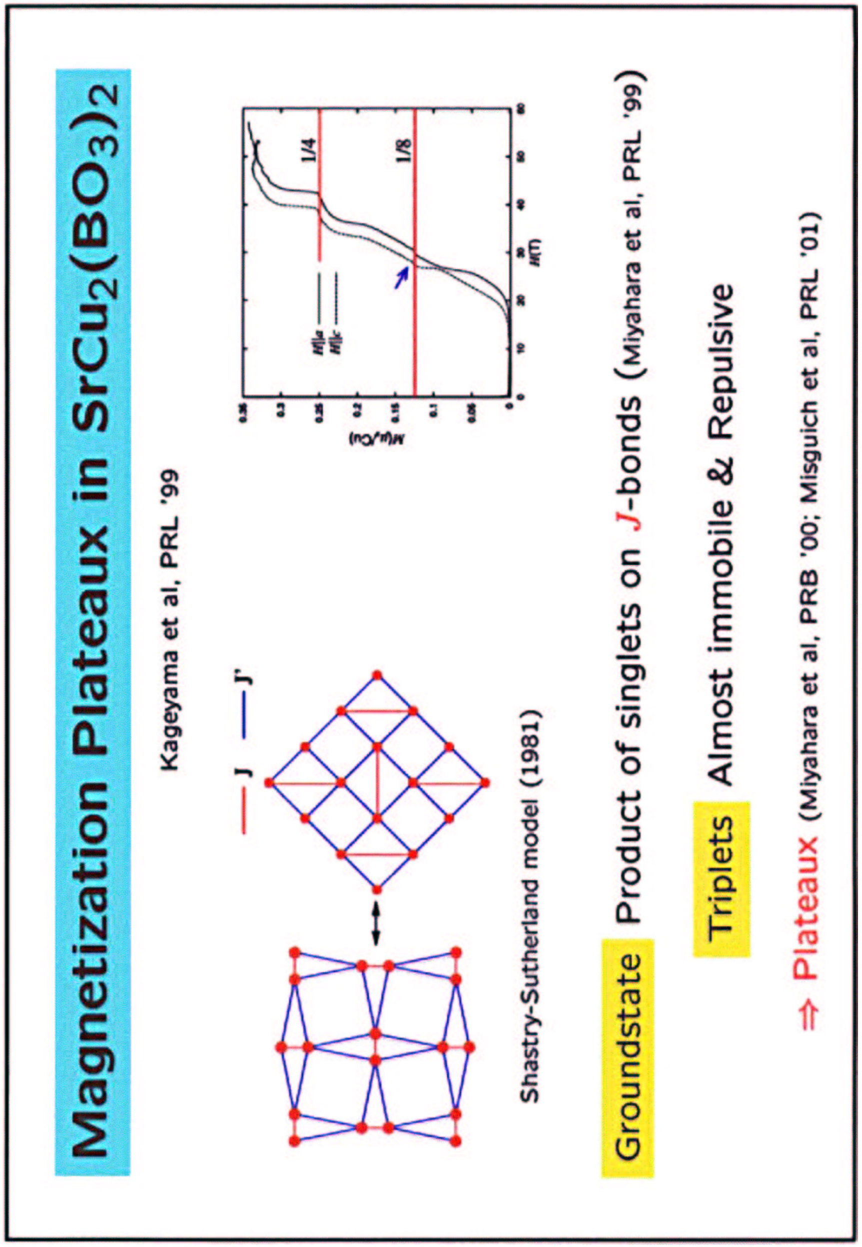
T. Tonegawa et al, PRB '99

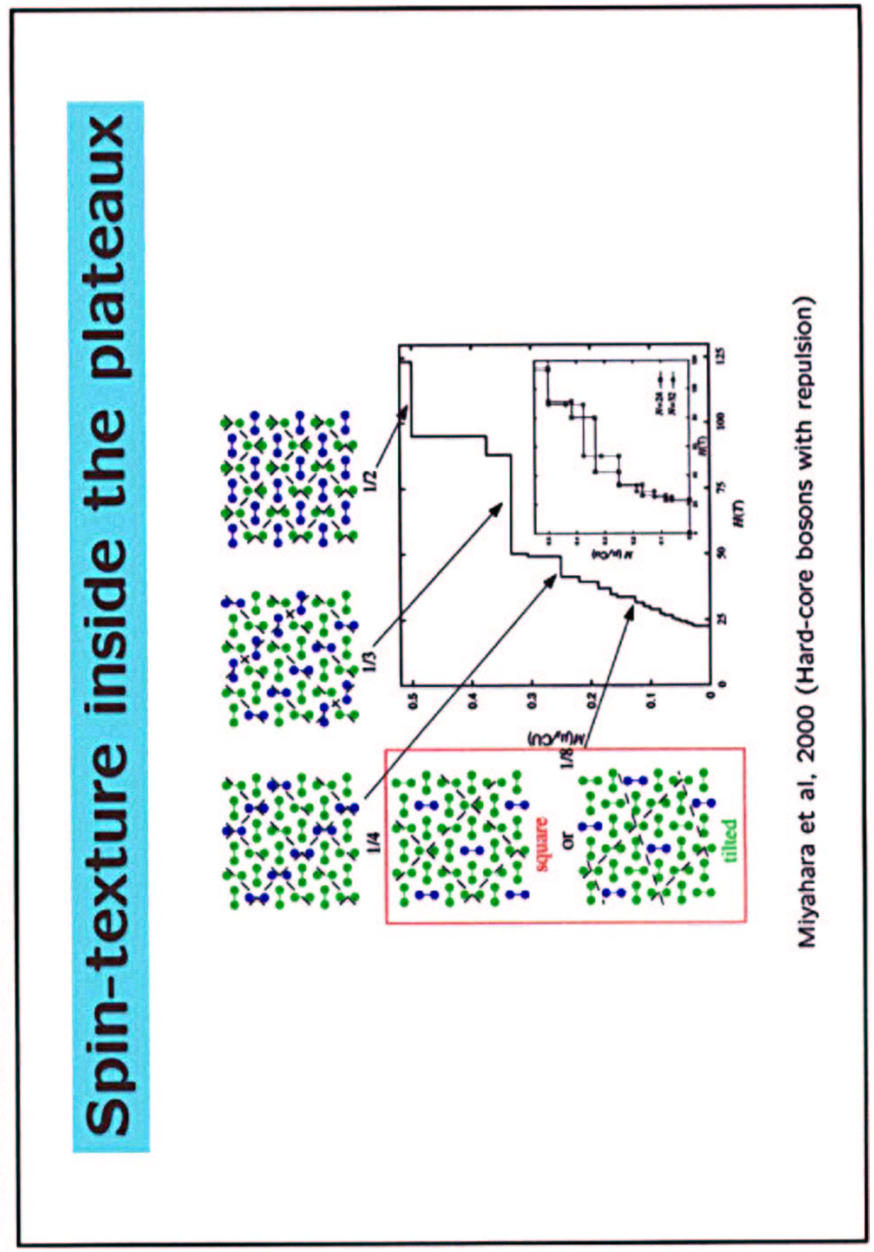
F. Mila, EPJ B '98



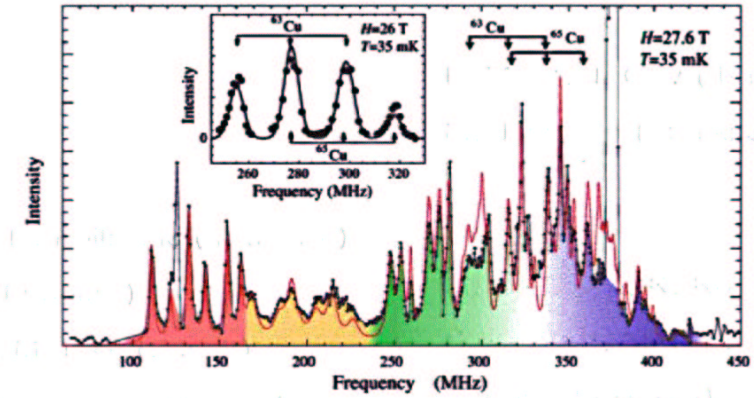
Conclusion

Frustration ⇒ Kinetic energy ↘ & Repulsion ↗ ⇒ Magnetization Plateau


8-èmes JMC, Marseille, 27-30 août 2002
Magnetization Plateaux in $SrCu_2(BO_3)_2$
Kageyama et al, PRL '99
1/4
1/8
M(μB/Cu)
H(T)
J
J'
Shastry-Sutherland model (1981)
Groundstate Product of singlets on J-bonds (Miyahara et al, PRL '99)
Triplets Almost immobile & Repulsive
⇒ Plateaux (Miyahara et al, PRB '00; Misguich et al, PRL '01)


8-èmes JMC, Marseille, 27-30 août 2002
Spin-texture inside the plateaux
1/2
