Frustrated magnetism, magnetization plateaux, and the mystery of $SrCu_2(BO_3)_2$

Solving a 15-year old puzzle

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Ecole Polytechnique Fédérale de Lausanne
Switzerland

Collaborators

Theorists

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K. Schmidt (Dortmund), J. Dorier (Lausanne)
S. Manmana, A. Honecker (Göttingen)
P. Corboz (Amsterdam)

Experimentalists

M. Takigawa, Y. Matsuda (ISSP, Tokyo)
C. Berthier, M. Horvatic, S. Krämer, I. Sheikin,
F. Lévy-Bertrand (Grenoble)

Scope

- A few words about frustrated magnetism

  → degeneracy, enhanced fluctuations, spin liquids

- Magnetization of frustrated magnets

  → plateaus (1/3 on triangular lattice, etc.)

- The intriguing case of $SrCu_2(BO_3)_2$

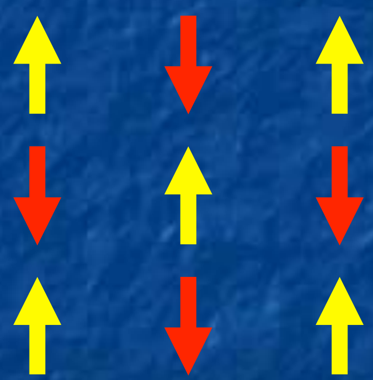
  → plateaus at 1/8, 2/15, 1/6, 1/4, 1/3, 1/2 ?!

- Conclusions

Heisenberg model

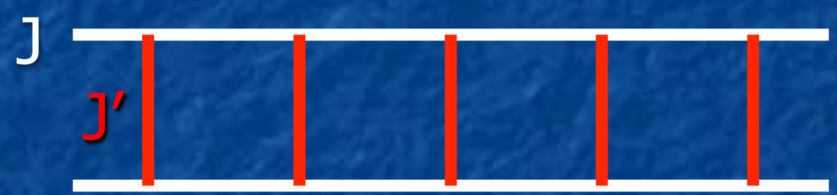
$$\mathcal{H} = \sum_{(i,j)} J_{ij} \, \vec{S}_i \cdot \vec{S}_j$$

Long-range order



Goldstone modes:
Spin waves

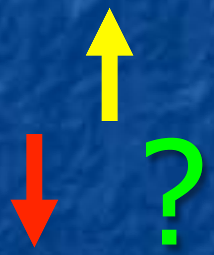
Spin gap



Singlet-triplet gap

$\Delta = J' + O(J) \quad (J \ll J')$

Frustration

Not frustrated  

Frustrated

Antiferromagnetic coupling + odd loops

⬇

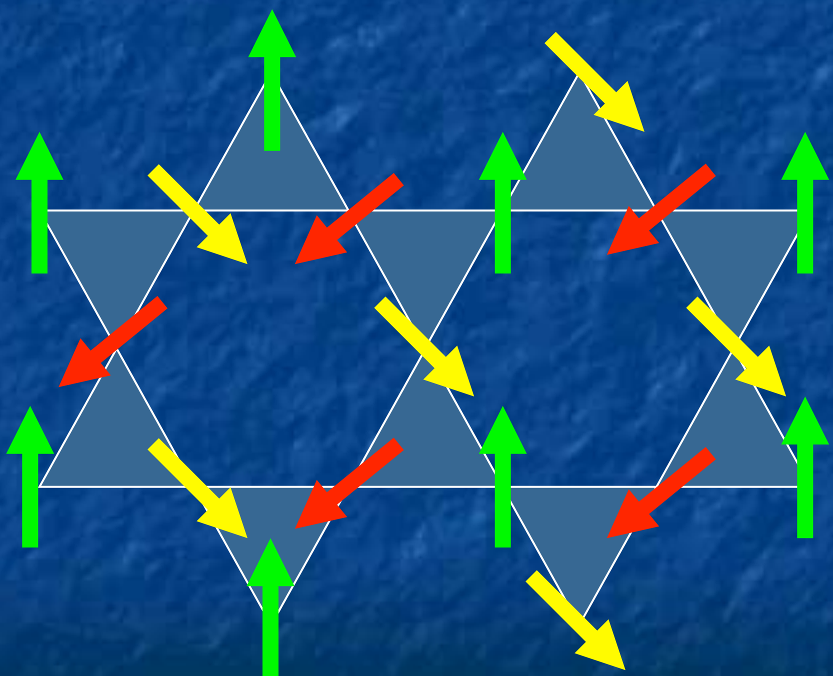
Competition between exchange paths = frustration

Possible consequences of frustration

- Infinite degeneracy of classical ground state (kagome, pyrochlore,…)
- Enhanced quantum fluctuations
  → Resonating Valence Bond spin liquids
  → Algebraic spin liquids
- Magnetization plateaus

Kagome antiferromagnet

Infinite degeneracy



RVB spin liquid?

→ Zeng, Elser, PRB 1995

→ FM, PRL 1998

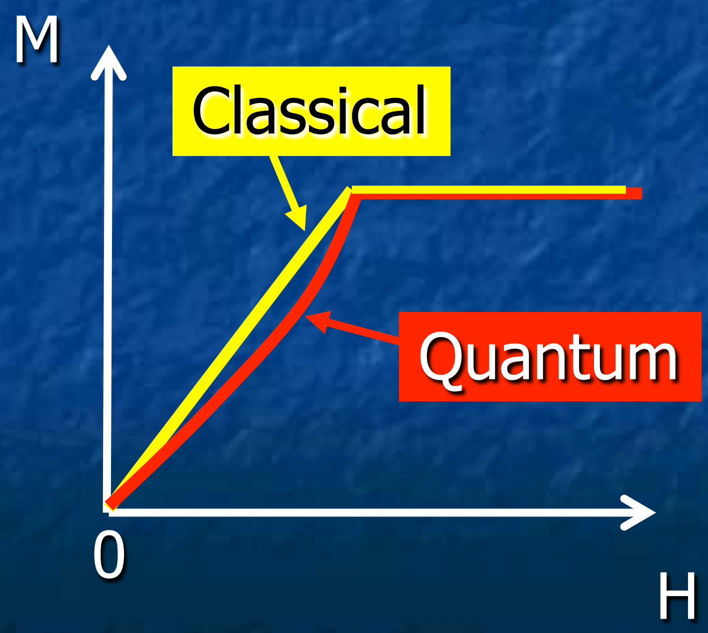
→ Han, Huse White, Science 2011

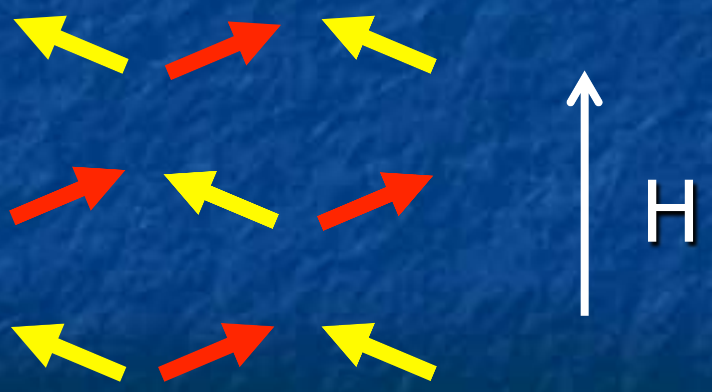
Algebraic spin liquid?

→ Ran, Hermele, Lee, Wen, PRL 2007

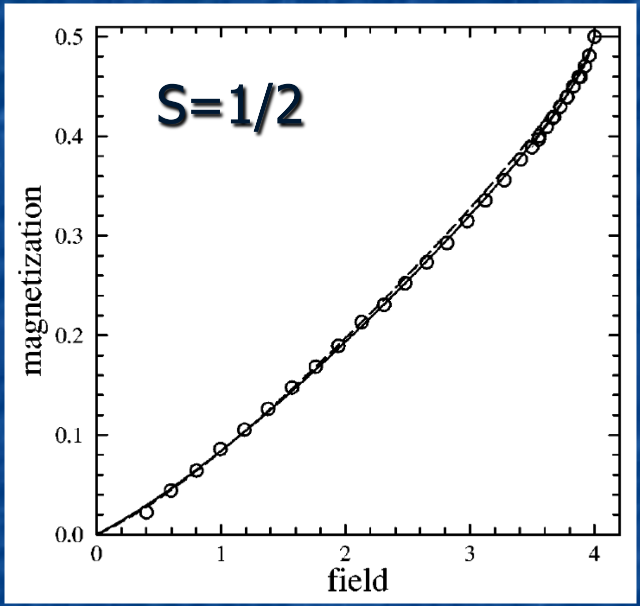
Magnetization of Néel AF

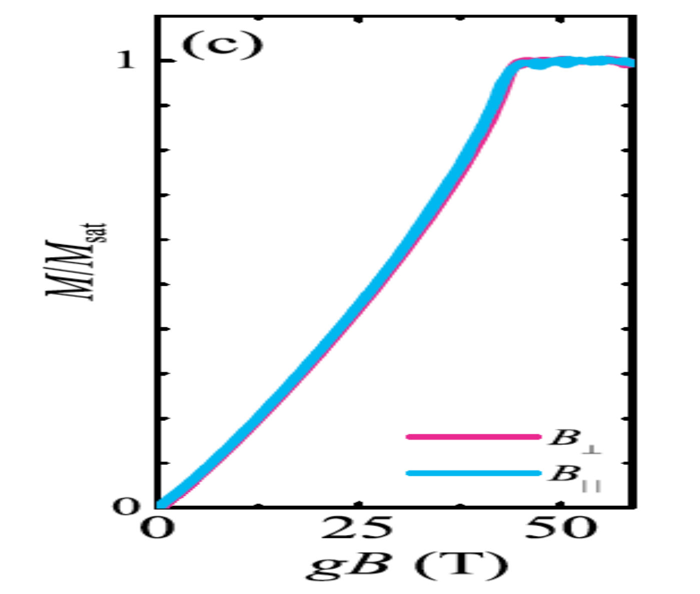
$$\mathcal{H} = J \sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j - g\mu_B H \sum_i S_i^z$$





Square lattice antiferromagnet





- Circles: QMC Igarashi, 1992
- Dashed and solid lines:

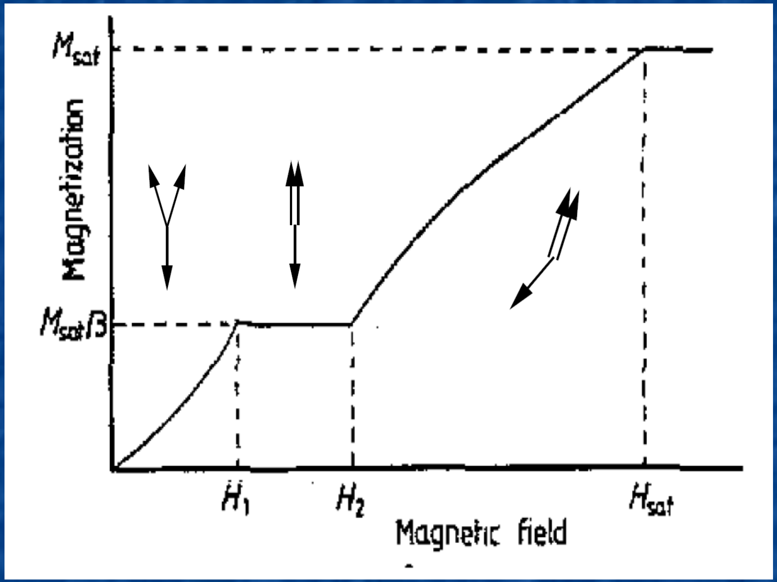
  1st and 2nd order spin-wave

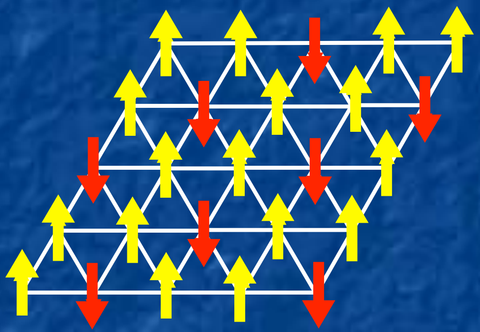
  Zhitomirsky, Nikuni, 1998

$[Cu(HF_2)(pyz)_2]ClO_4$

Goddard et al, 2008

1/3 magnetization plateau



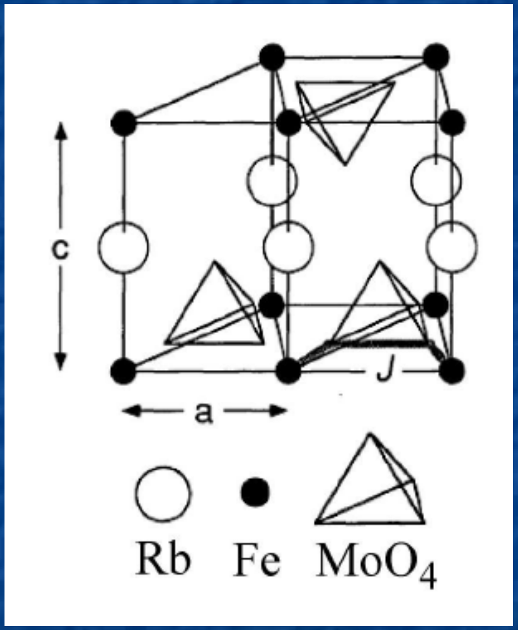


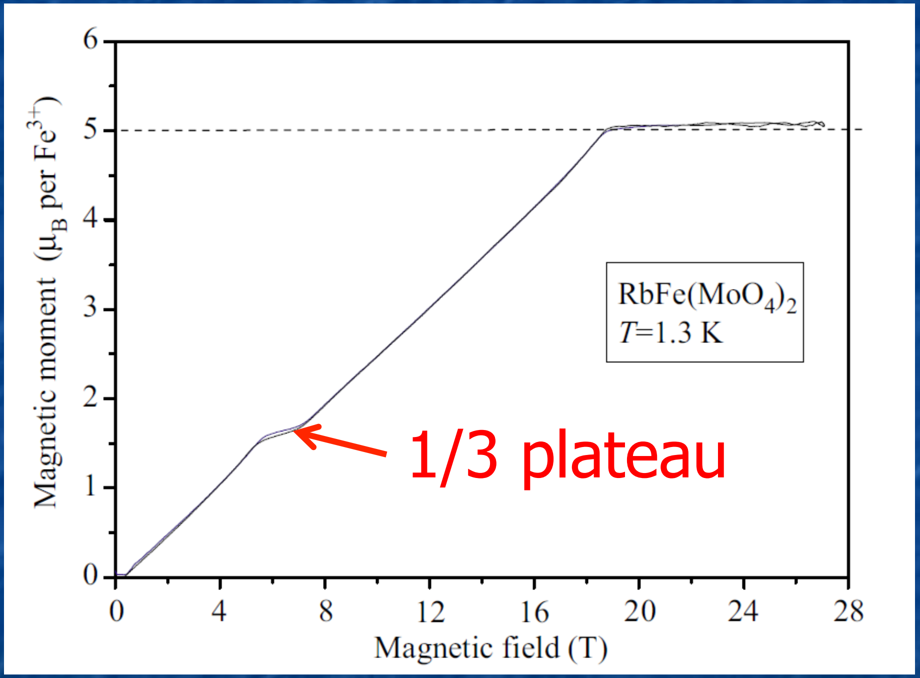
Chubukov and Golosov
JPMC 1991

Collinear structure stabilized in a field range by zero-point fluctuations

$RbFe(MoO_4)_2$

Smirnov et al, PRB 2007

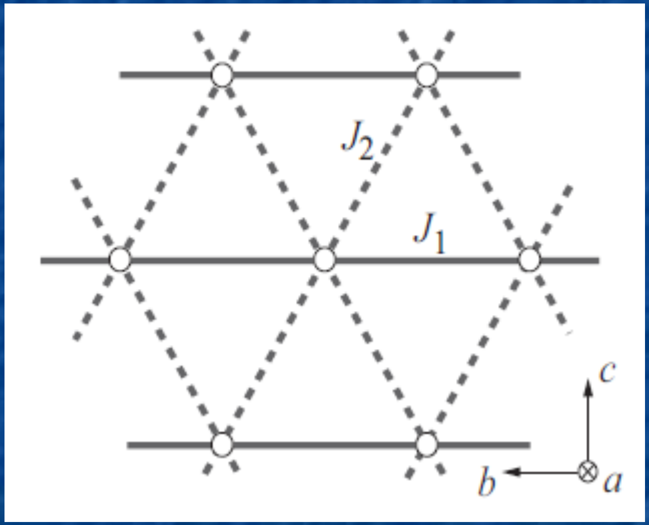




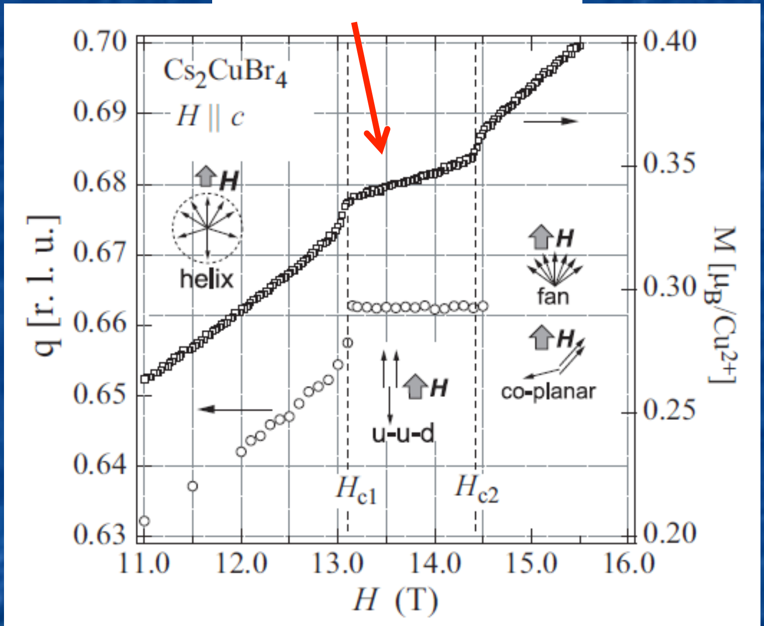
Spin 5/2, triangular lattice

Cs_2CuBr_4

Ono et al,
J. Phys. Cond. Mat. 2004



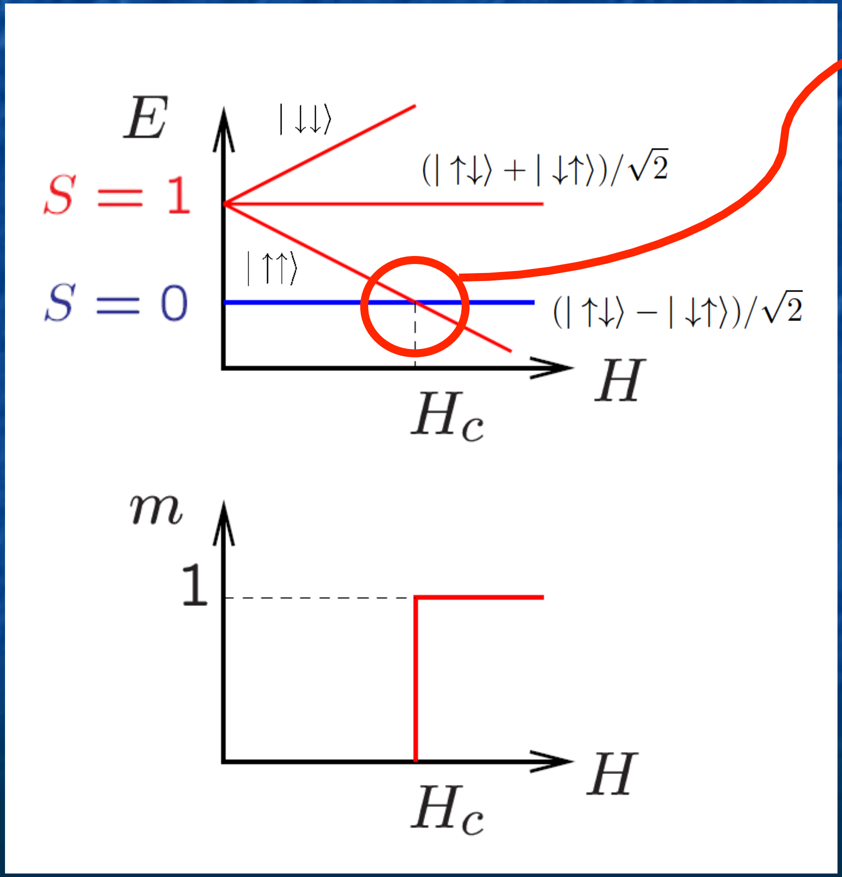
1/3 plateau



Spin 1/2, anisotropic triangular lattice

Gapped systems (ladders,...)

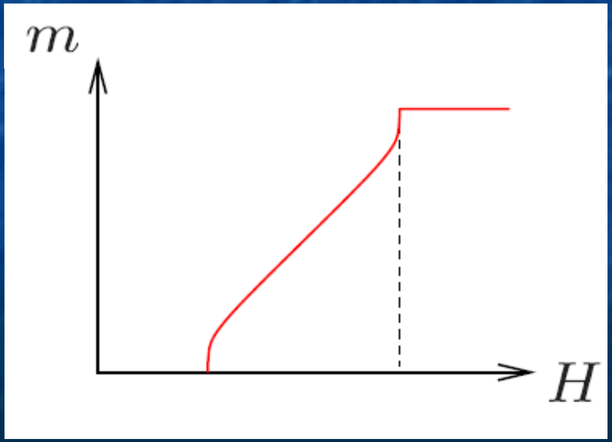
Isolated dimers



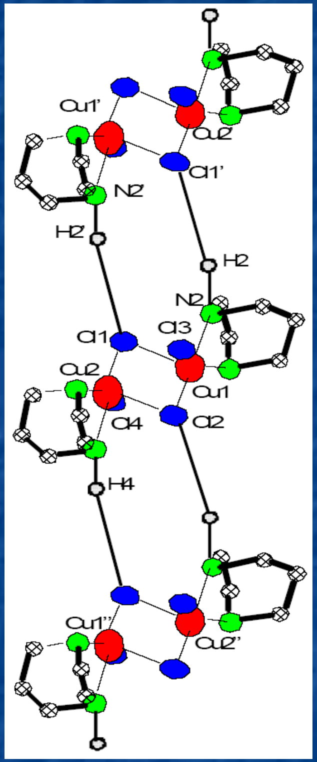
S=0: empty site

S_z=1: boson

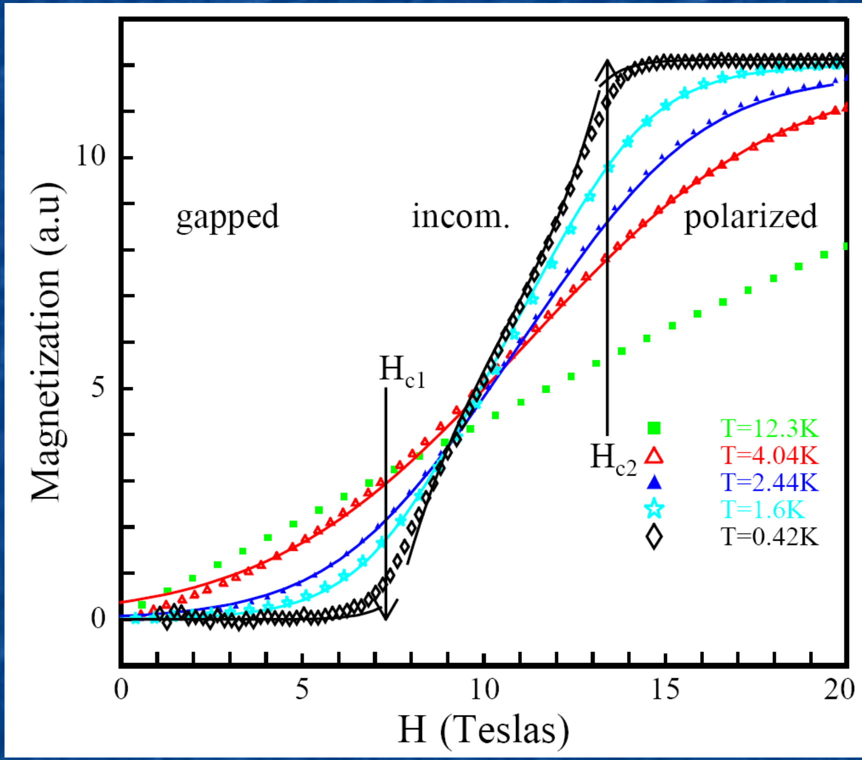
Coupled dimers (ladders,...)



Magnetization of CuHpCl



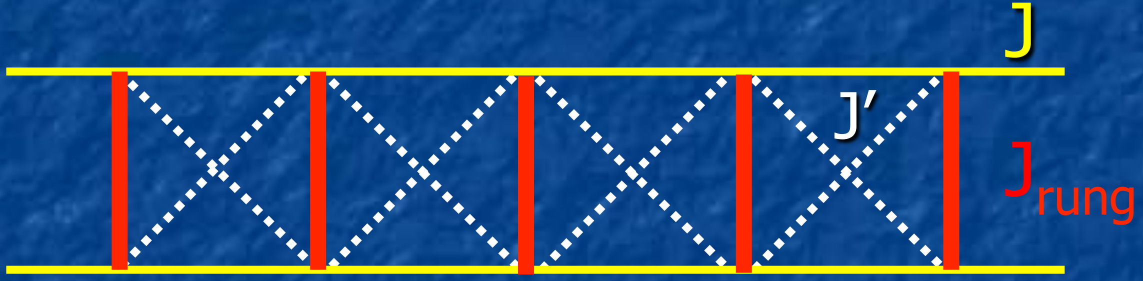
CuHpCl



Chaboussant et al, EPJB '98

No magnetization up to critical field H_{c1}

Frustrated ladder

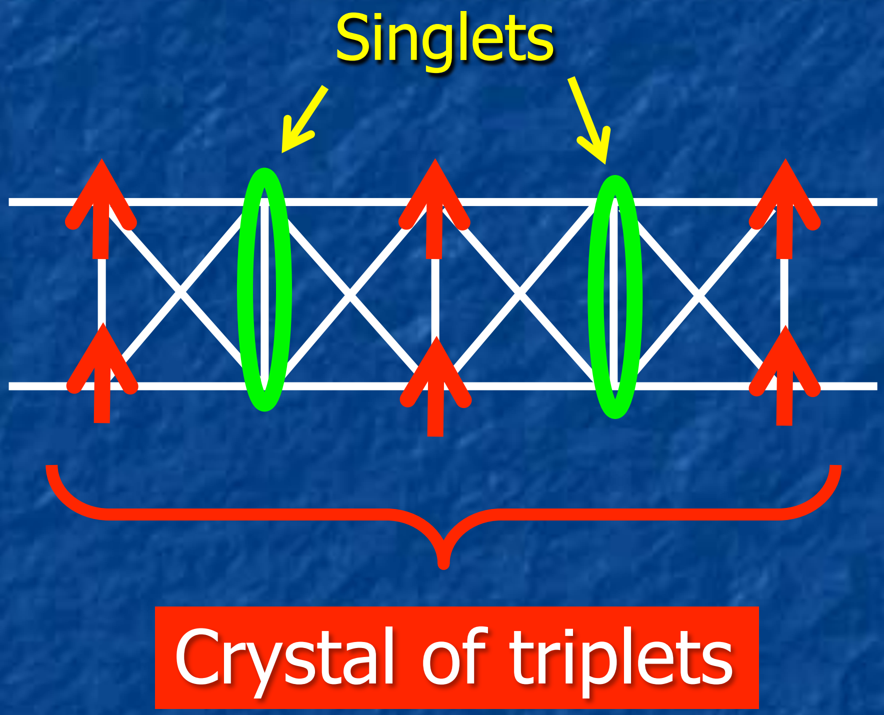


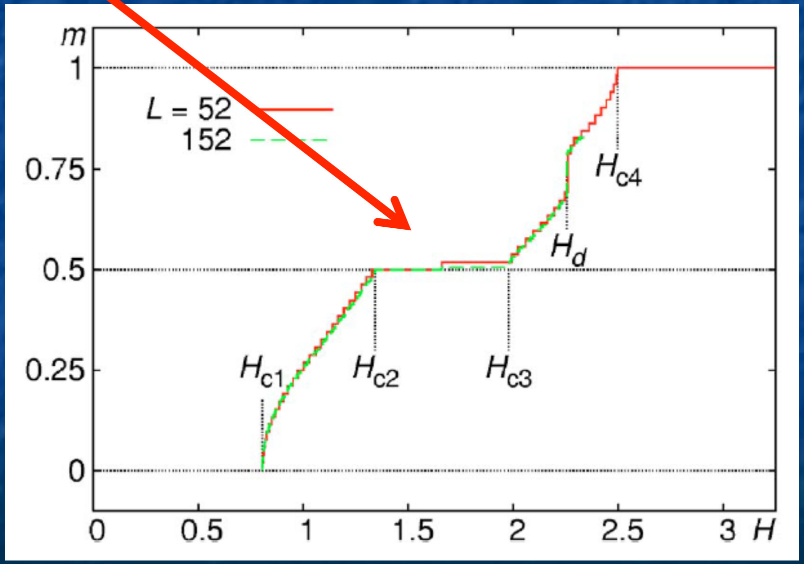
$$H = t\sum_{\langle i,j\rangle}(a_i^\dagger a_j + a_j^\dagger a_i) - \mu\sum_i n_i + V\sum_{\langle i,j\rangle} n_i n_j$$

t=(J-J')/2 $\mu=H-H_c$ V=(J+J')/2

- Mott insulator for V>2t Ovchinnikov 1971
- 1/2 plateau for 1/3<J'/J<3 Totsuka PRB 98; FM EPJB 98

