Current Frontiers in Condensed Matter Physics
ICTS, Bangalore, 2016.6.20-29

Magnetic properties of volborthite determined by a coupled-trimer model

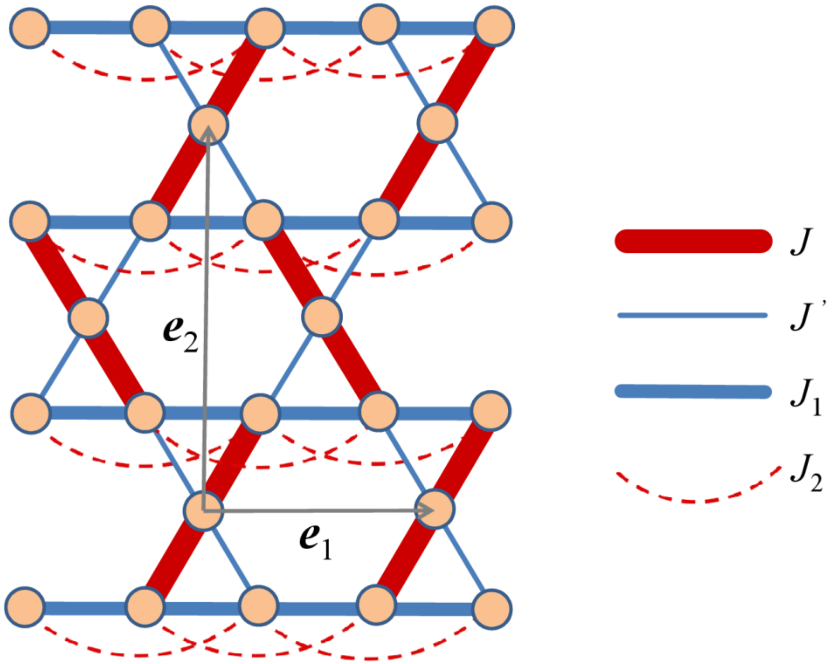
Shunsuke Furukawa
Dept. of Physics, Univ. of Tokyo

東京大学
THE UNIVERSITY OF TOKYO

O. Janson *et al.*, arXiv:1509.07333
(to appear in Phys. Rev. Lett.)

✓Volborthite = coupled trimers

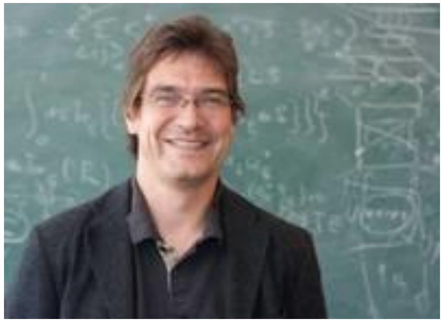
✓Scenario for a bond nematic state in a magnetic field



Collaboration

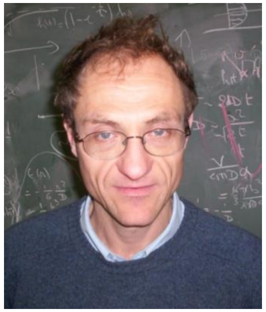
DFT+U modeling

O. Janson (TU Wien)



K. Held (TU Wien)

Exact diagonalization



P. Sindzingre (Paris VI)

J. Richter (Magdeburg)

Effective model analysis

S. F.
(Univ. of Tokyo)

T. Momoi (RIKEN)

Introduction: spin-1/2 kagome antiferromagnet (AFM)

- Candidate of a quantum spin liquid

Schwinger boson: Sachdev, PRB 45, 12377 (1992)

DMRG: Gapped Z_2 spin liquid (S_{topo}=log 2)

S. Yan *et al*., Science **332**, 1173 (2011)
Depenbrock *et al*., PRL **109**, 067201 (2012)
H.-C. Jiang *et al*., Nat. Phys. **8**, 902 (2012)