1/3
1/4
1/8
square
or
tilted
Miyahara et al, 2000 (Hard-core bosons with repulsion)

NMR at 1/8-plateau



Kodama, Takigawa, Horvatic, Berthier (unpublished)

At least 11 different sites!

Theory: The Problem

Assumption

Eight-fold degenerate Ground State (thermodynamic limit)

Calculation

Finite system with Periodic Boundary Conditions

↓

Uniform Magnetization!

Solution: break translational symmetry

Coupling to the lattice

S. Miyahara, F. Becca, F. Mila (unpublished)

Motivation

Anomalies of sound velocitiy before plateaux (Wolf et al, PRL '01)

Calculation

Coupling to adiabatic, classical phonons (cf spin-Peierls transition)

Results

GS still 8-fold degenerate, but no coupling between groundstates

→ Spin Texture

Details of the model

Variables = Bond lengths ($\delta_{ij} = d_{ij} - d_{ij}^0$)

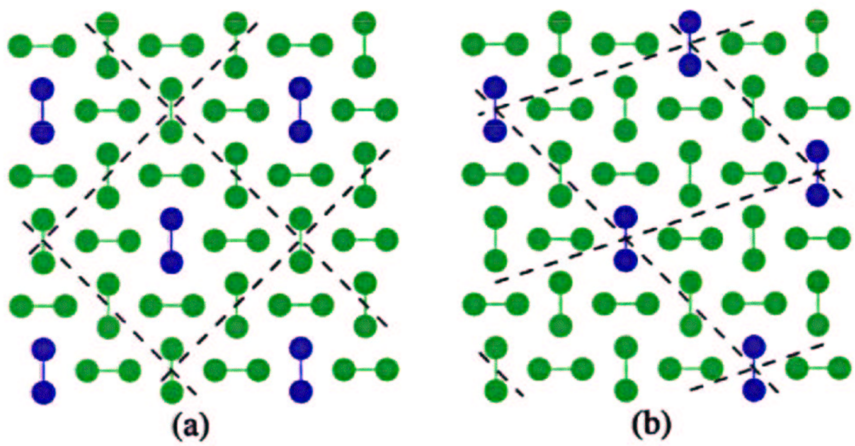
$$\mathcal{H} = \sum_{(i,j)} J_{ij}\, \vec{S}_i . \vec{S}_j + \frac{1}{2} K \sum_{(i,j)} \delta_{ij}^2$$

$$J_{ij} = J_{ij}^0\ (1 - \alpha\ \delta_{ij}) \quad (J \propto \frac{1}{d^\alpha},\ \ \alpha \simeq 7)$$

Minimize total energy with respect to bond lengths

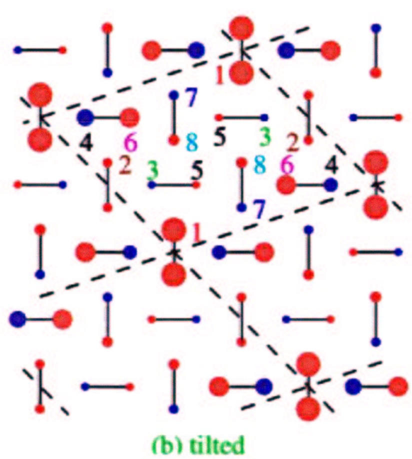
New Periodicity ⇒ Magnetization Pattern

Square vs Tilted



Only 6 different sites 8 with interplane coupling	8 different sites 12 different sites with interplane coupling