Frustrated ladder

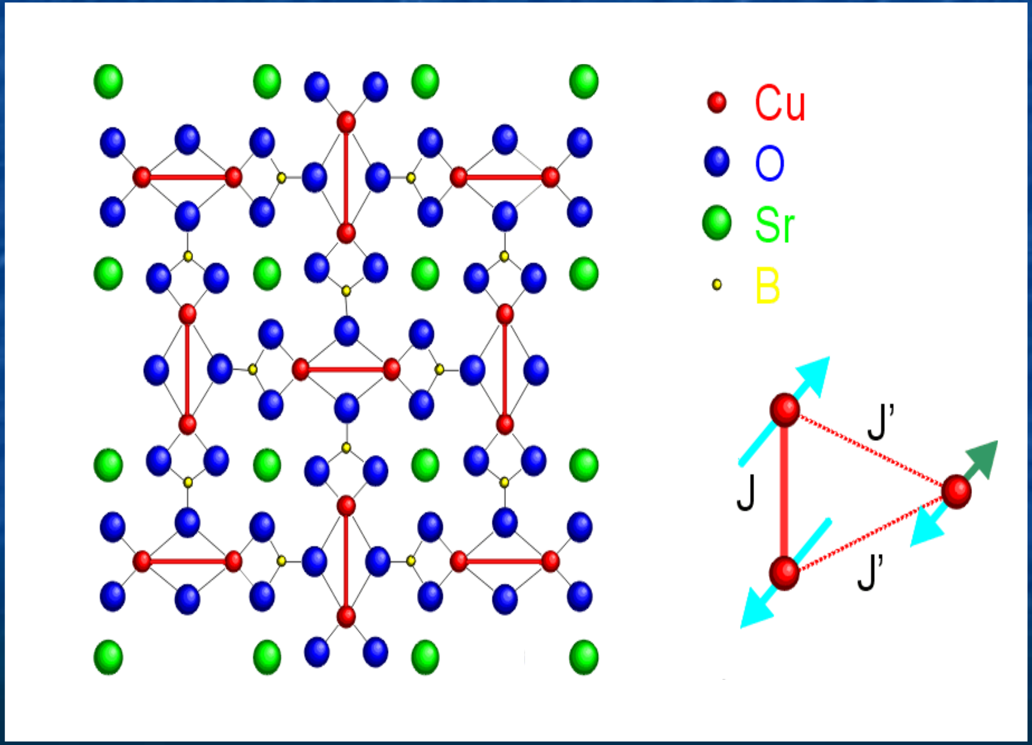




Fouet et al, 2006

$SrCu_2(BO_3)_2$

Smith and Keszler, JSSC 1991

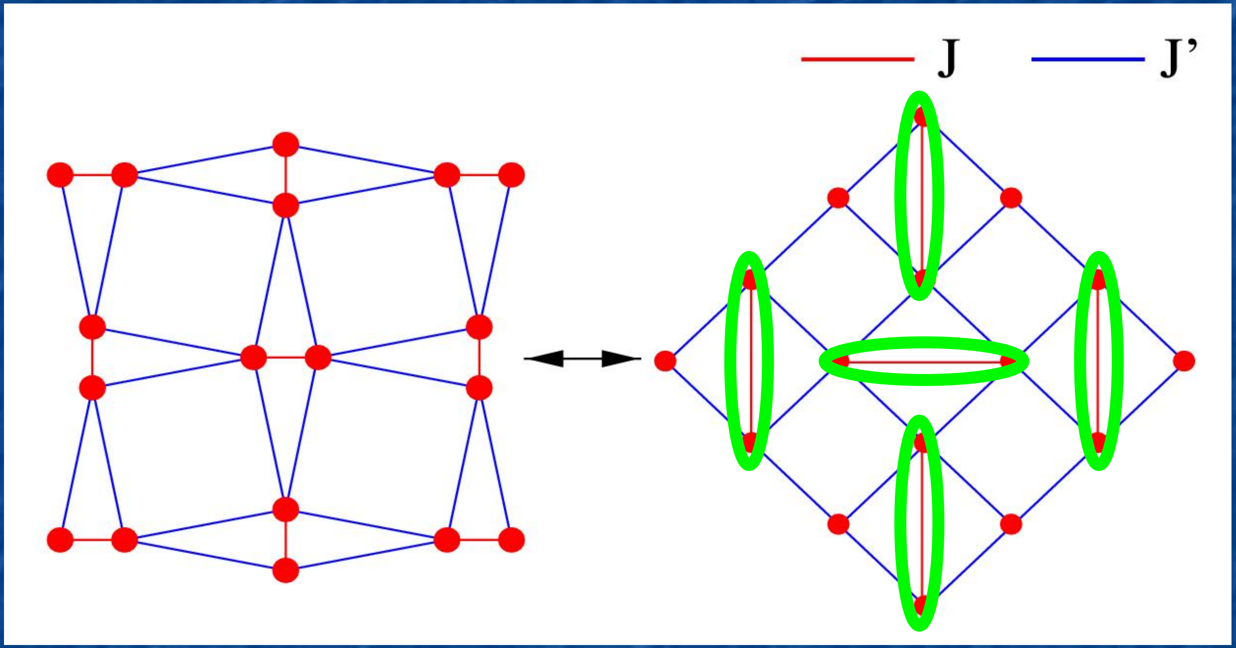


Cu^{2+} -> Spin 1/2

$J \approx 85$ K

$J'/J \approx 0.65$

Shastry-Sutherland model



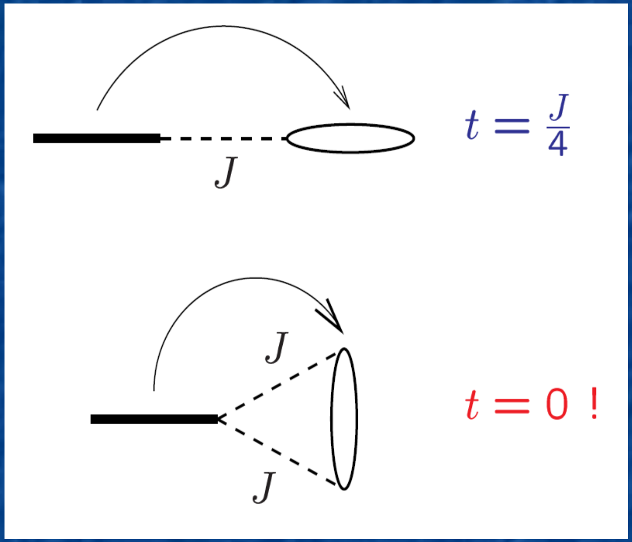
Ground-state = Product of singlets on J-bonds

Shastry and Sutherland, '81

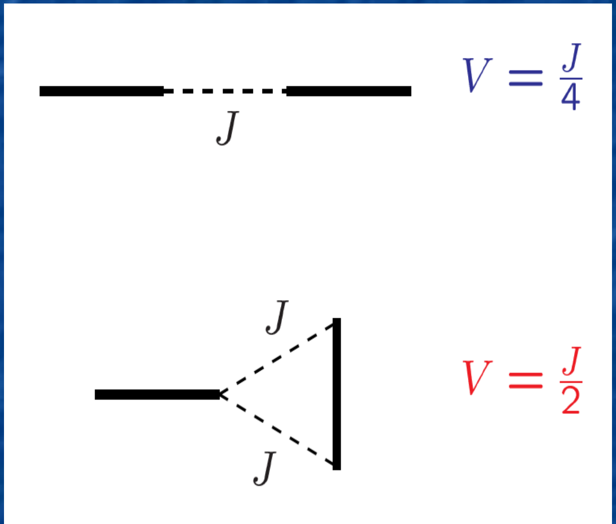
→ Spin gap, and plateau at 0

Effect of frustration

Triplet Hopping



Triplet Repulsion

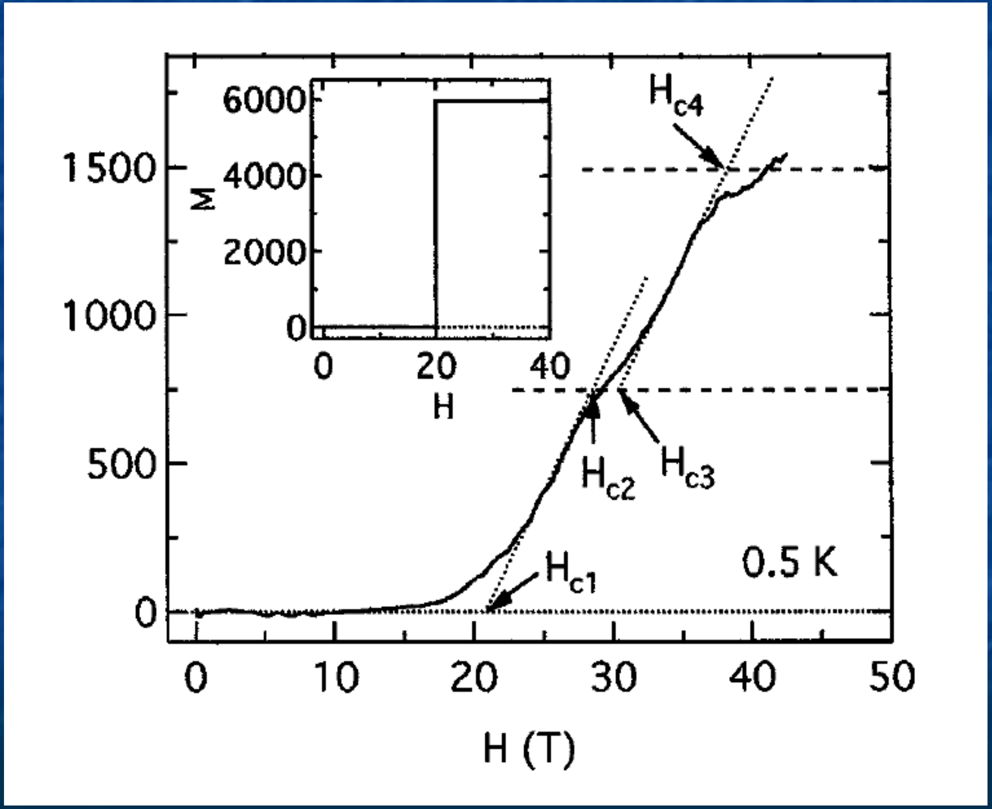


- Kinetic energy << potential energy
- Long-range repulsion
  → Crystals of triplets with high commensurability
  → Magnetization plateaux

VOLUME 82, NUMBER 15 PHYSICAL REVIEW LETTERS 12 APRIL 1999

Exact Dimer Ground State and Quantized Magnetization Plateaus in the Two-Dimensional Spin System $SrCu_2(BO_3)_2$

H. Kageyama,[1,2,*] K. Yoshimura,[1,3,†] R. Stern,[3] N. V. Mushnikov,[2] K. Onizuka,[2] M. Kato,[1] K. Kosuge,[1] C.P. Slichter,[3] T. Goto,[2] and Y. Ueda[2]



