

GDR 3183 MICO:
Matériaux et Interactions
en COmpétition

Autrans, 1-4 décembre 2008



New Quantum Magnetic Phases in $\text{SrCu}_2(\text{BO}_3)_2$:
A Route to Supersolid Phases ?

M. Horvatić

high-field **NMR** in Solids

<http://ghmfl.grenoble.cnrs.fr/spip.php?rubrique33>



Grenoble High Magnetic
Field Laboratory

<http://ghmfl.grenoble.cnrs.fr>



CENTRE NATIONAL
DE LA RECHERCHE
SCIENTIFIQUE



NMR investigation of plateaus in the $\text{SrCu}_2(\text{BO}_3)_2$ system

[K. Kodama *et al.*, *Science* **298**, 395 (2002) and *J. Phys.: Condens. Matter* **17**, L61 (2005); S. Miyahara, F. Becca, F. Mila, *Phys. Rev. B* **68**, 024401 (2003); F. Lévy *et al.*, *EPL* **81**, 67004 (2008) and unpublished; M. Takigawa *et al.*, *PRL* **101**, 037202 (2008) and unpublished]

Samples: H. Kageyama* Y. Ueda * *Dpt. of Chemistry, Kyoto Univ.*

NMR: K. Kodama† S. Matsubara‡ T. Waki M. Takigawa
M. Horvatić C. Berthier S. Krämer

Torque: F. Lévy I. Sheikin ** *Dpt. of Physics, Aoyama Gakuin Univ.*

Theory: S. Miyahara** F. Becca†† J. Dorier ... F. Mila

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



*Grenoble High Magnetic Field Laboratory, CNRS,
B.P. 166X, F-38042 Grenoble Cedex 9, France*



Institut de Physique Théorique, Ecole Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland

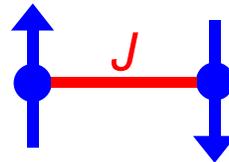
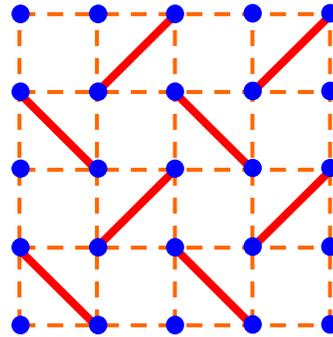
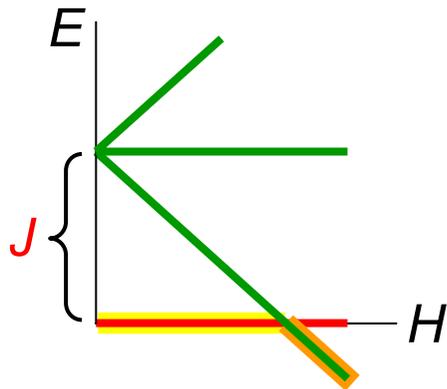


Low-D, quantum, AF spin systems - Definition:

Heisenberg Hamiltonian:
$$\mathcal{H}_H = \sum_{i,\delta} \mathbf{J}_{i,\delta} \vec{S}_{i+\delta} \cdot \vec{S}_i$$

(+ $\vec{D} \cdot \vec{S}_{i+\delta} \times \vec{S}_i + J_{3D} \vec{S}_i \cdot \vec{S}_j$)

$S = 1/2$
spin-dimer
systems:



$\text{SrCu}_2(\text{BO}_3)_2$:

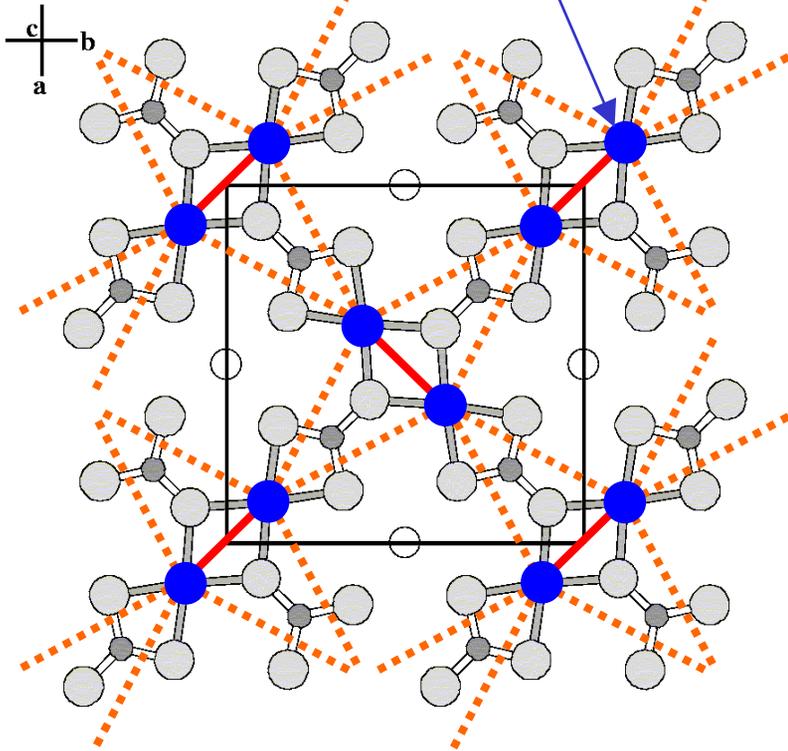
Shastry-
Sutherland

(plateaus,
supersolid?)

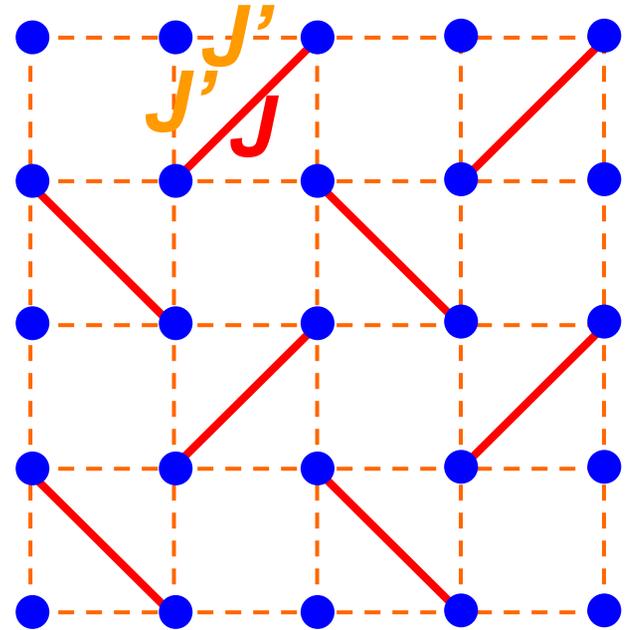
System: $S = 1/2$, 2D, AF, frustrated, highly symmetric !



Cu: $S = 1/2$ spins



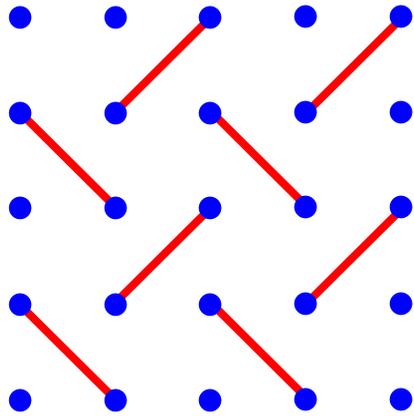
Shastry-Sutherland
Hamiltonian



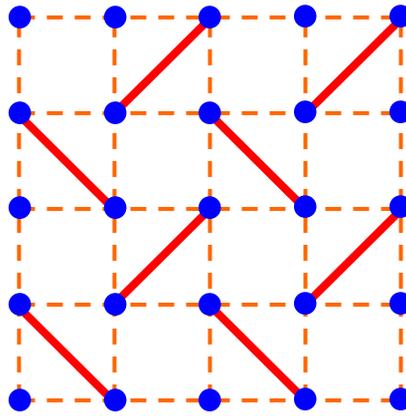
$$\mathcal{H} = \sum_{\text{"diagonal" bonds}} J \vec{S}_i \cdot \vec{S}_j + \sum_{\text{"square" bonds}} J' \vec{S}_i \cdot \vec{S}_j - B \sum_i S_i^z$$

[B.S. Shastry, B. Sutherland, Physica (Amsterdam) B108, 1069 (1981)]

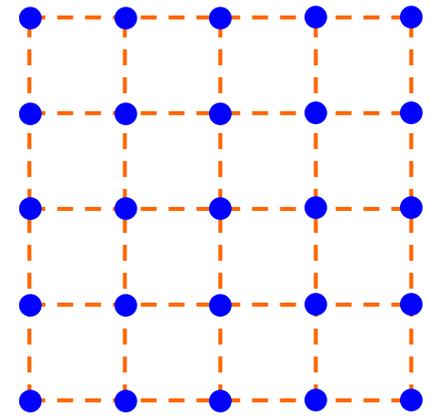
Separated Dimers (J)



Shastry-Southerland

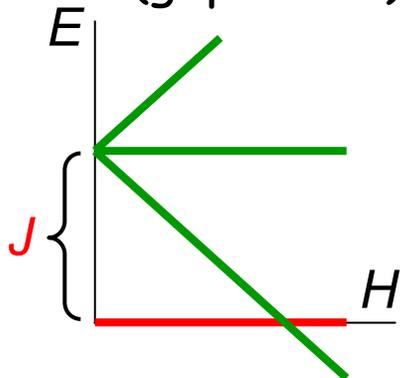


2D square lattice (J')