Variational MC: Gapless spin liquid

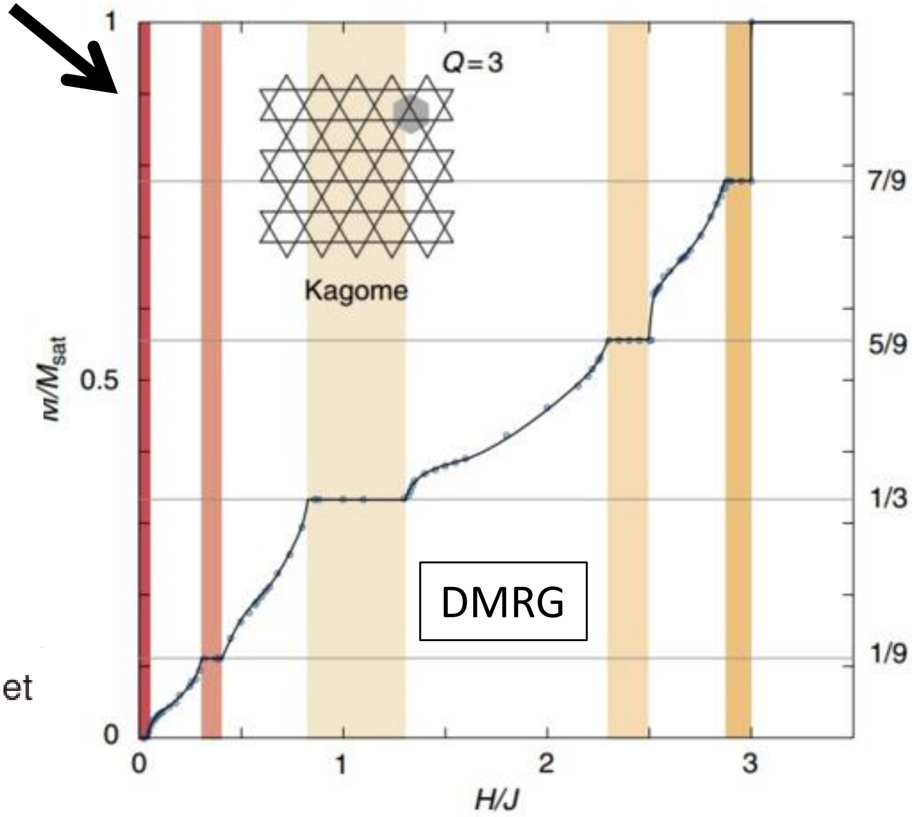
Y. Ran et al., PRL 98, 117205 (2007)
Y. Iqbal et al., PRB 89, 020407 (2014)

- Unusual magnetization process

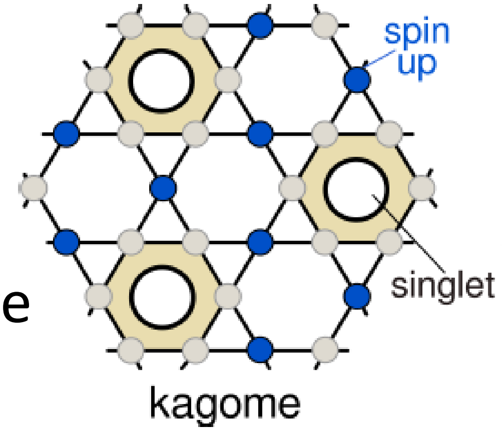
Nishimoto *et al*. Nat. Comm. **4**, 2287 (2013)
Capponi *et al*., PRB **88**, 144416 (2013)
Schlenburg *et al*., PRL **88**, 167207 (2002)