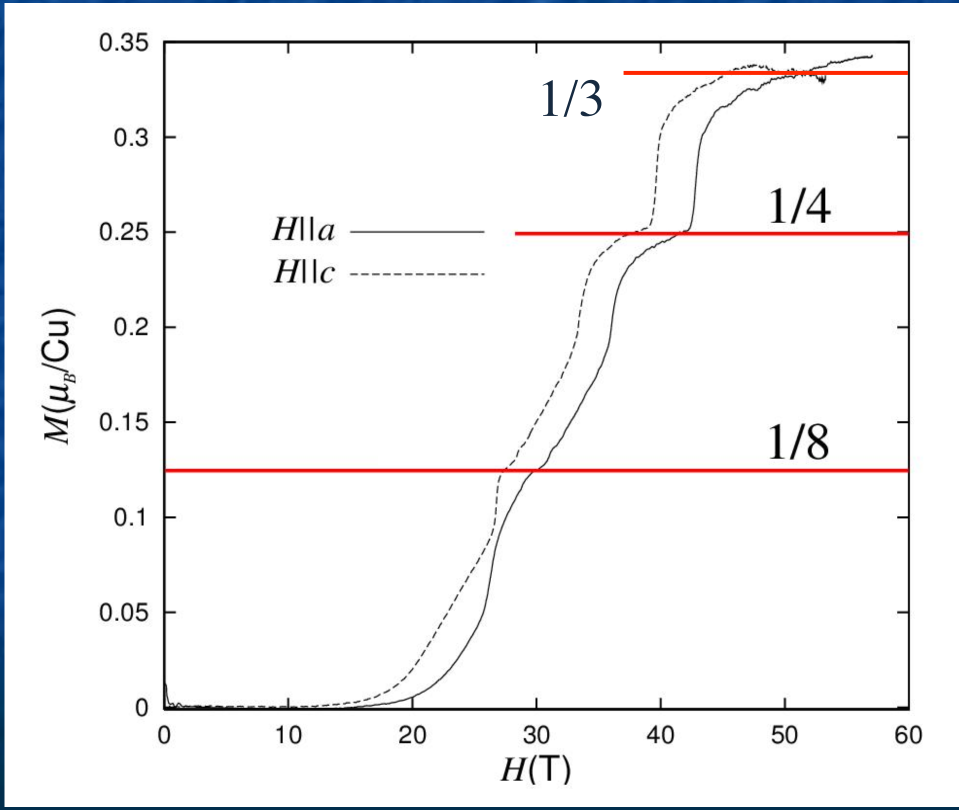
Anomalies

- M=0
- M=1/8
- M=1/4

Journal of the Physical Society of Japan
Vol. 69, No. 4, April, 2000, pp. 1016-1018

1/3 Magnetization Plateau in $SrCu_2(BO_3)_2$ - Stripe Order of Excited Triplets -

Kenzo ONIZUKA, Hiroshi KAGEYAMA*, Yasuo NARUMI[1,2],
Koichi KINDO[2,1], Yutaka UEDA and Tsuneaki GOTO

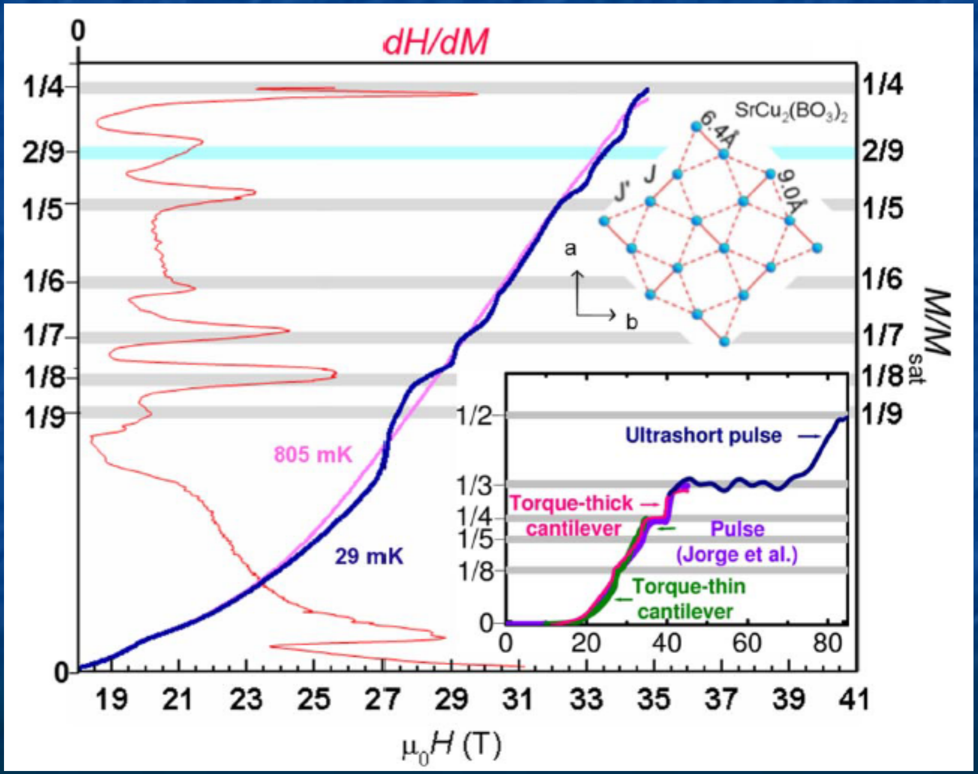


Plateaus
- M=0
- M=1/8
- M=1/4
- M=1/3

Fractalization drives crystalline states in a frustrated spin system

Suchitra E. Sebastian[a,1], N. Harrison[b], P. Sengupta[c], C. D. Batista[c], S. Francoual[b], E. Palm[d], T. Murphy[d], N. Marcano[a], H. A. Dabkowska[e], and B. D. Gaulin[e]

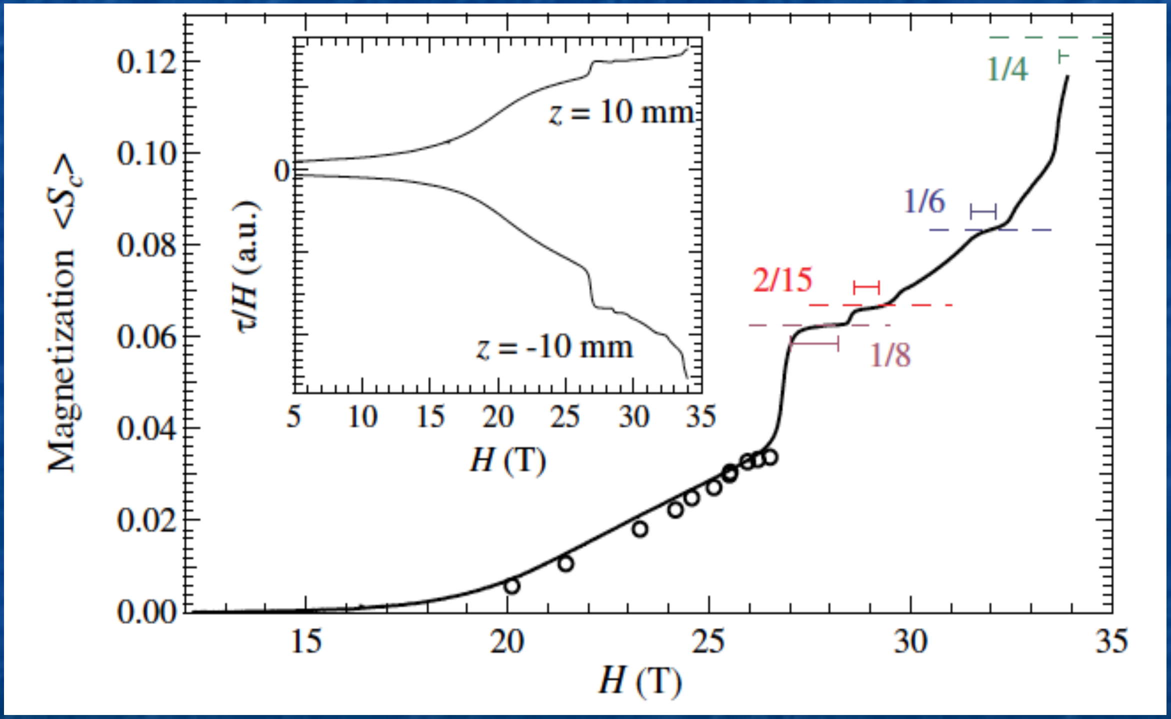
www.pnas.org/cgi/doi/10.1073/pnas.0804320105 PNAS | December 23, 2008 | vol. 105 | no. 51 | 20157–20160



1/9, 1/8, 1/7, 1/6, 1/5, 2/9, 1/4

Incomplete Devil's Staircase in the Magnetization Curve of $SrCu_2(BO_3)_2$

M. Takigawa,[1,*] M. Horvatić,[2] T. Waki,[3] S. Krämer,[2] C. Berthier,[2] F. Lévy-Bertrand,[2,†] I. Sheikin,[2] H. Kageyama,[4] Y. Ueda,[1] and F. Mila[5]

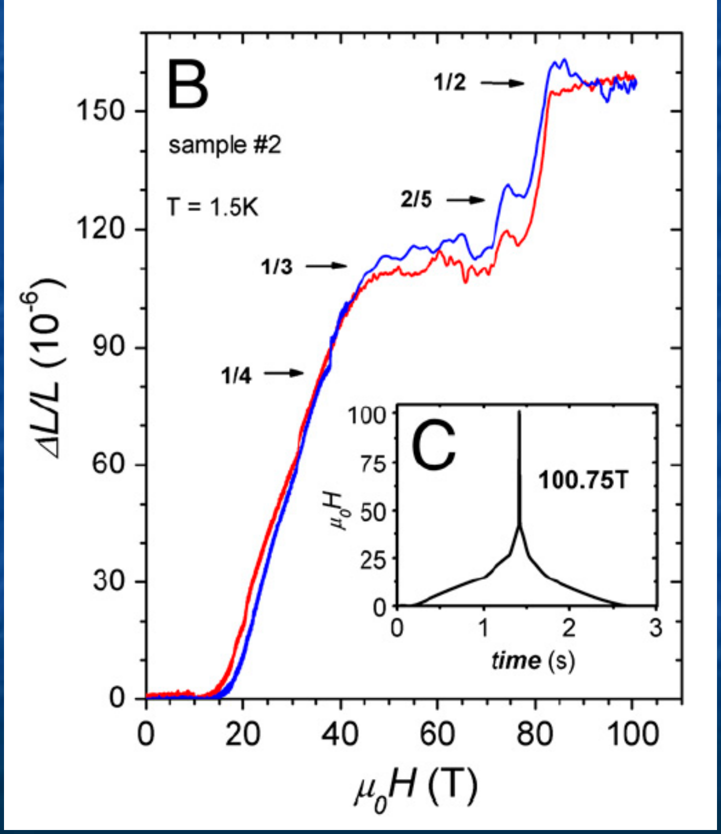


1/8, 2/15, 1/6, 1/4,...

Magnetostriction and magnetic texture to 100.75 Tesla in frustrated $SrCu_2(BO_3)_2$

Marcelo Jaime[a,b,1], Ramzy Daou[c,d], Scott A. Crooker[a,b], Franziska Weickert[b], Atsuko Uchida[a,b], Adrian E. Feiguin[e], Cristian D. Batista[f], Hanna A. Dabkowska[g], and Bruce D. Gaulin[g,h]

12404–12407 | PNAS | July 31, 2012 | vol. 109 | no. 31
www.pnas.org/cgi/doi/10.1073/pnas.1200743109

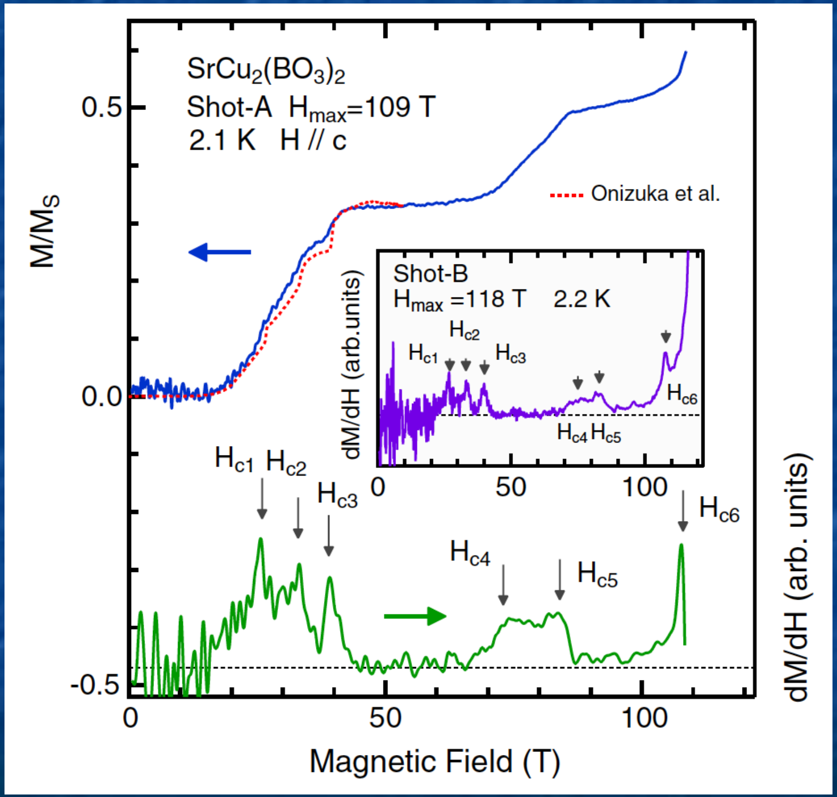


…,1/4, 1/3, 2/5, 1/2

PRL 111, 137204 (2013) PHYSICAL REVIEW LETTERS week ending 27 SEPTEMBER 2013

Magnetization of $SrCu_2(BO_3)_2$ in Ultrahigh Magnetic Fields up to 118 T

Y. H. Matsuda,[1,*] N. Abe,[1] S. Takeyama,[1] H. Kageyama,[2] P. Corboz,[3] A. Honecker,[4,5] S. R. Manmana,[4] G. R. Foltin,[6] K. P. Schmidt,[6] and F. Mila[7]



...1/3, 1/2

Critical summary I

0, 1/9, 1/8, 2/15, 1/7, 1/6, 1/5, 2/9, 1/4, 1/3, 2/5, 1/2

Too many suspects!