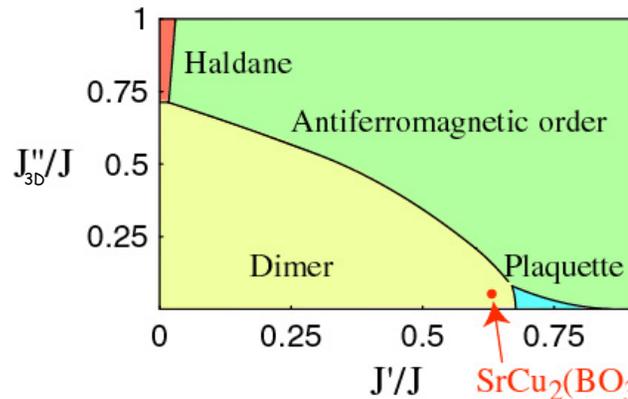
$$|\Psi\rangle = \prod_i |\Psi_{\text{dimer}}\rangle_i$$

(product of) localized
singlets +
 triplet excitations
 (gap $\Delta = J$)



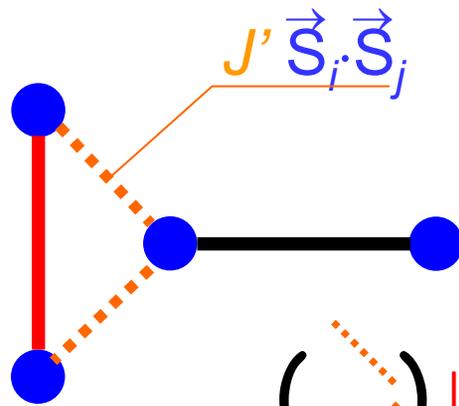
product of singlets is
 always an eigenstate!
 and at $J' < 0.7 J$
 = ground state

AF ground state



A. Koga, JPSJ **69**,
 3509 (2000)

$$|s\rangle = \frac{|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle}{\sqrt{2}}$$



$$|\Psi_{GS}\rangle = \prod_i |s\rangle_i$$

$$\left(\begin{array}{c} \text{---} \\ \text{---} \\ \text{---} \end{array} \right) |s\rangle|?\rangle = 0$$

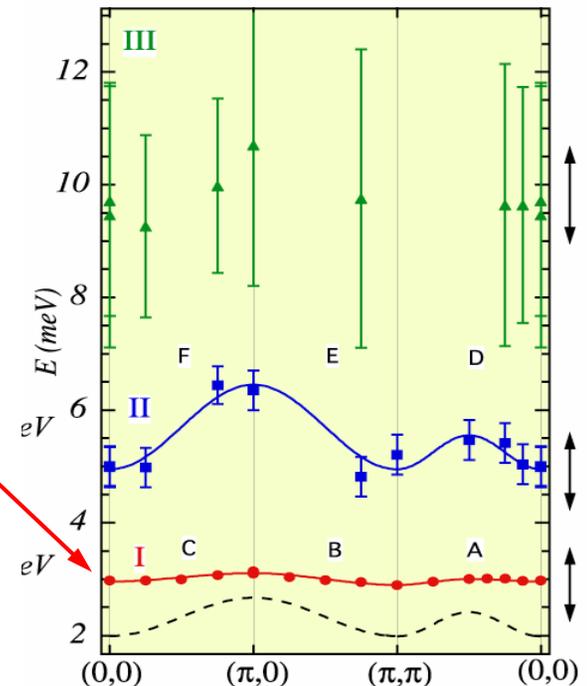
$$\left(\begin{array}{c} \text{---} \\ \text{---} \\ \text{---} \end{array} \right) |t_1\rangle|s\rangle = \frac{J'}{2} (|t_1\rangle|t_0\rangle - |t_0\rangle|t_1\rangle)$$

correction to **localized triplets** very small - starting only in 6-th order of perturbation $(J'/J)^6$

(negligible dispersion)

very small kinetic energy ($E_k \approx \hbar^2 k^2 / 2m$)

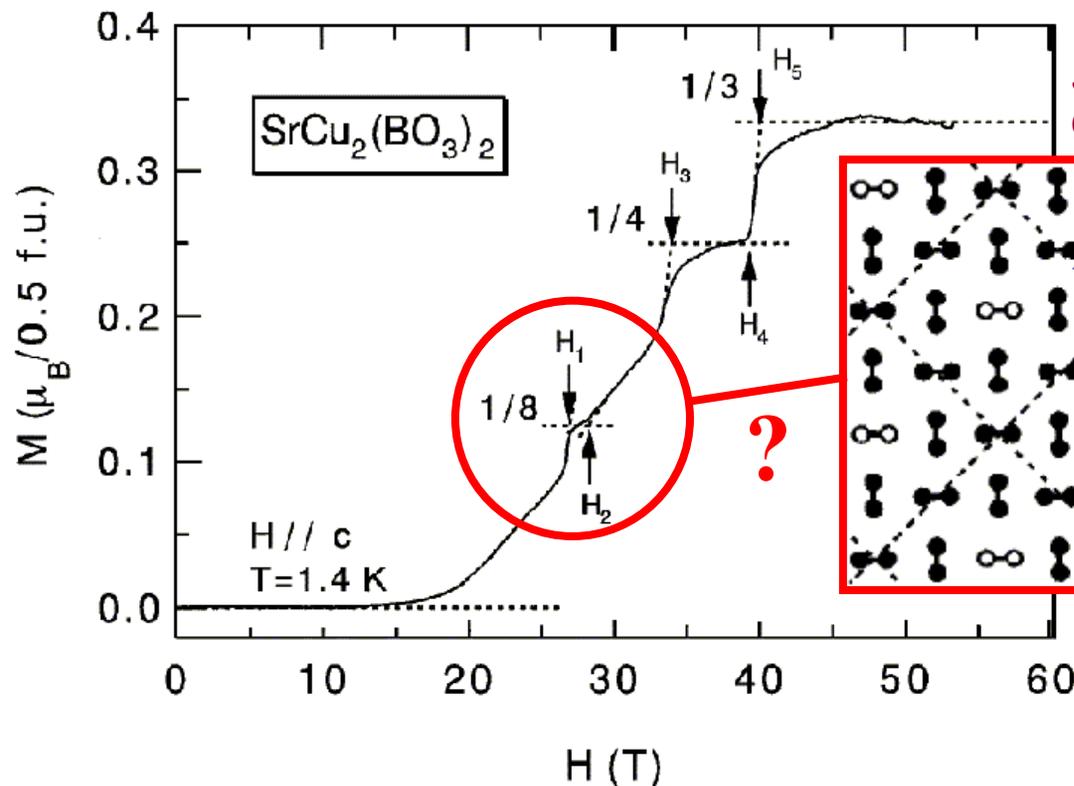
⇒ can easily crystallize !



"Magnetic crystal" ?

1/8
plateau ?

Problem:

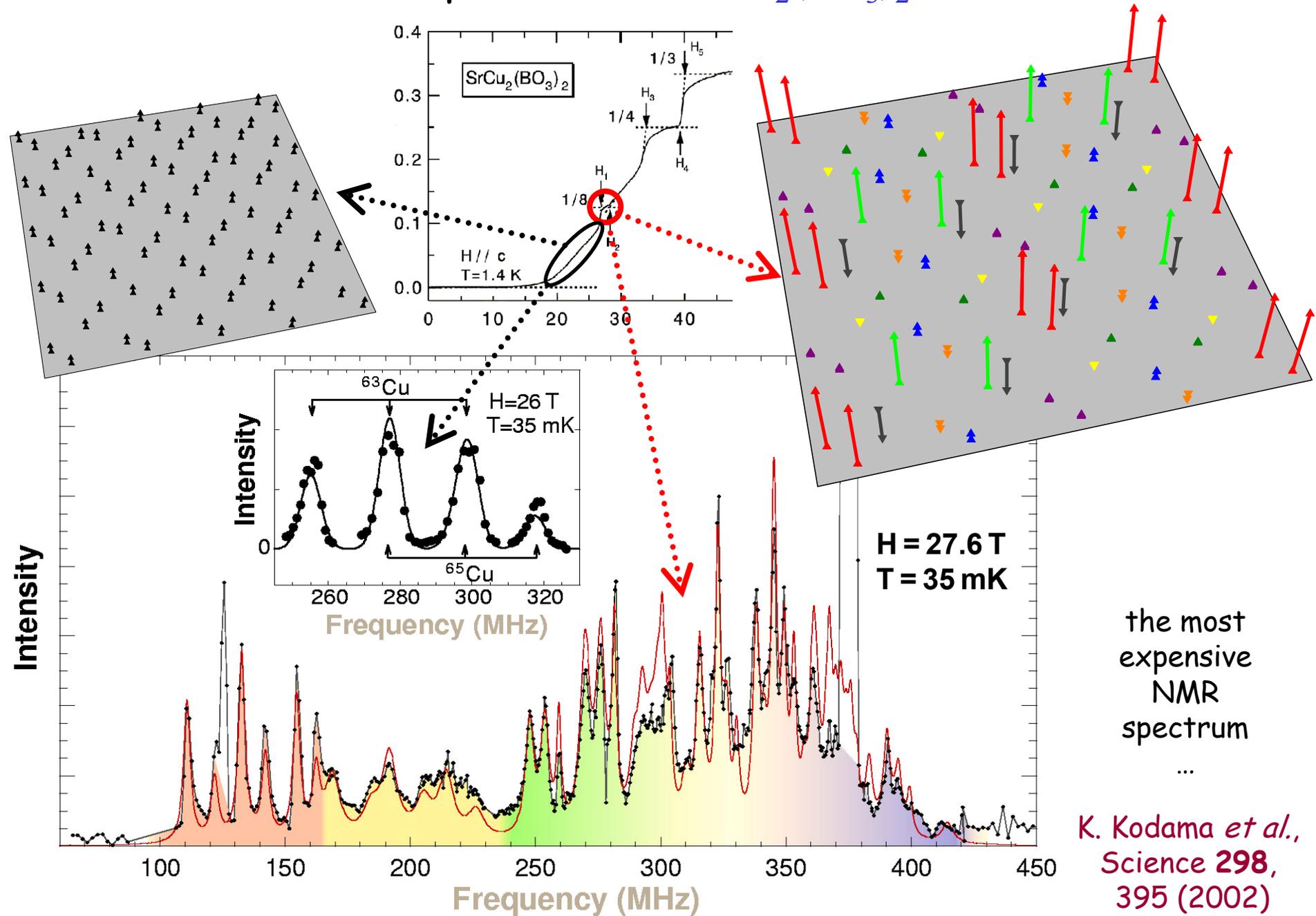


K. Onizuka, *et al.*,
J. Phys. Soc. Jpn.
69, 1016 (2000)

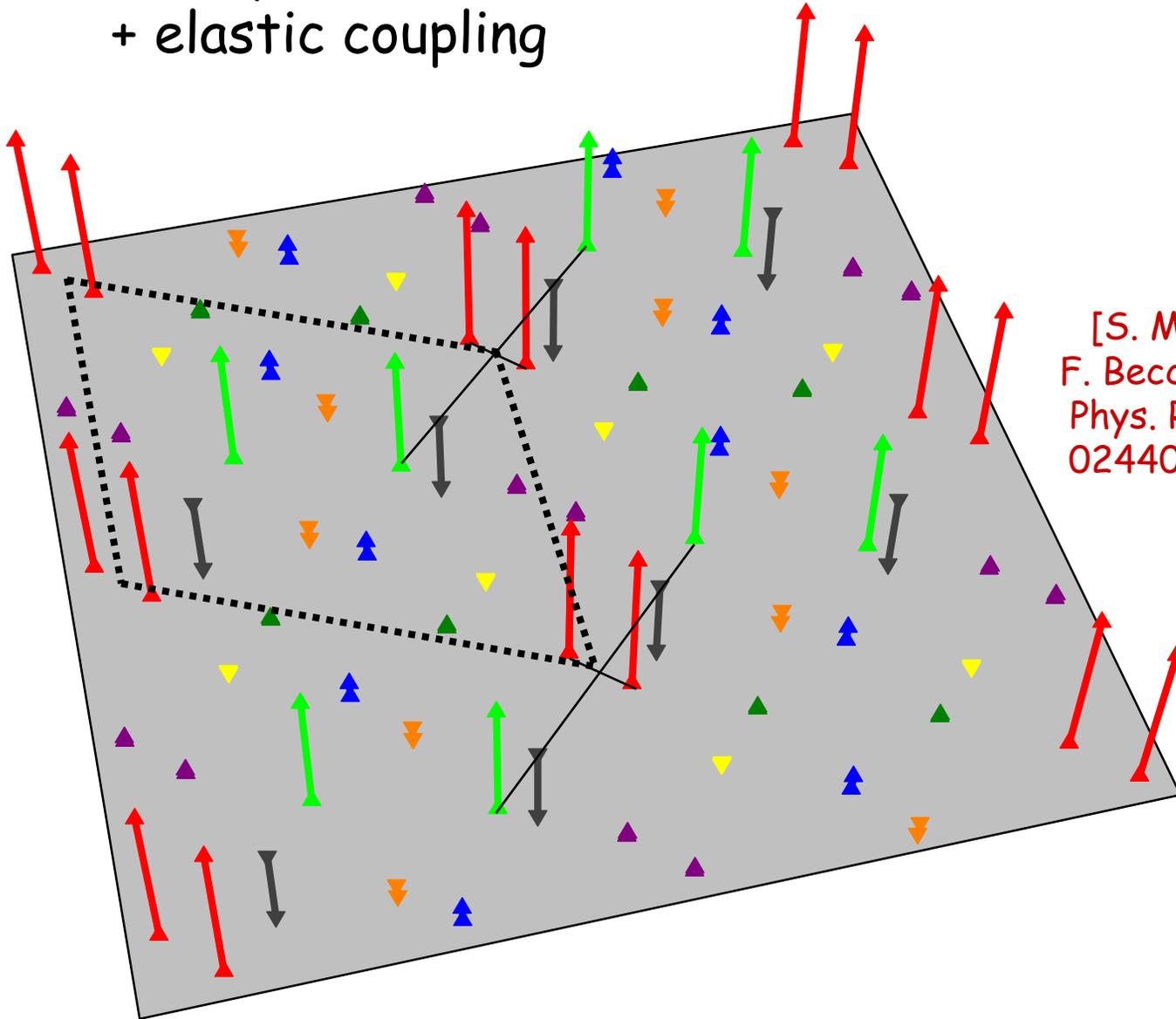
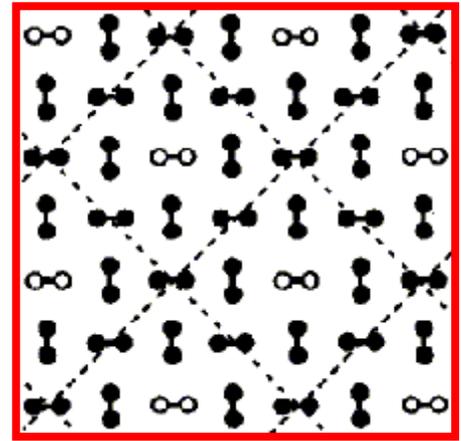
Plateaus of "fractional" magnetisation are supposed to be particularly stable, commensurate, spin-textured states.

Experimentally, due to high magnetic field involved, their magnetic structure can be accessed only by NMR !

$^{63,65}\text{Cu}$ ($I=3/2$) NMR spectrum of $\text{SrCu}_2(\text{BO}_3)_2$ at 35 mK



numerics on
Shastry-Sutherland
+ elastic coupling



[S. Miyahara,
F. Becca, F. Mila,
Phys. Rev. B 68,
024401 (2003)]

$$\langle S_z \rangle =$$

0.386

-0.215

0.312

0.010

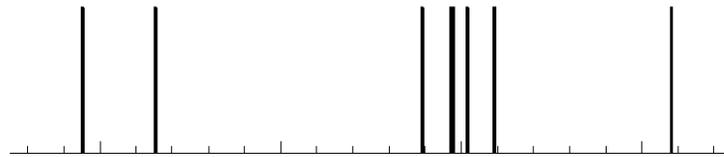
-0.035

0.040

-0.0063

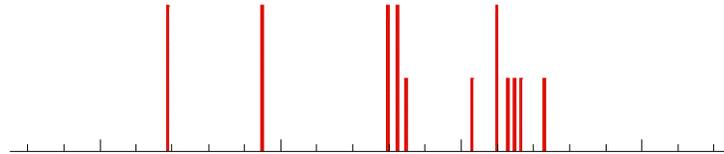
0.0073

Theory + on-site
hyperfine coupling:

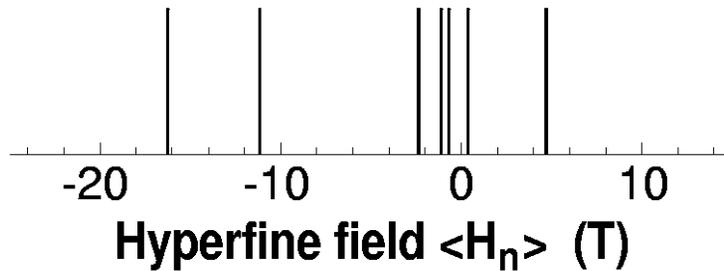
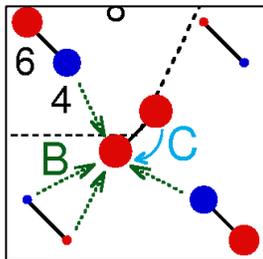


$$A = -23.8 \text{ T}/\mu_B$$

Experiment:



Theory + on-site
and NN coupling:



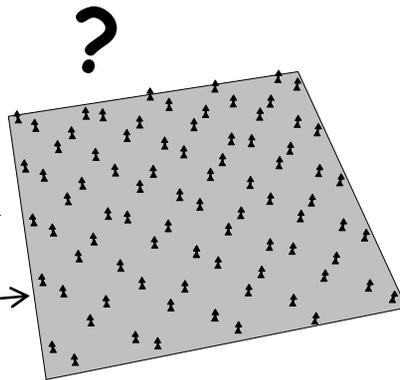
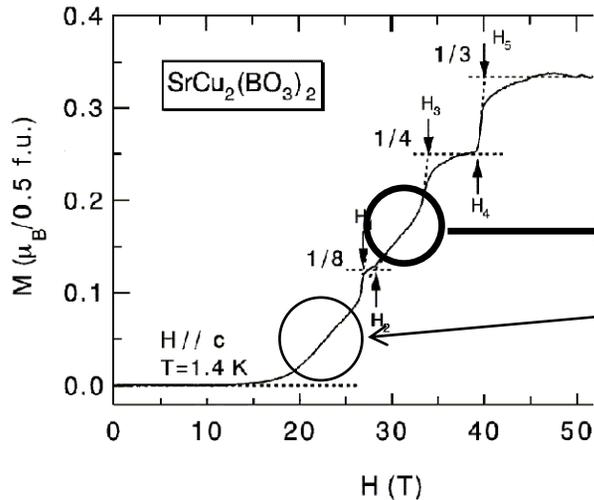
$$A + 4B + C = -23.8 \text{ T}/\mu_B$$