Widest plateau at M/M_{sat}=1/3

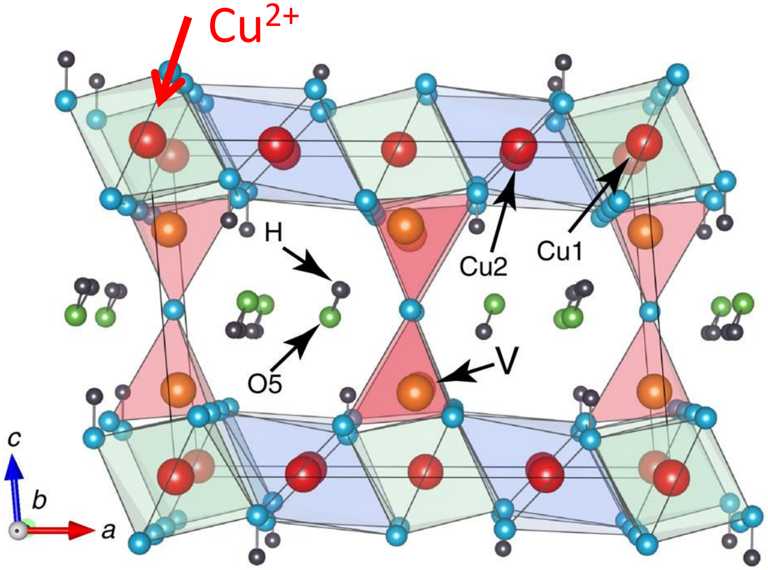
Hida, JPSJ **70**, 3673 (2001)
Cabra *et al*., PRB **71**,144420 (2005)