Critical summary II

Only pulsed field   Only magnetostriction

0, 1/9, 1/8, 2/15, 1/7, 1/6, 1/5, 2/9, 1/4, 1/3, 2/5, 1/2

0, 1/8, 2/15, 1/6, 1/4, 1/3, 1/2

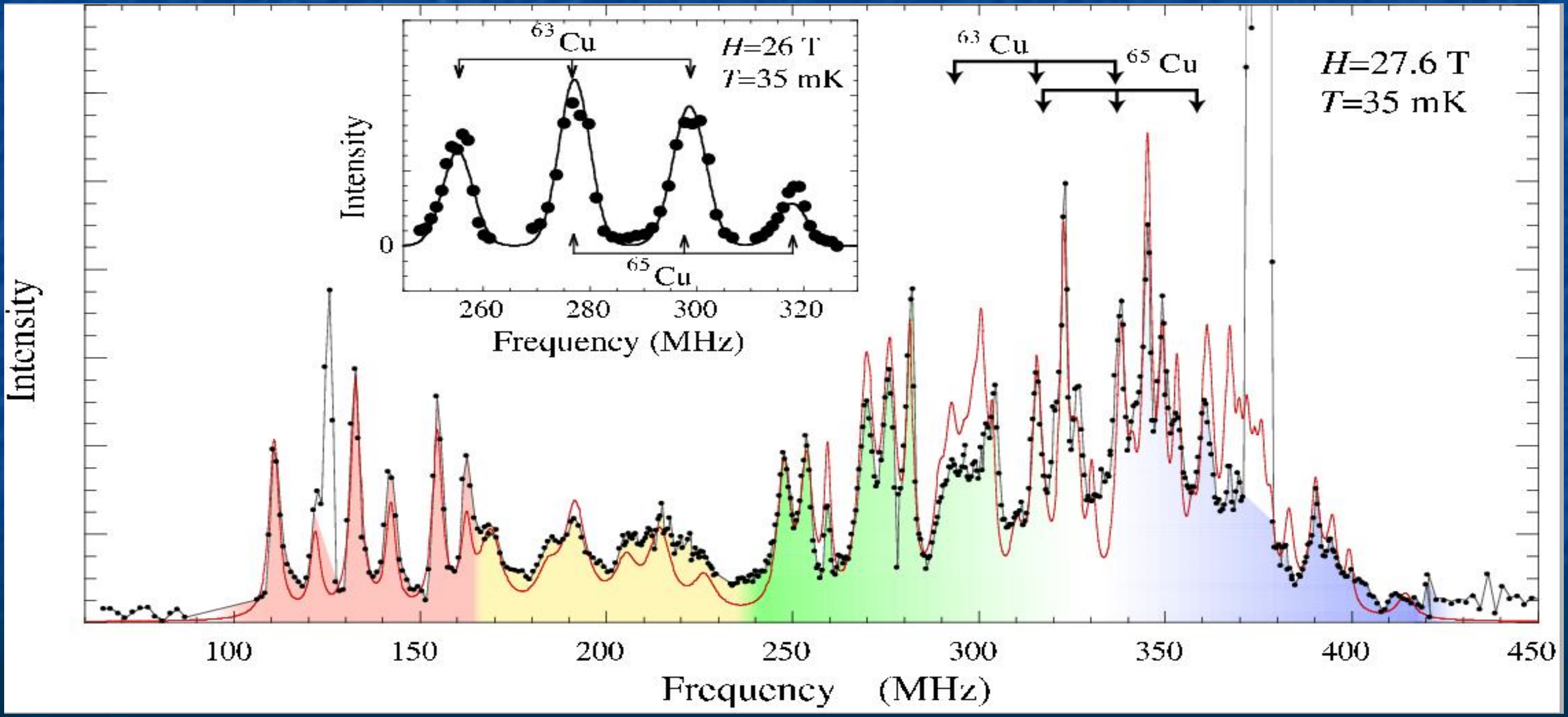
Quantum Hall Effect or broken spatial symmetry?

Magnetic Superstructure in the Two-Dimensional Quantum Antiferromagnet $SrCu_2(BO_3)_2$

K. Kodama,[1] M. Takigawa,[1*] M. Horvatić,[2] C. Berthier,[2,3]
H. Kageyama,[1] Y. Ueda,[1] S. Miyahara,[1,4] F. Becca,[4] F. Mila[4]

www.sciencemag.org SCIENCE VOL 298 11 OCTOBER 2002

Broken symmetry in 1/8 plateau



Incomplete Devil's Staircase in the Magnetization Curve of $SrCu_2(BO_3)_2$

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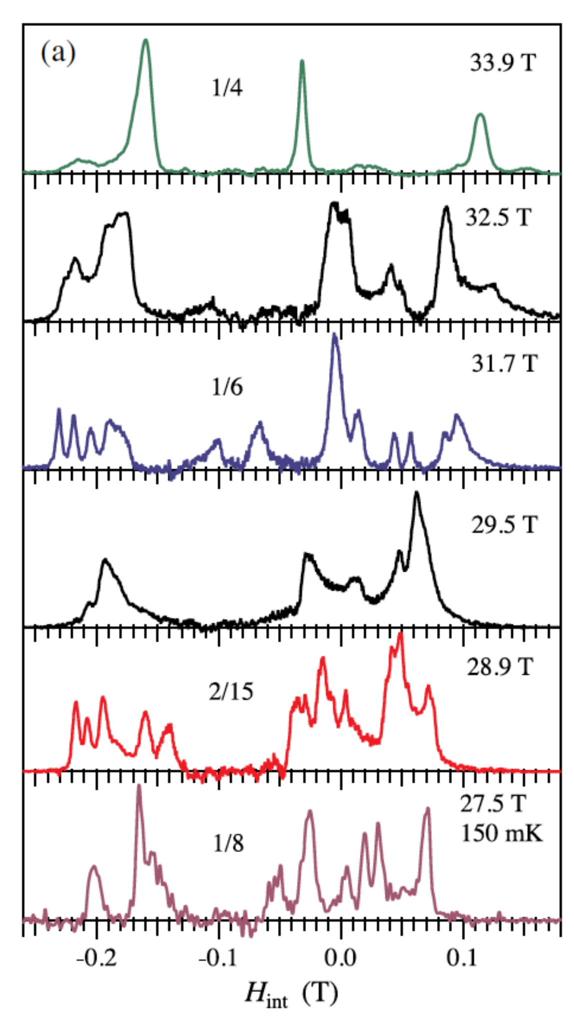
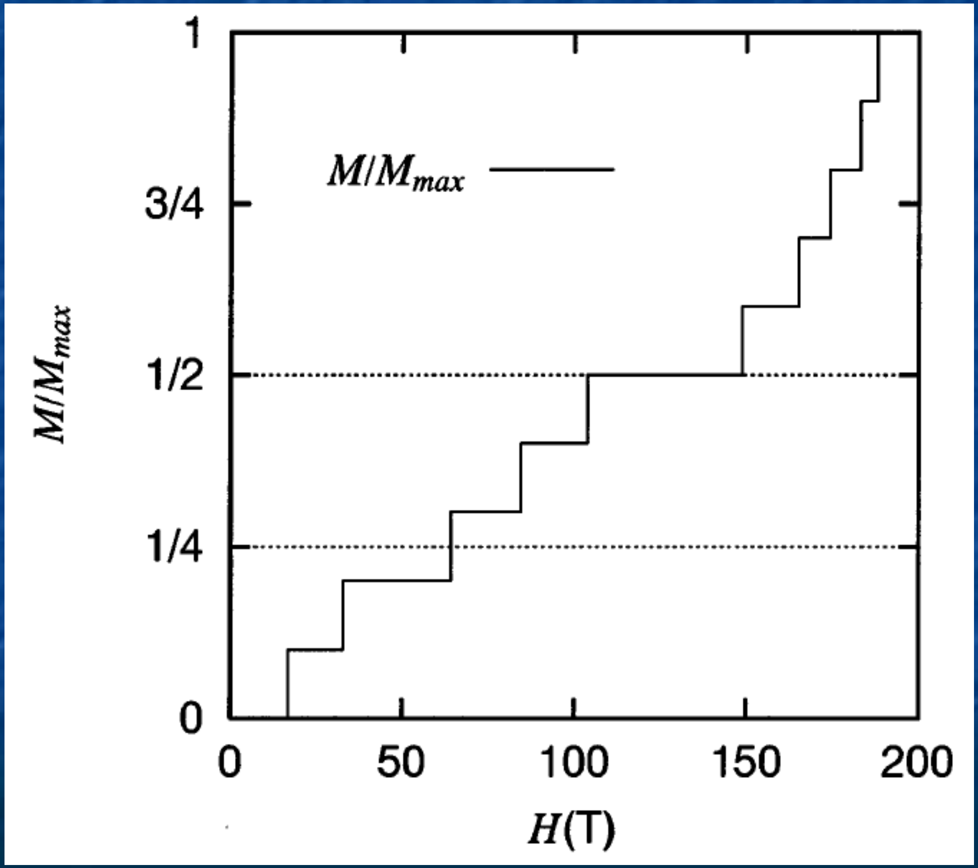


Broken symmetry in
1/8, 2/15, 1/6 and 1/4 plateaus

VOLUME 82, NUMBER 18 PHYSICAL REVIEW LETTERS 3 MAY 1999

Exact Dimer Ground State of the Two Dimensional Heisenberg Spin System $SrCu_2(BO_3)_2$

Shin Miyahara and Kazuo Ueda

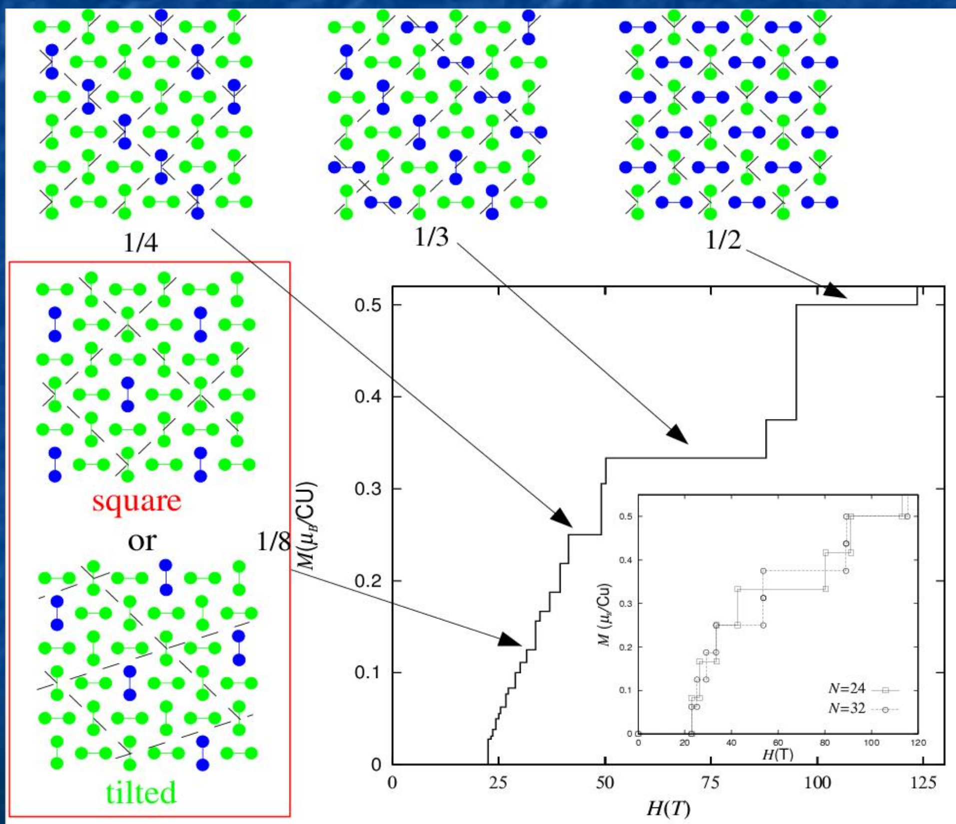


- 20-site cluster
  → finite-size effects

PHYSICAL REVIEW B VOLUME 61, NUMBER 5 1 FEBRUARY 2000-I

Superstructures at magnetization plateaus in $SrCu_2(BO_3)_2$

Shin Miyahara and Kazuo Ueda



- Simple ansatz for long-range triplet-triplet interaction
- Many plateaus

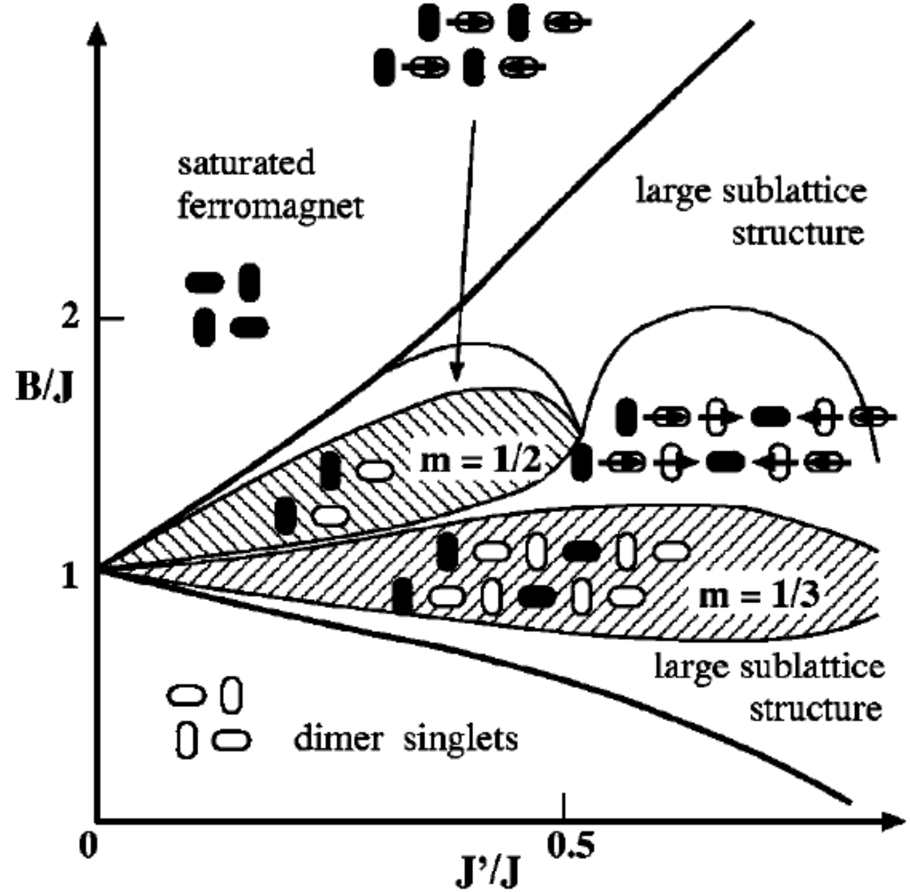
PHYSICAL REVIEW B VOLUME 62, NUMBER 22 1 DECEMBER 2000-II