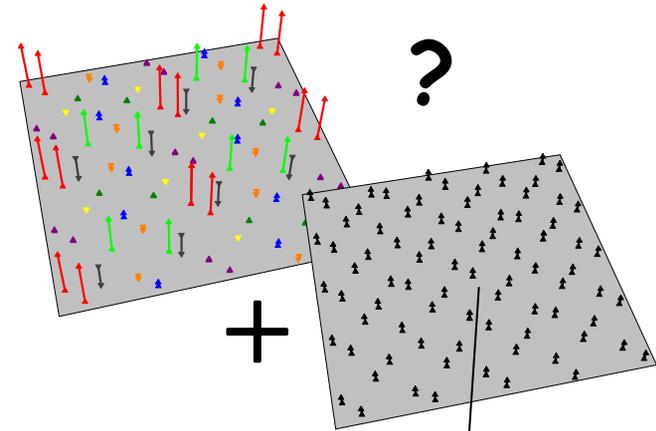
$$B = -1.04 \text{ T}/\mu_B$$

$$C = -2.45 \text{ T}/\mu_B$$

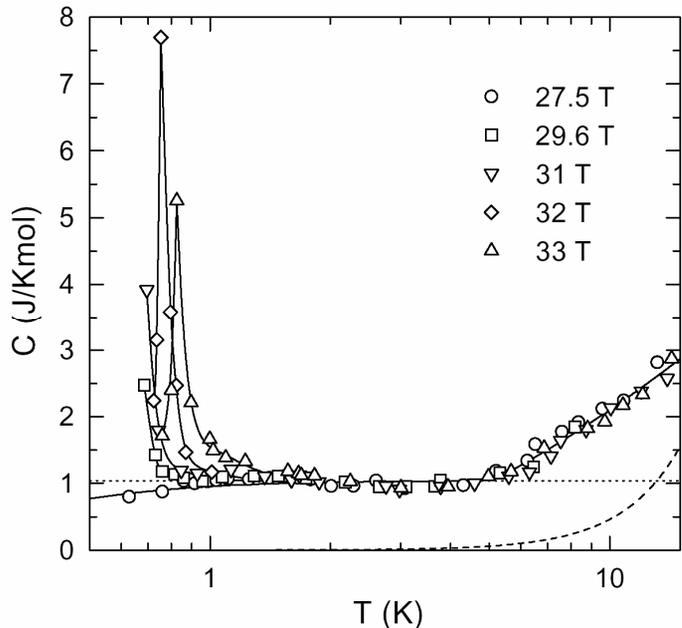
What is expected above the 1/8 plateau, at 28 - 33 T ?



or



if BEC \Rightarrow
supersolid



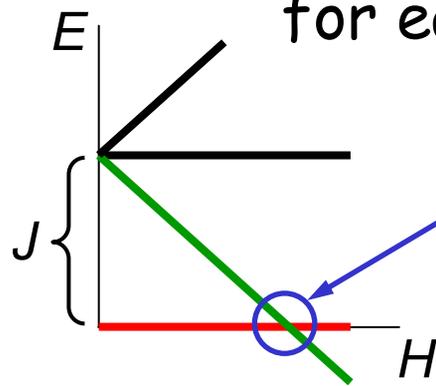
At and above plateau, **specific heat** sees only the same 2D gas of 1/8 of triplets, and not the particles that carry additional magnetization !

[H. Tsujii *et al.*, cond-mat/0301509]

Bose condensation of hard core bosons ?

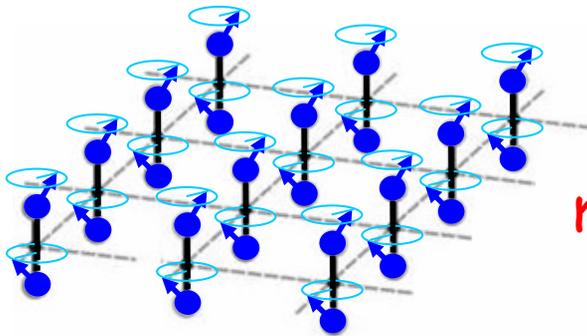
for each dimer:

[Momoi and Totsuka,
PRB 62, 15067 (2000)]

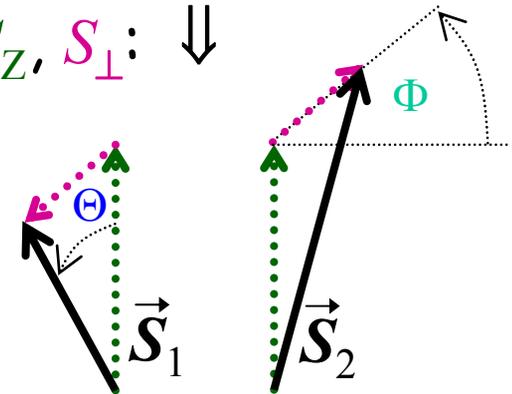


$$|\Phi, \Theta\rangle = e^{-i\Phi/2} \cos \Theta |\uparrow\uparrow\rangle + e^{+i\Phi/2} \sin \Theta \{|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle\} / \sqrt{2}$$

local projections of S_Z, S_{\perp} : \Downarrow

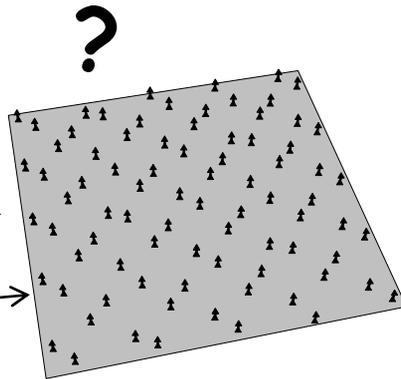
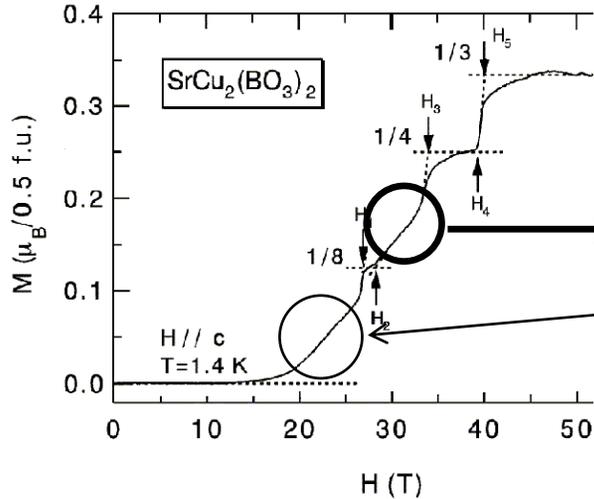


staggered S_{\perp}
moments - may order !
(gapless ODLRO)

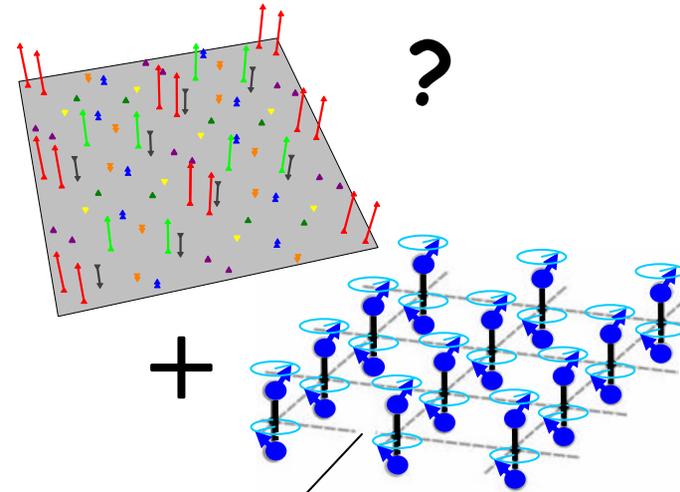


NMR should detect a **line-splitting** (into two lines)
corresponding to a Néel type order !

What is expected above the 1/8 plateau, at 28 - 33 T ?