Quantum nature



Volborthite $Cu_3V_2O_7(OH)_2 2H_2O$

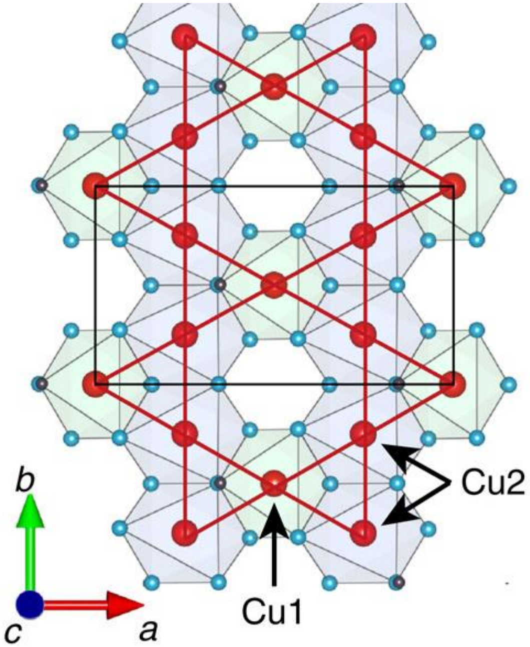


Spin-1/2 (Cu^{2+}) kagome layer

Non-magnetic V_2O_7 pillars

Spin-1/2 (Cu^{2+}) kagome layer

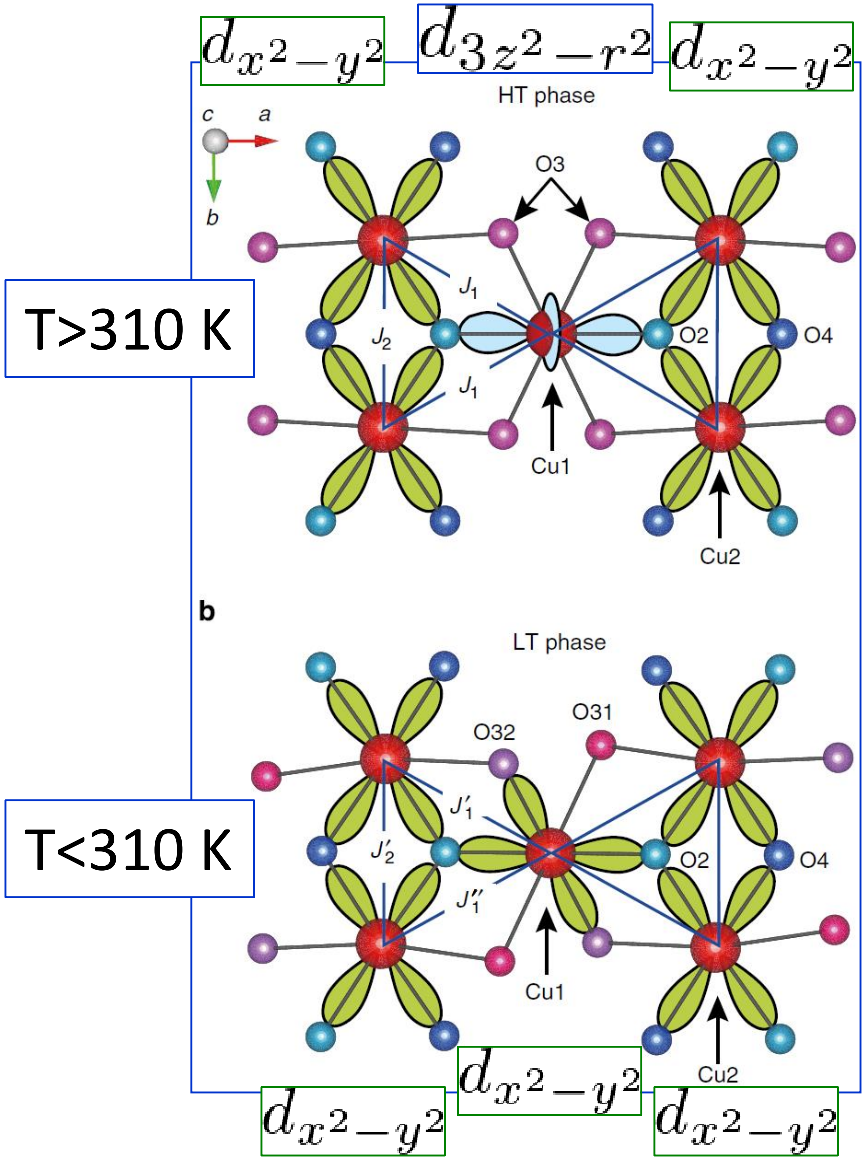
www.webmineral.com



A spin-1/2 kagome antiferromagnet ?

Hiroi *et al.*, JPSJ **70**, 3377 (2001)