Magnetization plateaus of the Shastry-Sutherland model for $SrCu_2(BO_3)_2$: Spin-density wave, supersolid, and bound states

Tsutomu Momoi*
Lyman Laboratory of Physics, Harvard University, Cambridge, Massachusetts 02138

Keisuke Totsuka
Department of Physics, Kyushu University, Hakozaki, Higashi-ku, Fukuoka-shi 812-8581, Japan

(Received 1 June 2000)

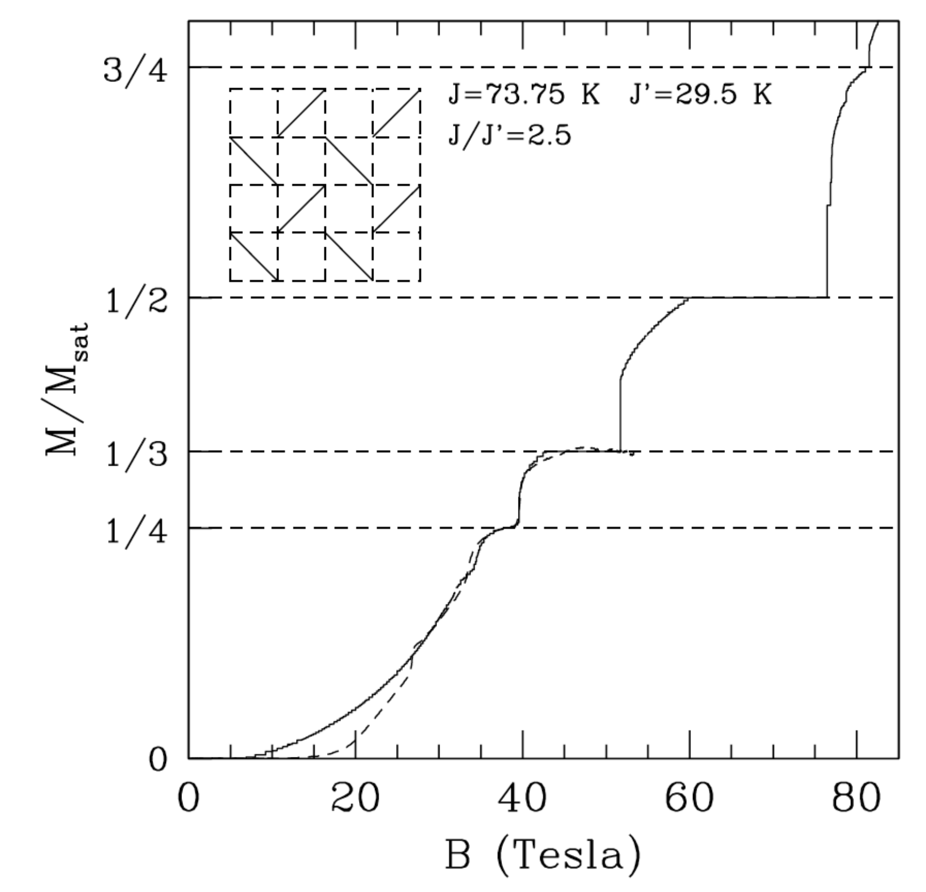


- Effective model to third order in $(J'/J)^3$
- Only short-range triplet-triplet interaction
- Only 1/3 and 1/2 plateaus
- Spin-supersolids

VOLUME 87, NUMBER 9 PHYSICAL REVIEW LETTERS 27 AUGUST 2001

Magnetization Plateaus of $SrCu_2(BO_3)_2$ from a Chern-Simons Theory

G. Misguich,[1] Th. Jolicoeur,[2] and S. M. Girvin[3,4]



- No breaking of spatial symmetry
- Incompatible with NMR

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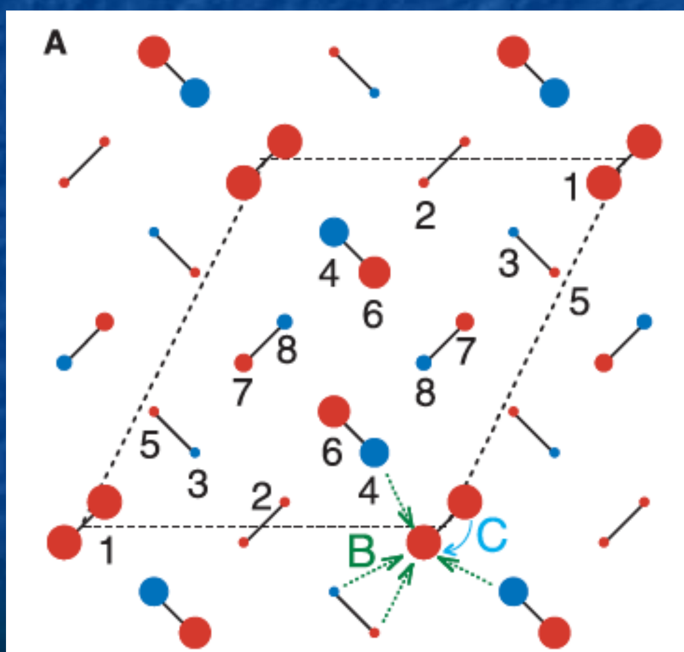
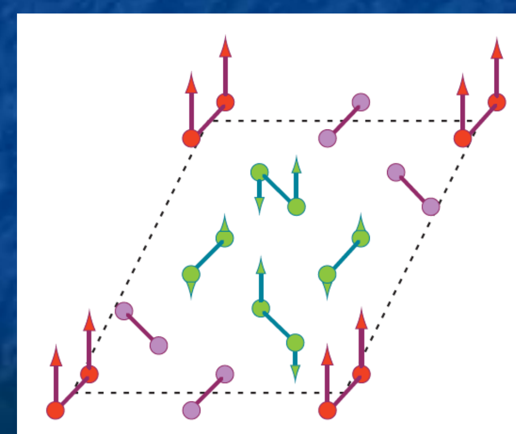
H. Kageyama,[1] Y. Ueda,[1] S. Miyahara,[1,4] F. Becca,[4] F. Mila[4]

www.sciencemag.org SCIENCE VOL 298 11 OCTOBER 2002



1/8 plateau
= crystal of (dressed) triplets

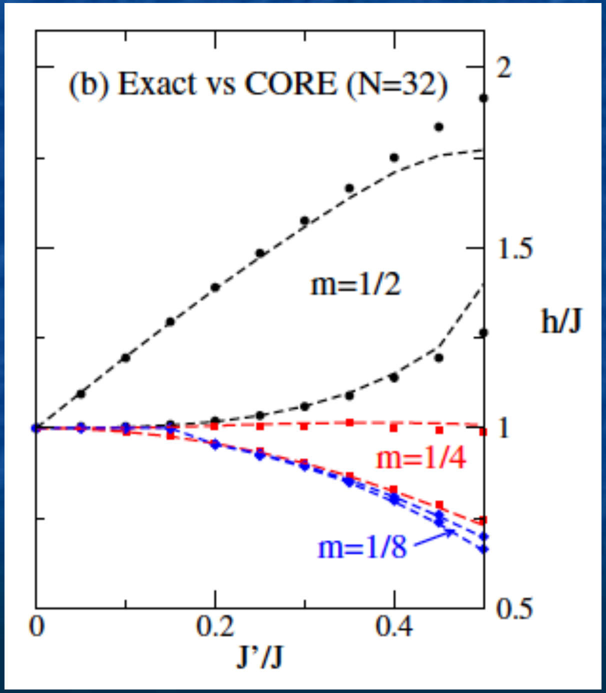


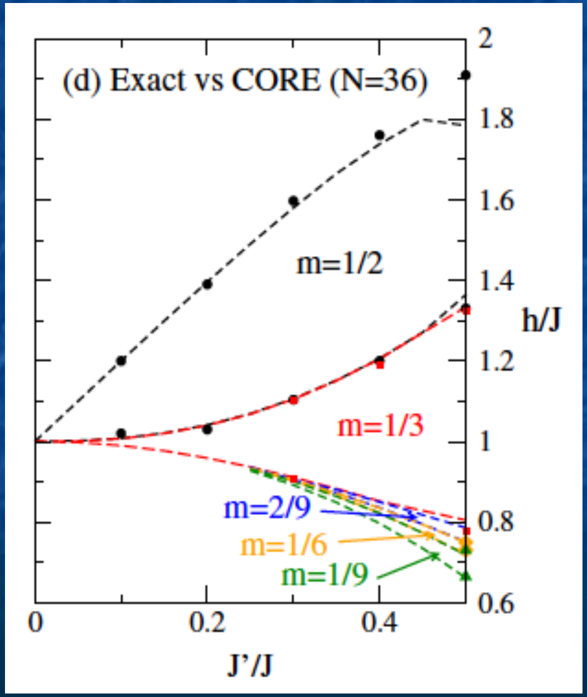
PRL 101, 227201 (2008) PHYSICAL REVIEW LETTERS week ending 28 NOVEMBER 2008

Effective Theory of Magnetization Plateaux in the Shastry-Sutherland Lattice

A. Abendschein[1,2] and S. Capponi[1,2,*]

Effective model with CORE (Contractor Renormalization)
Exact diagonalization on small clusters





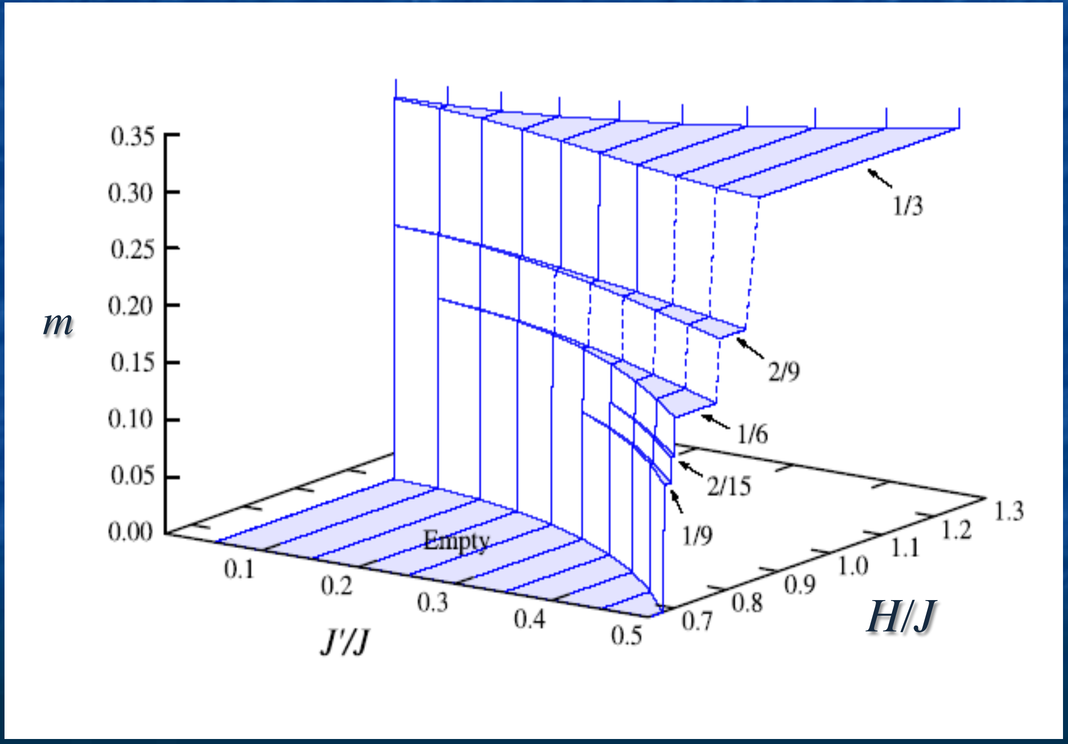
- Large finite-size effects
- No 2/15 plateau
- No information on plateau structures

PRL 101, 250402 (2008) PHYSICAL REVIEW LETTERS week ending 19 DECEMBER 2008

Theory of Magnetization Plateaux in the Shastry-Sutherland Model

J. Dorier,[1] K. P. Schmidt,[2,*] and F. Mila[1]

Long-range triplet-triplet interaction with high-order perturbation theory in J'/J



Triplet crystals:
not the right sequence
(at least up to J'/J=0.5)