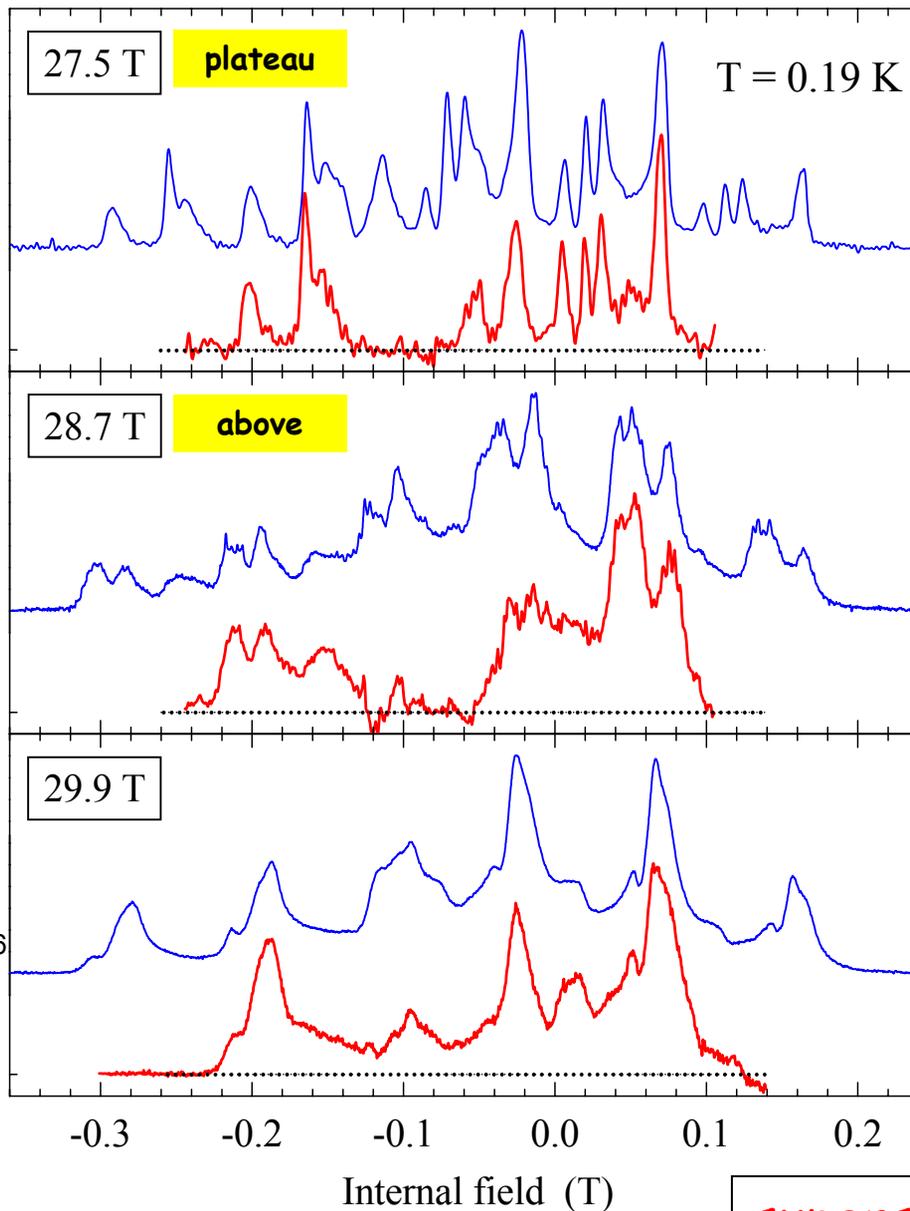
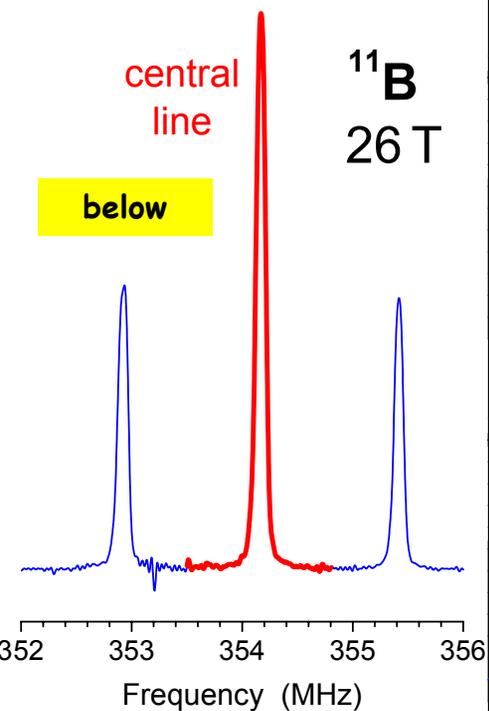
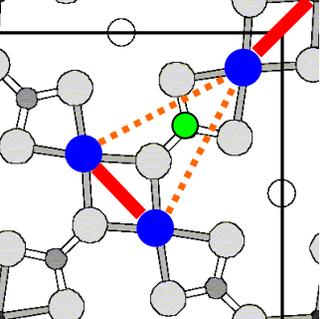


or

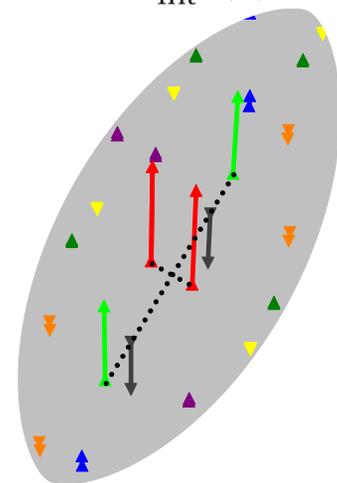
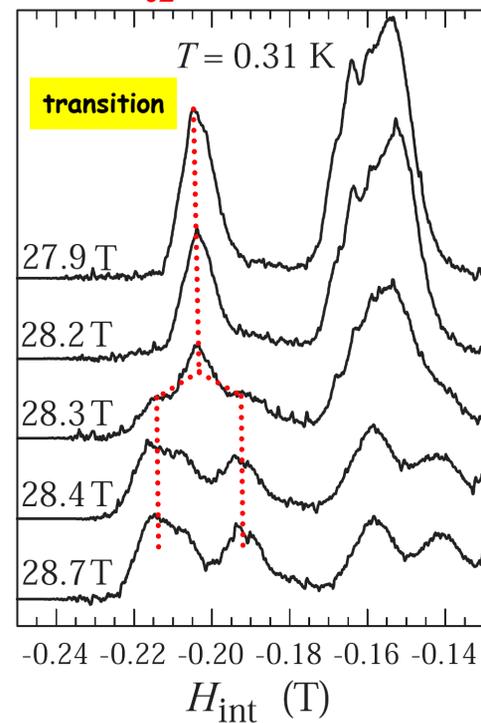


if BEC \Rightarrow **supersolid**
 but DM terms $\vec{D} \cdot \vec{S}_{i+\delta} \times \vec{S}_i$
 break the U(1) symmetry

^{11}B NMR above the 1/8 plateau



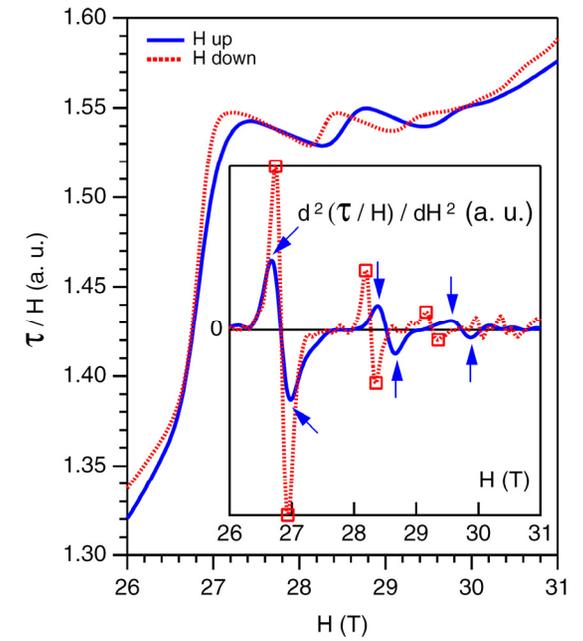
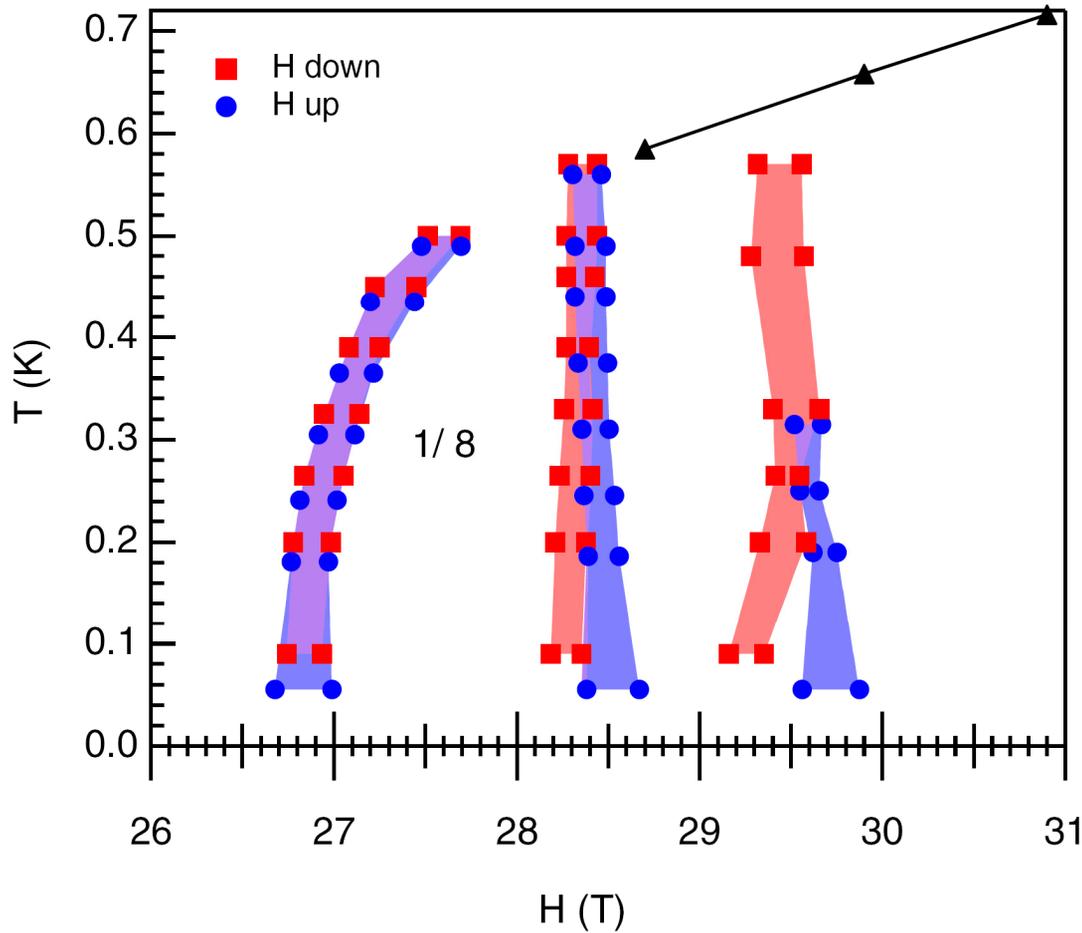
$$H_{c2} = 28.35 \text{ T}$$