Spin texture at 1/8

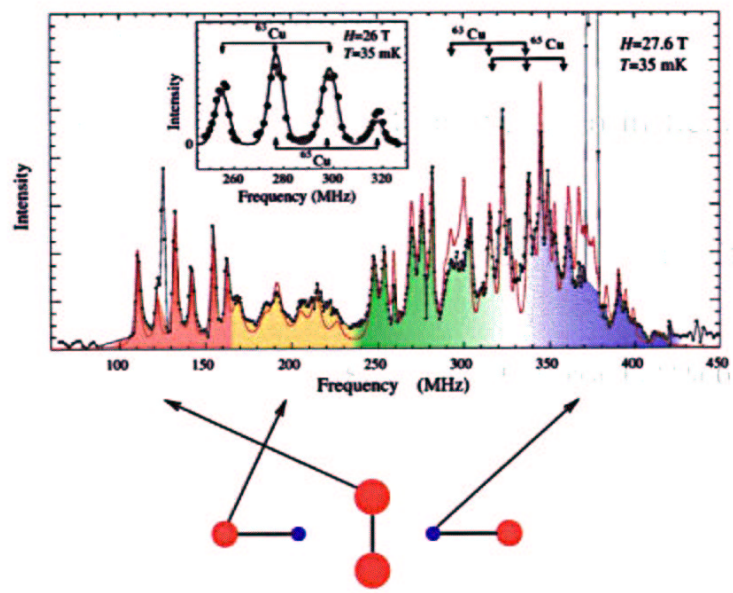


- Magnetization in field direction
- Magnetization opposite to field

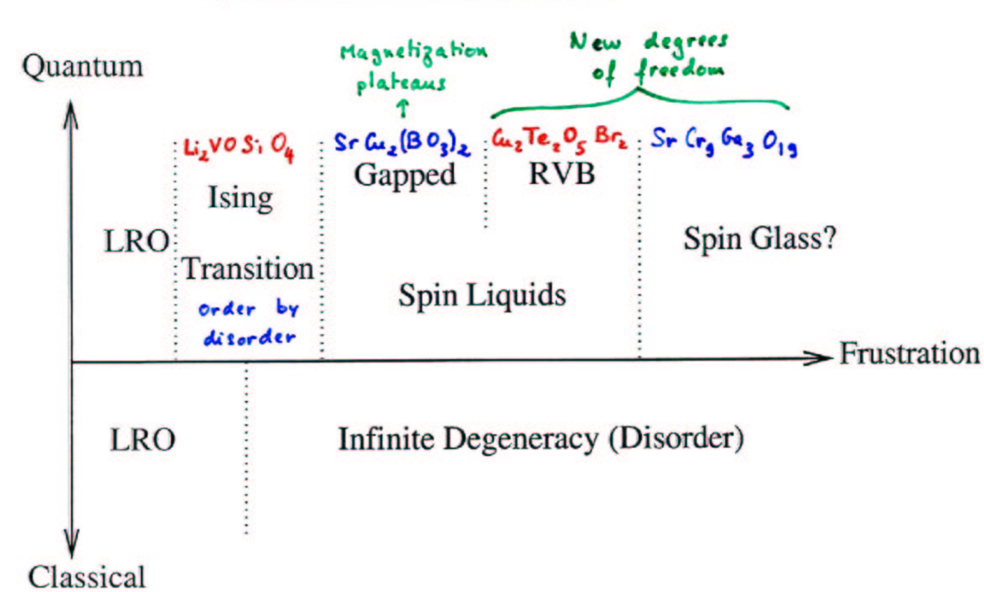
S. Miyahara, F. Becca, F. Mila (unpublished)

Friedel-like oscillations

Comparison with experiment



Conclusions



Collaborators

Theorists

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C. Gros, R. Valenti (Saarbrücken)
A. Honecker, W. Brenig (Braunschweig)

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M. Johnsson (Stockholm)

Experimentalists

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