PRL 111, 137204 (2013) PHYSICAL REVIEW LETTERS week ending 27 SEPTEMBER 2013

Magnetization of $SrCu_2(BO_3)_2$ in Ultrahigh Magnetic Fields up to 118 T

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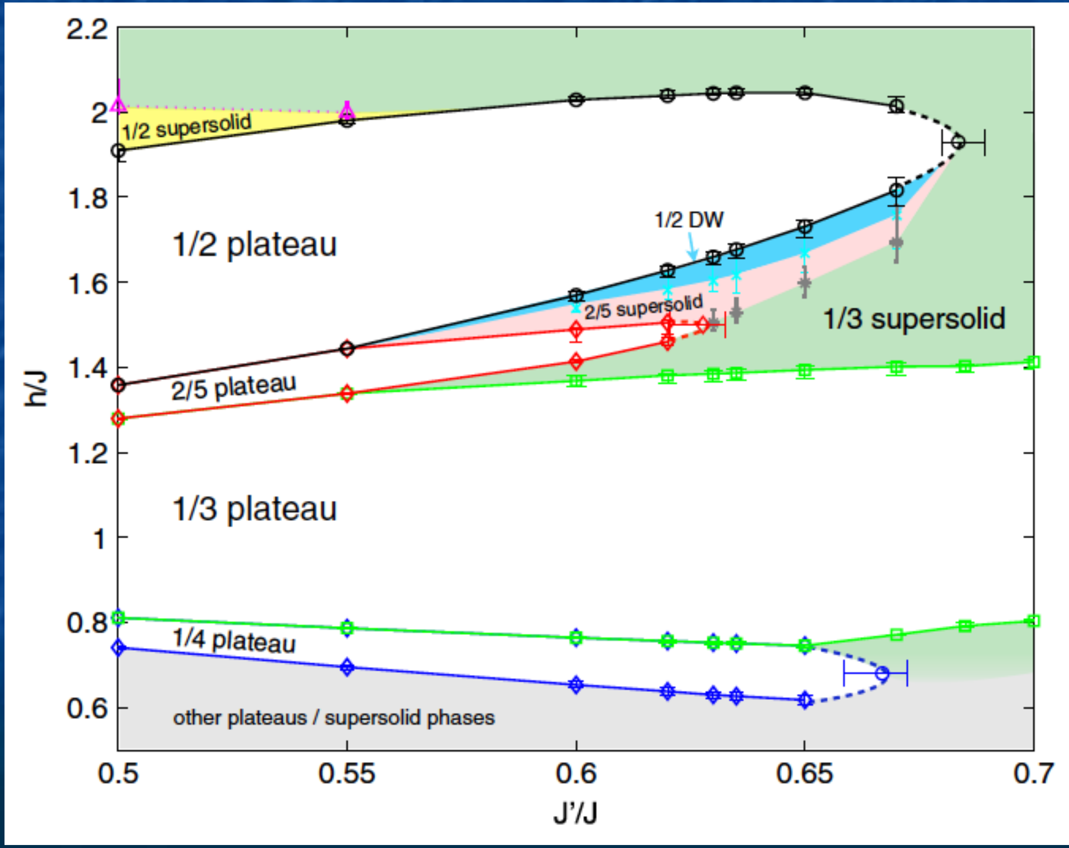


iPEPS

A variational approach derived from quantum information

Verstraete & Cirac, 2004

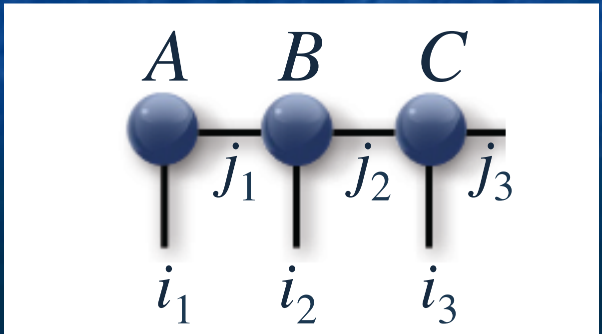
Tensor network ansatz

$$|\psi\rangle = \sum_{i_1 \dots i_N} c_{i_1 \dots i_N} |i_1\rangle \otimes \cdots \otimes |i_N\rangle$$

$c_{i_1 \dots i_N} \simeq$ trace over a product of tensors

Example: Matrix product state in 1D

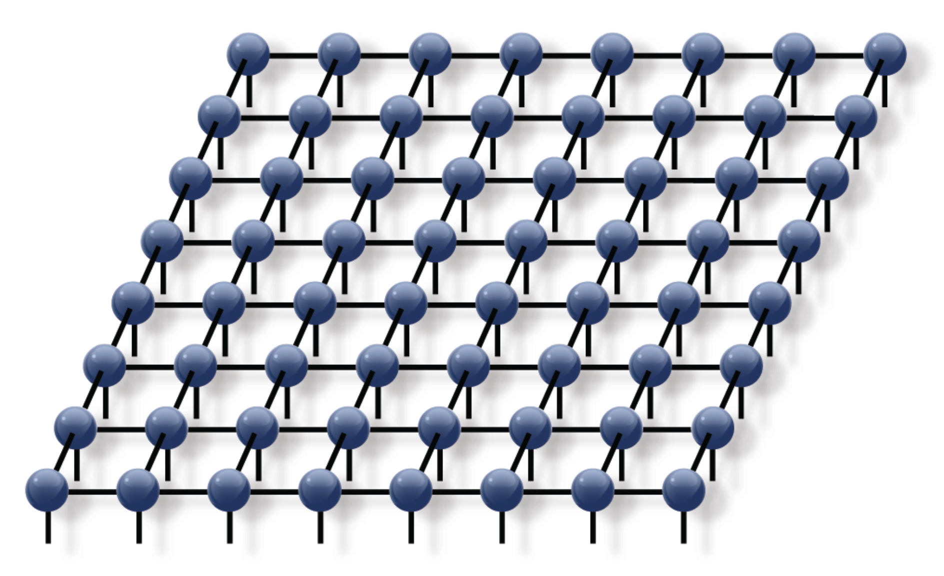
$$c_{i_1 i_2 i_3 \dots} \simeq \sum_{j_1 j_2 \dots} A_{i_1}^{j_1} B_{i_2}^{j_1 j_2} C_{i_3}^{j_2 j_3} \cdots$$



Generalization to 2D

PEPS = product of entangled pair states

Verstraete and Cirac, 2004



$$A_i^{j_1 j_2 j_3 j_4} = \text{rank-5 tensor}$$

$$j_1, j_2, j_3, j_4 = 1, \ldots, D$$

Variational approach

- PEPS: minimize the energy w.r.t. tensor elements
- Other schemes: renormalization (MERA,…)
- Advantage: dim=pol(D,N), not exp(N)
- Why can it work?

→ reproduces the 'area law' for the entanglement entropy in the GS of a local Hamiltonian

$$S = -\mathrm{tr}\left(\rho_A \log \rho_A\right) \sim \partial A$$

- How large should D be? It depends…

Results on frustrated magnets

- Kagome: difficult
- J_1-J_2 on square lattice: intermediate phase around $J_2/J_1=1/2$, but no consensus on its nature

  → Plaquette?

  Yu and Kao, PRB 2012

  → Topological spin liquid?

  Mezzacapo, PRB 2012; Wang, Gu, Verstraete, Wen 2012
- Checkerboard: intermediate plaquette phase, in agreement with previous results

  Chan, Han, Duan, PRB 2011

Shastry-Sutherland

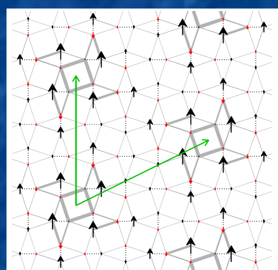
- Isacsson and Syljuasen, PRE 2006
  → D=2, no intermediate phase in zero field
- Lou, Suzuki, Harada, Kawashima, arxiv 2012
  → MERA
  → intermediate plaquette phase
  → plateaux and supersolid phases in a field
- P. Corboz and FM, PRB 2013, PRL 2014
  → plateau sequence of $SrCu_2(BO_3)_2$

PRL 112, 147203 (2014) PHYSICAL REVIEW LETTERS week ending 11 APRIL 2014

Crystals of Bound States in the Magnetization Plateaus of the Shastry-Sutherland Model

Philippe Corboz[1] and Frédéric Mila[2]

1/8



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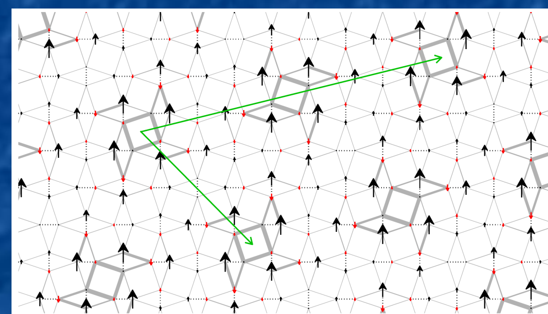
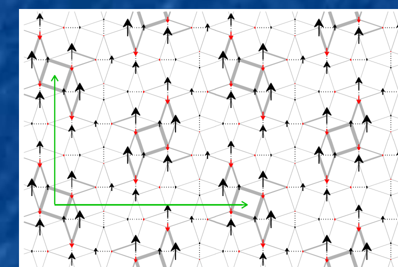
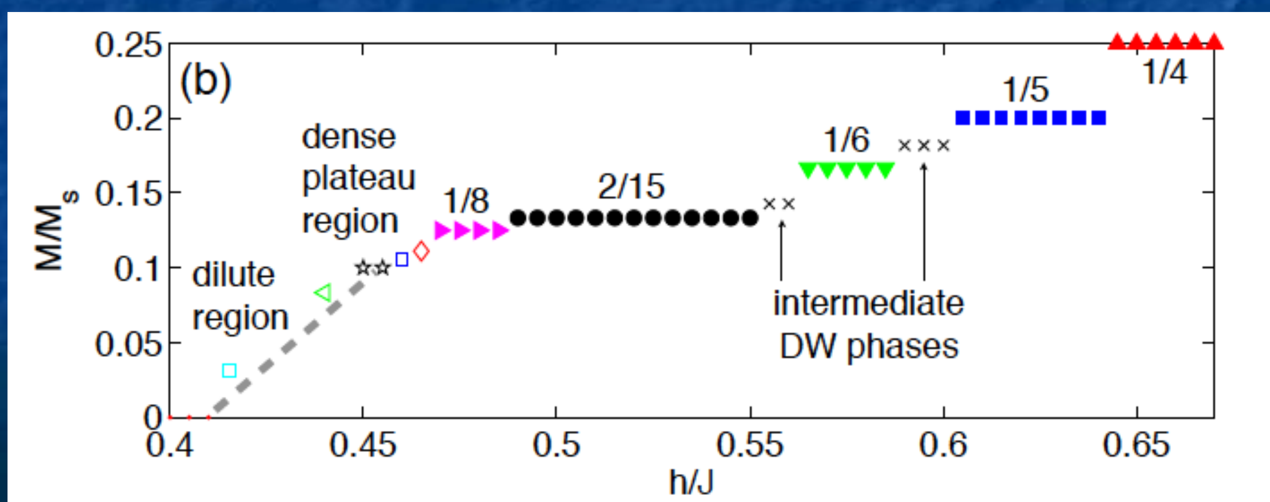
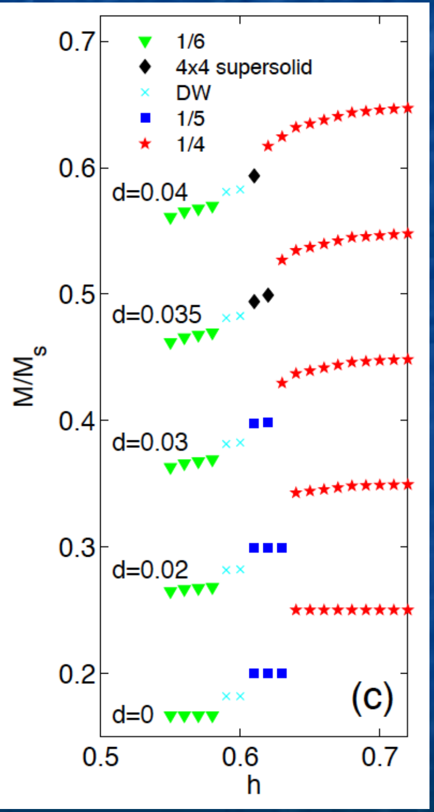


1/6





Why no 1/5 plateau?

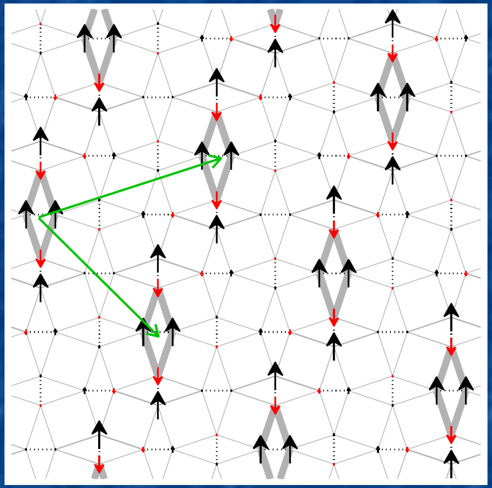


Killed by Dzyaloshinskii-Moriya Interaction

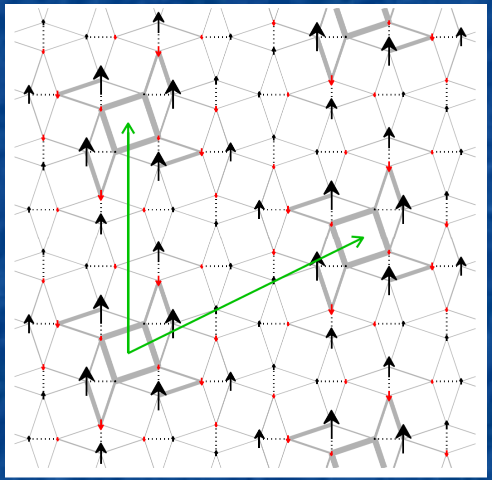
P. Corboz, FM, PRL 2014

Triplets versus bound-states

Triplet crystal



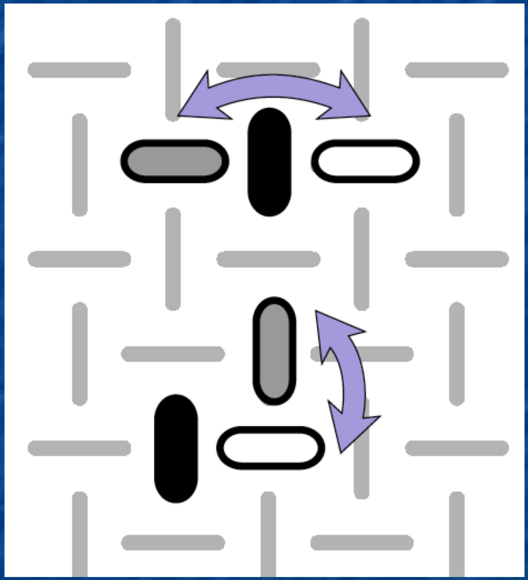
Bound state crystal



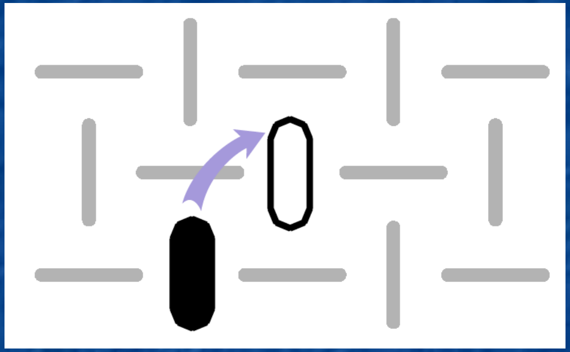
- Why are bound state crystals favoured?
- Why did it take 15 years to identify them?