M. Takigawa *et al.*,
PRL 101, 037202 (2008)

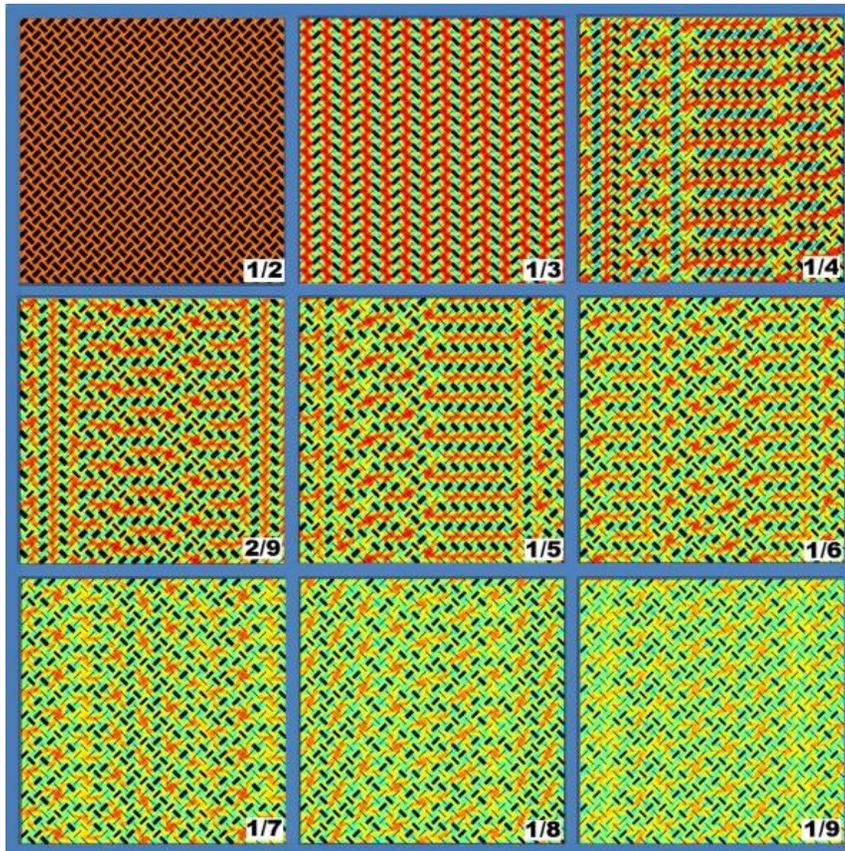
superstructure persists !

Torque:

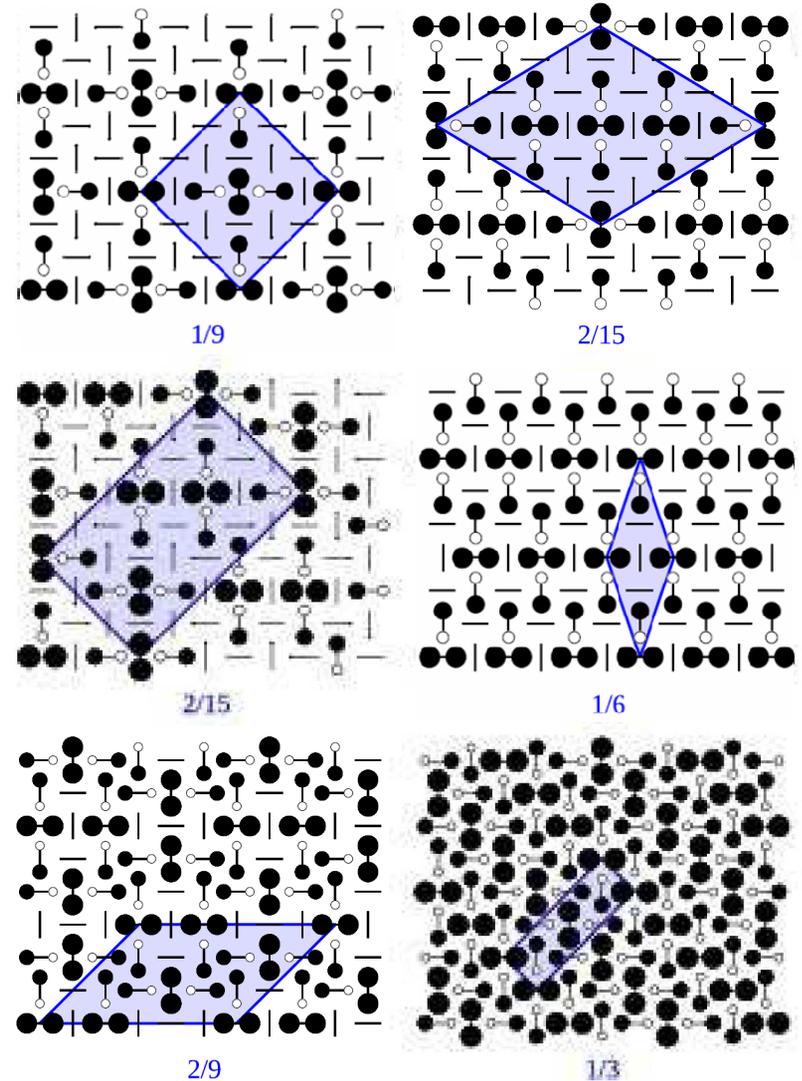


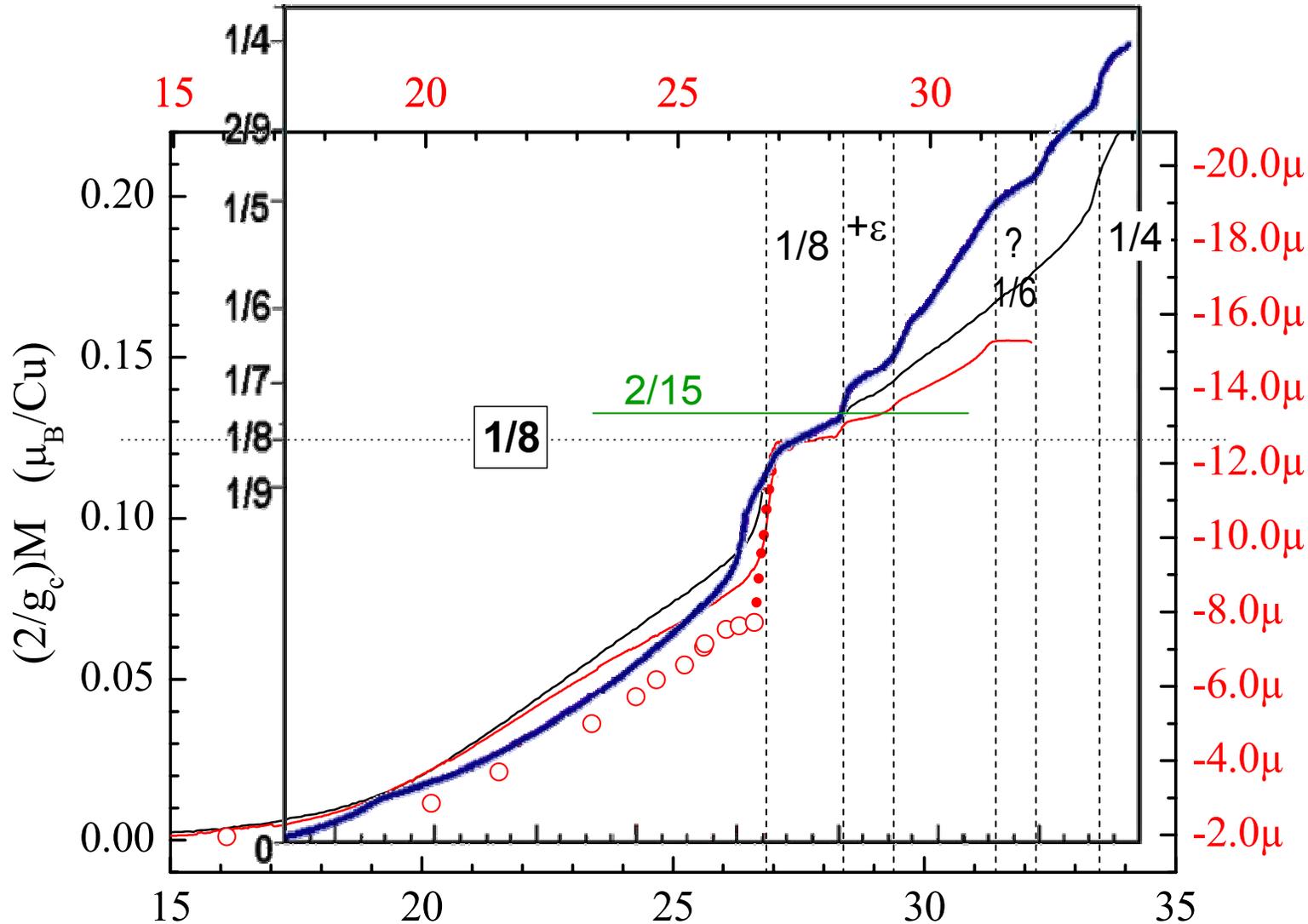
"only" $1/8$, $1/4$ and $1/3$?
or more ?

Sebastian S.E. *et al.*,
arXiv:0707.2075



J. Dorier, K. P. Schmidt & F. Mila,
arXiv:0806.3406





○ NMR lineposition

• NMR intensity in the mixed phase

— pulsed field at 1.5 K (+ adiabatic cooling),
K. Onizuka *et al.*, J. Phys. Soc. Jpn. **69**, 1016 (2000).

— by torque @ GHMFL,
F. Lévy *et al.*, EPL **81**, 67004 (2008) and unpublished

— by torque @ NHMFL,
S. Sebastian *et al.*, arXiv:0707.2075

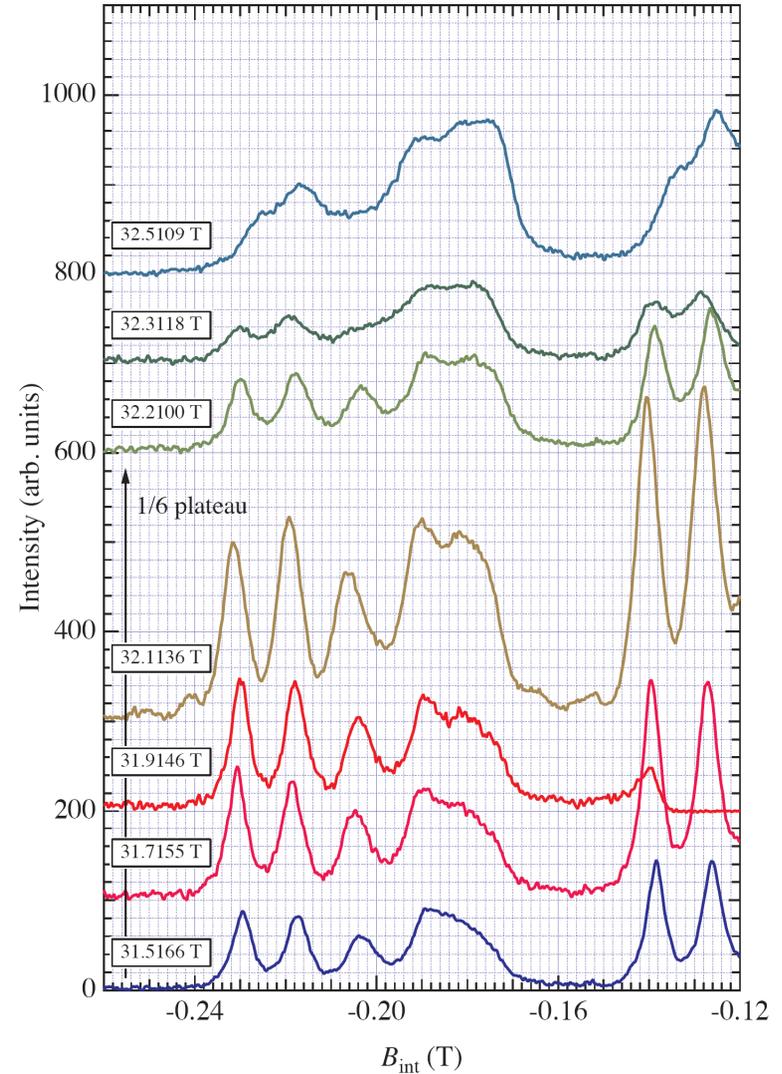
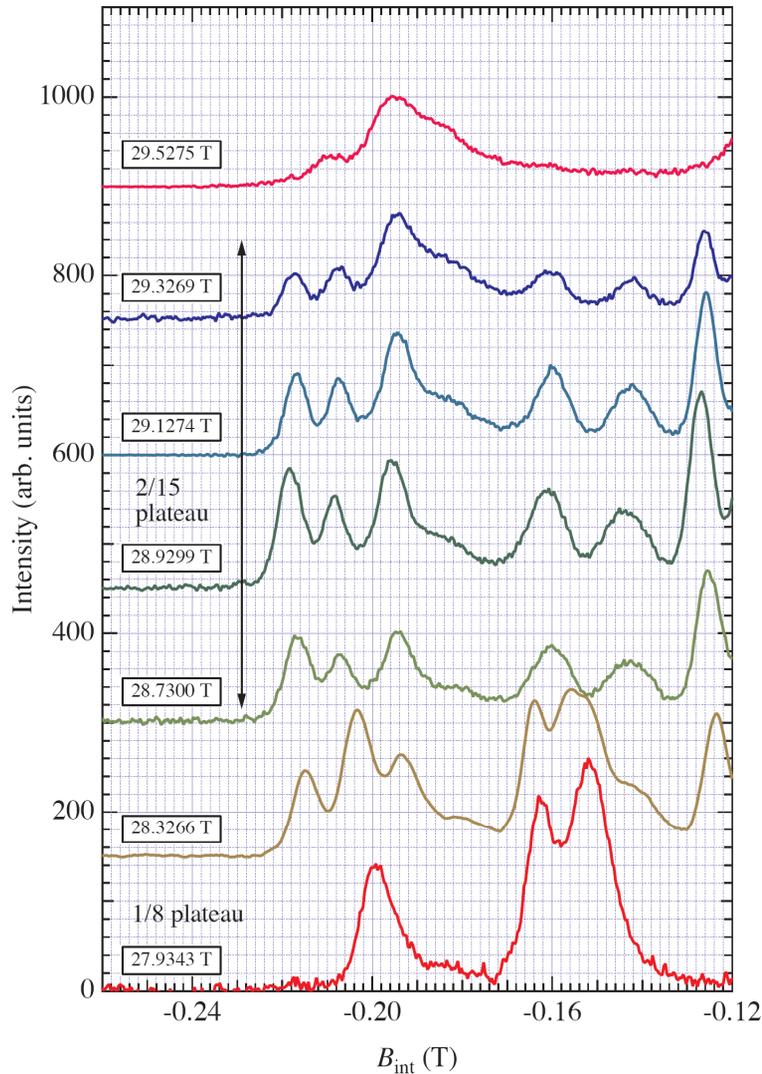
Theory: new fractions by Dorier, Schmidt & Mila, arXiv:0806.3406: **1/9, 2/15, 1/6, 2/9, 1/3**

Plateau: field independent spin superstructure / NMR spectra

$1/8 + \epsilon$

$T = 0.43 \text{ K}$

$1/6 ?$



Continuous field dependence in-between the plateaus

$1/8 + \epsilon$

\leftrightarrow

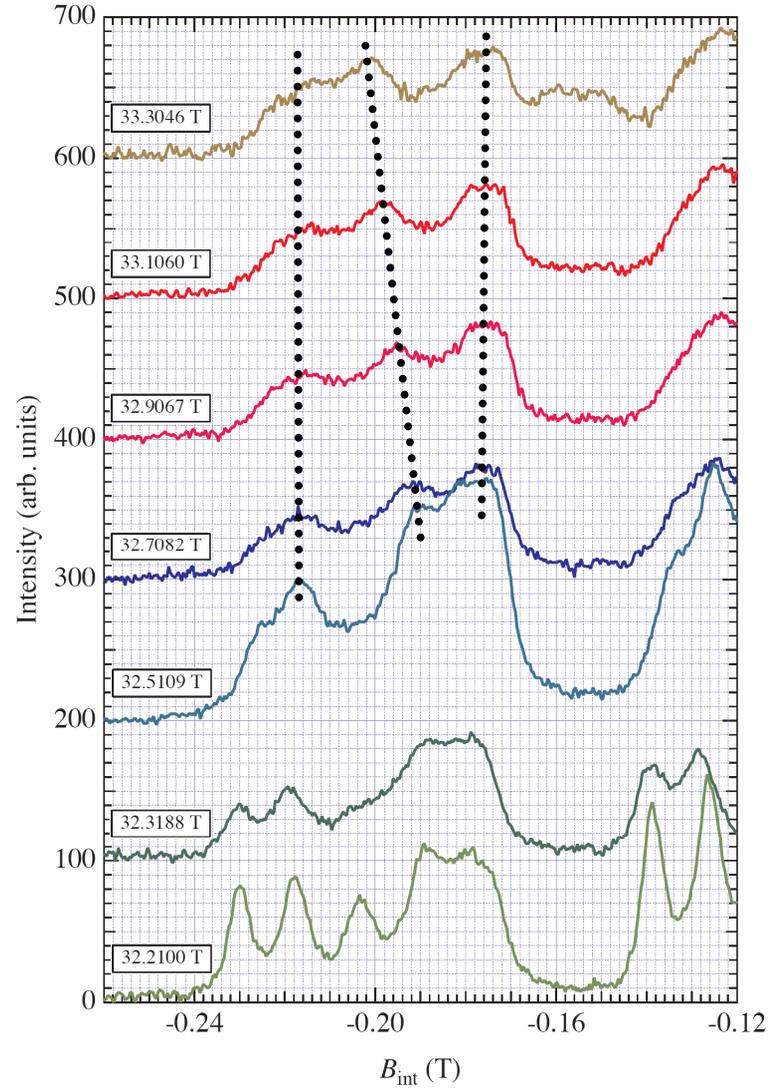
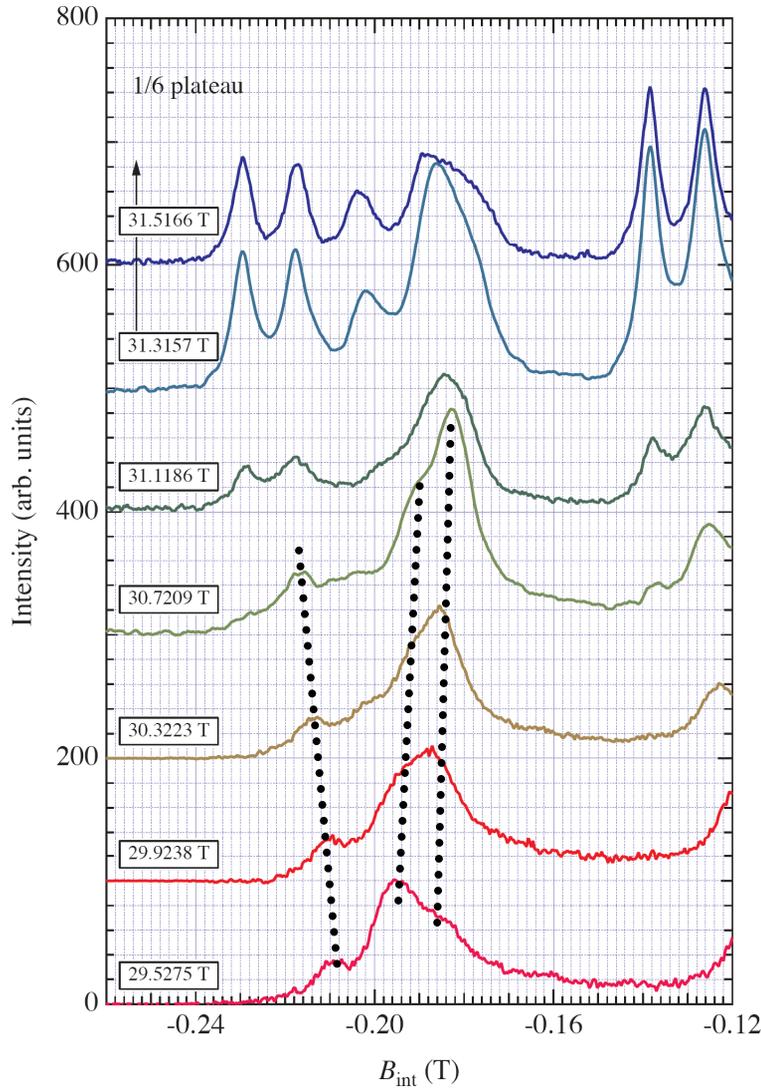
$1/6 ?$