Why are bound states favoured?

Correlated hopping



Single particle hopping

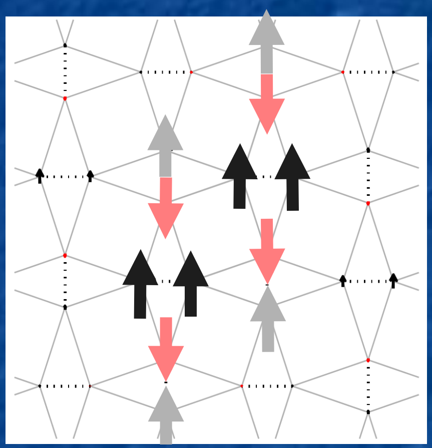


$\alpha\ (J'/J)^6$

$\alpha\ (J'/J)^2 \rightarrow$ gain in kinetic energy

Boundstates as pinwheels

Order $(J'/J)^3$



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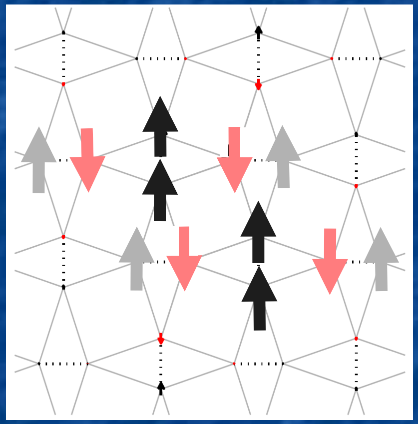

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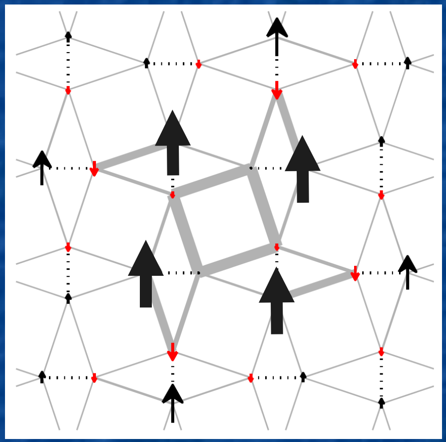

Higher order

PHYSICAL REVIEW B VOLUME 62, NUMBER 22 1 DECEMBER 2000-II

Magnetization plateaus of the Shastry-Sutherland model for $SrCu_2(BO_3)_2$: Spin-density wave, supersolid, and bound states

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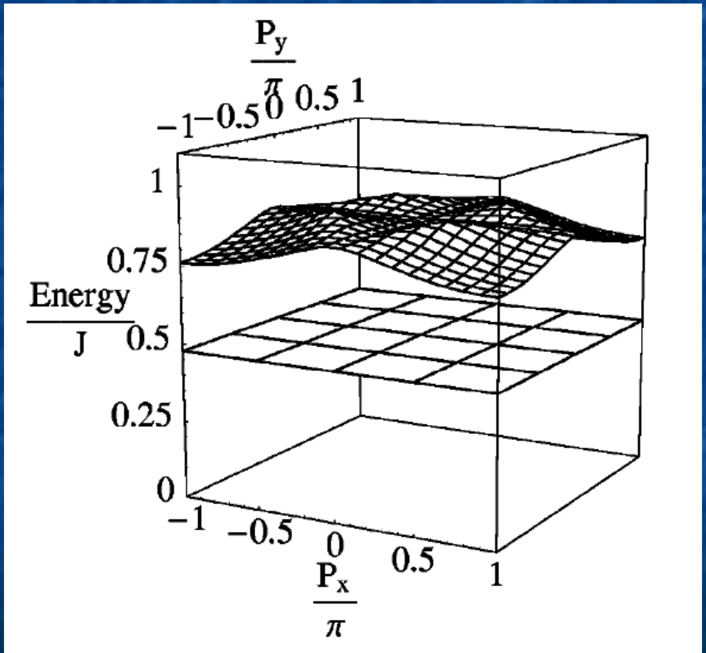
(Received 1 June 2000)



Bound state dispersion: minimum at the zone corner

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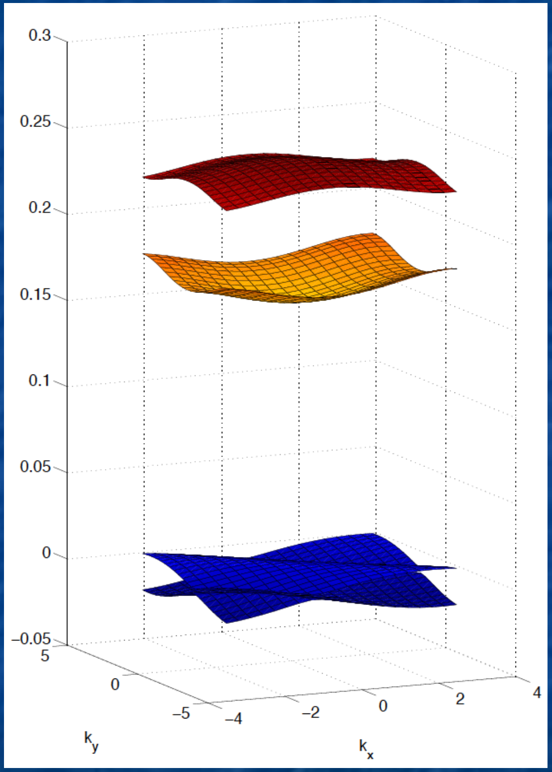
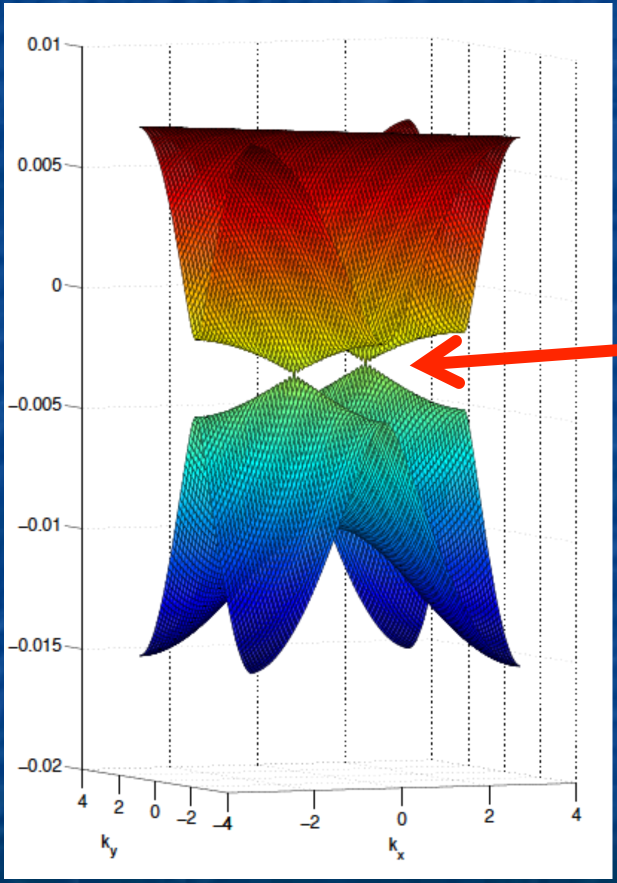
If bound states tend to crystallize, bound states loose kinetic energy and then they will be unbound by repulsive interaction between triplets.

Is that really true?

Localized bound state



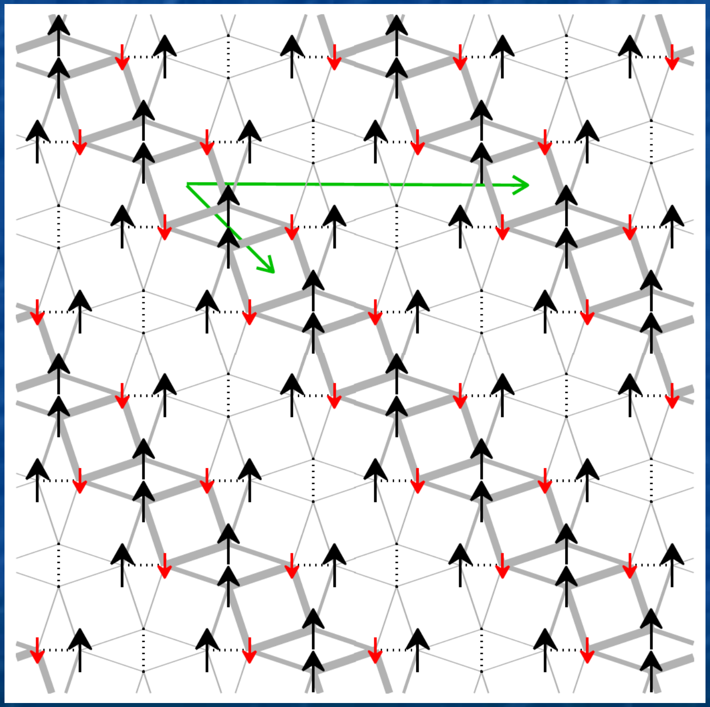


Negative reference energy!

P. Corboz, FM, PRL 2014

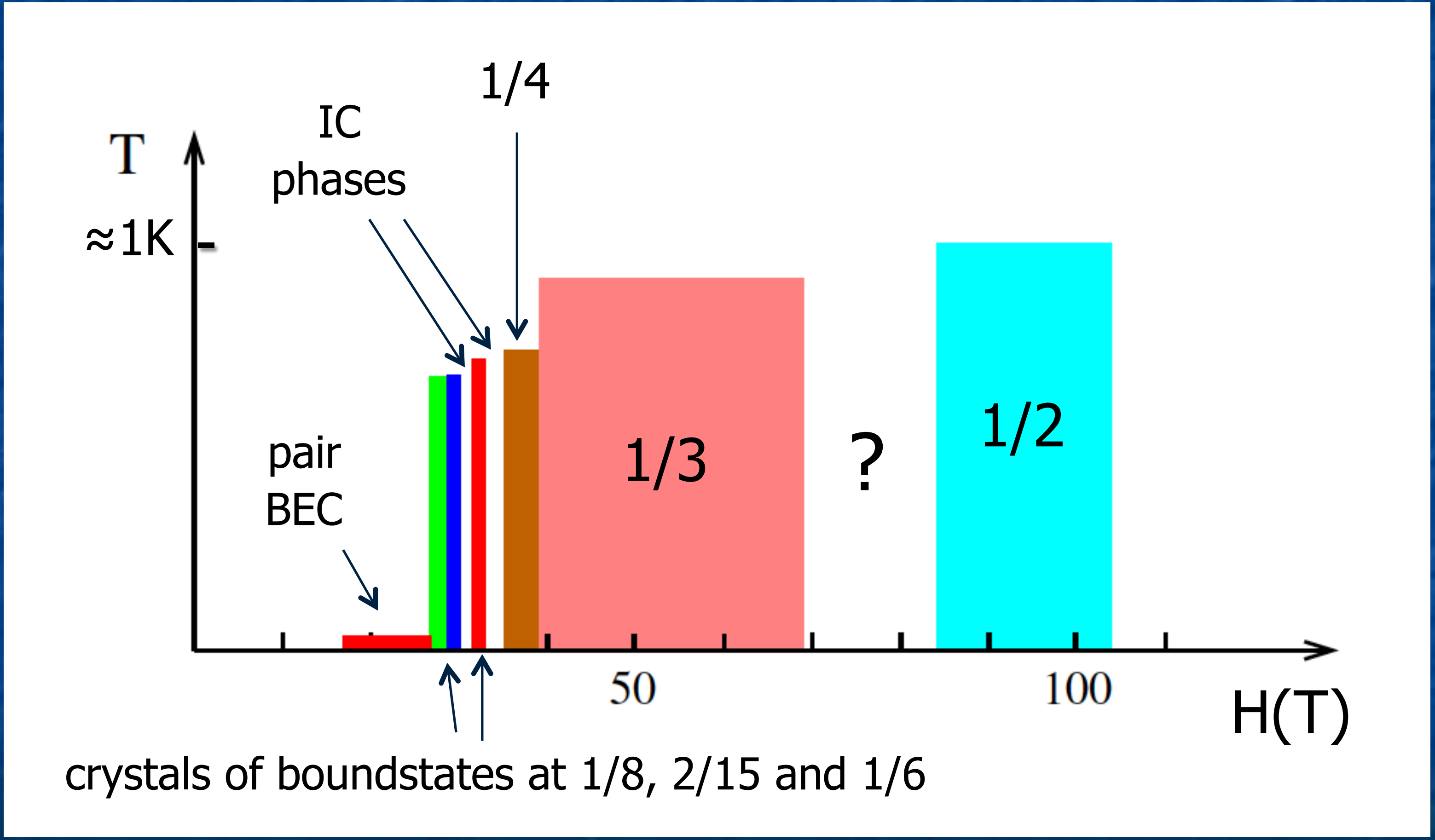
Dense triplets as bound states

1/4 plateau



1/4 , 1/3 and 1/2 plateaux: can be seen both as crystals of triplets and as crystals of bound states.

Tentative phase diagram



Pair BEC: Bendjama, Kumar, FM, PRL 2005; Tc: Takigawa et al, PRL 2008

Conclusions on $SrCu_2(BO_3)_2$

- Natural plateau sequence
  → 1/8, 2/15, 1/6: crystals of bound states
  → new predictions for their symmetry
- In progress
  → revisit effective triplet model
  → compare predictions with NMR
- Still to be done
  → X-ray or neutron in high field (>27 T)?

Conclusions

- Magnetization of frustrated magnets
  → very rich
  → reveals several fundamental aspects of
  strongly correlated systems