$T = 0.43 \text{ K}$

$1/6 ?$

\leftrightarrow

" $1/4$ "



phase diagram:

also by: **torque**
 F. Lévy *et al.*, EPL **81**, 67004 (2008) and unpublished

□ **specific heat**
 H. Tsujii *et al.*, cond-mat/0301509

linesplitting:

supercell
 of extended triplets

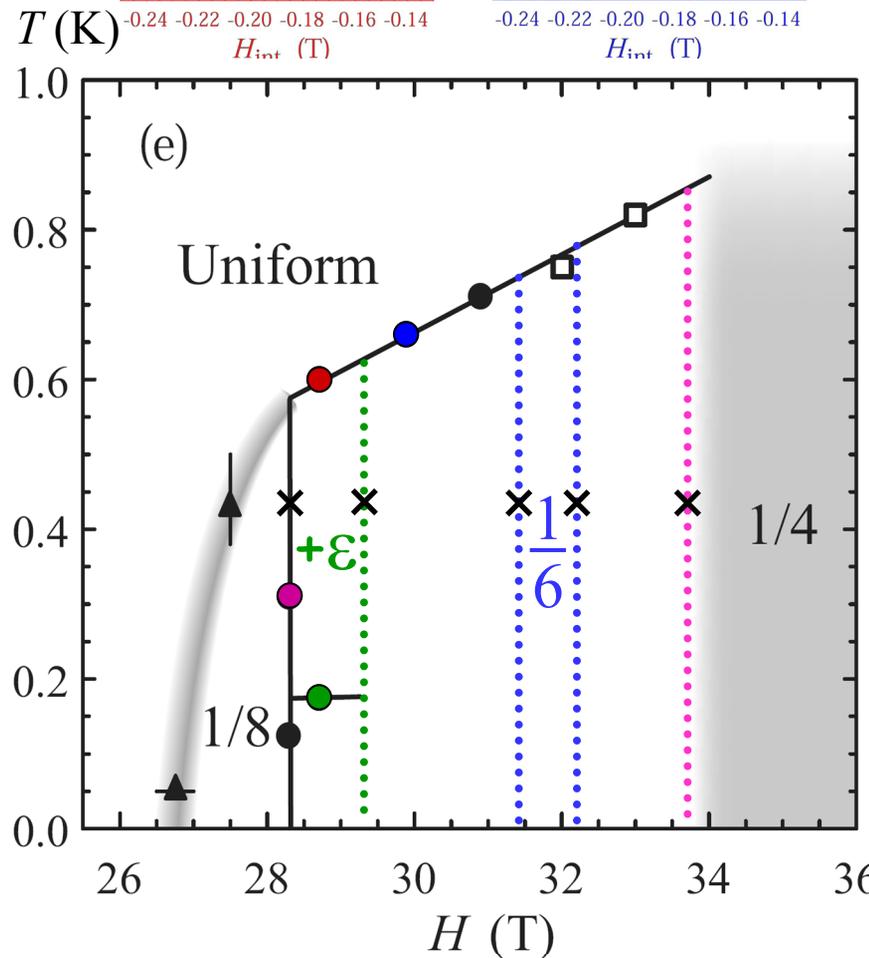
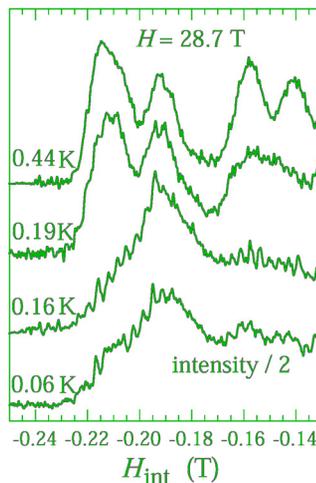
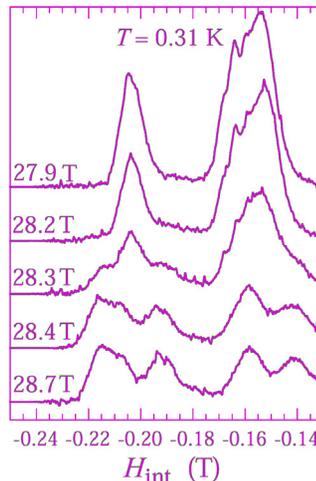
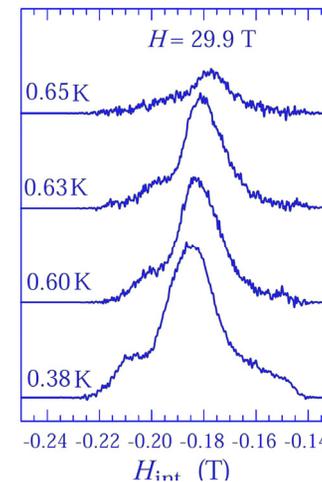
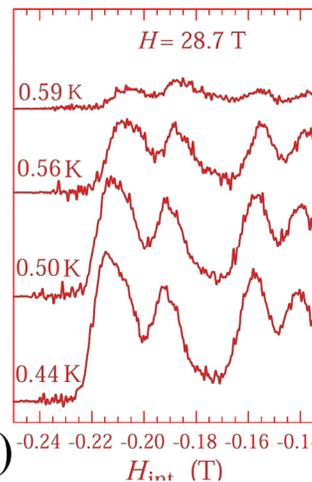
or

"quasi-BEC"
 staggered
 moments ?

[rotational symmetry
 already broken by DM
 $(\vec{D} \cdot \vec{S}_1 \times \vec{S}_2)$ interaction]

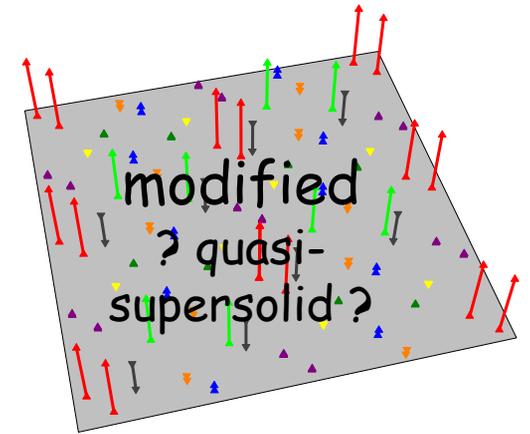
by **NMR**:

- ▲ K. Kodama *et al.*, Science **298**, 395 (2002)
- M. Takigawa *et al.*, PRL **101**, 037202 (2008) and
- × unpublished (June 2008)



SrCu₂(BO₃)₂ conclusions:

[M. Takigawa *et al.*, PRL 101, 037202 (2008) and unpublished]



Spin superstructure persists

in-between "1/8" and "1/4" plateau

New plateau phases discovered:

1/8+ ϵ or 2/15 (?) and 1/6 (?)

NMR line-splitting in these phases:

"supercell" or "quasi-supersolid" (BEC) ?



NMR: simulate spectra, importance of staggered moments ...

Theory: stable structures, supersolid in presence of symmetry breaking DM terms ?