Orbital arrangement



T>310 K

$d_{x^2-y^2}$ $d_{3z^2-r^2}$ $d_{x^2-y^2}$

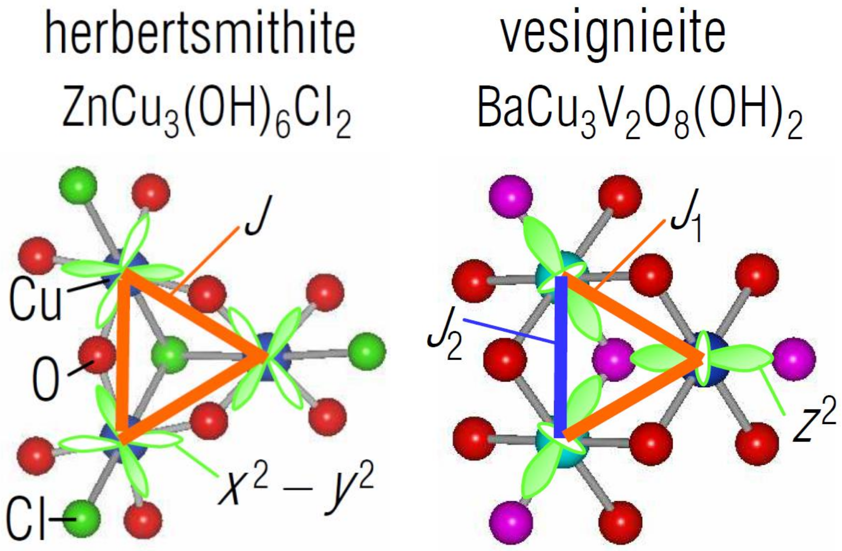
T<310 K

$d_{x^2-y^2}$ $d_{x^2-y^2}$ $d_{x^2-y^2}$

Single-crystal X-ray diffraction:
orbital switching at T=310 K

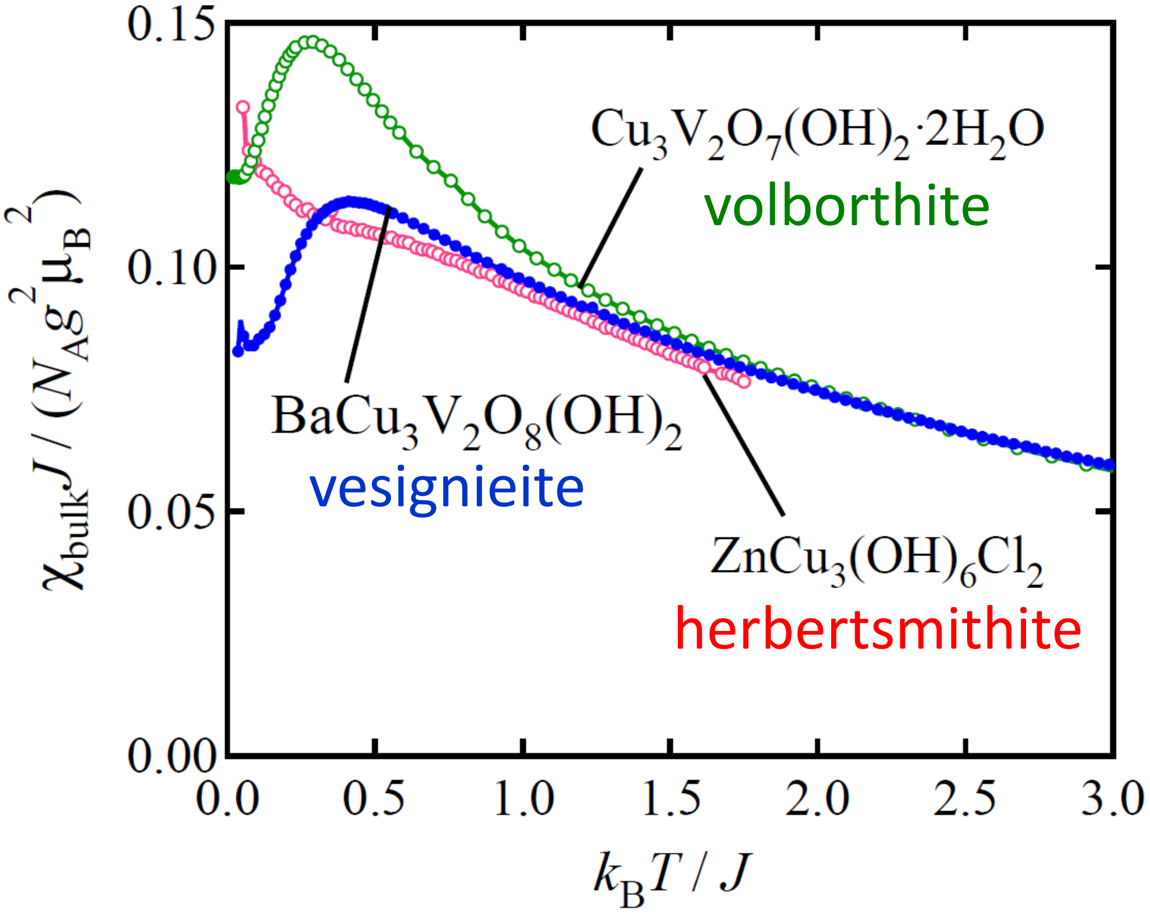
H. Yoshida *et al*., Nat. Comm. **3**, 860 (2012)

cf. Other kagome candidates



Y. Okamoto et al.,
J. Phys. Soc. Jpn. 78, 033701 (2009)

Comparisons of magnetic susceptibilities



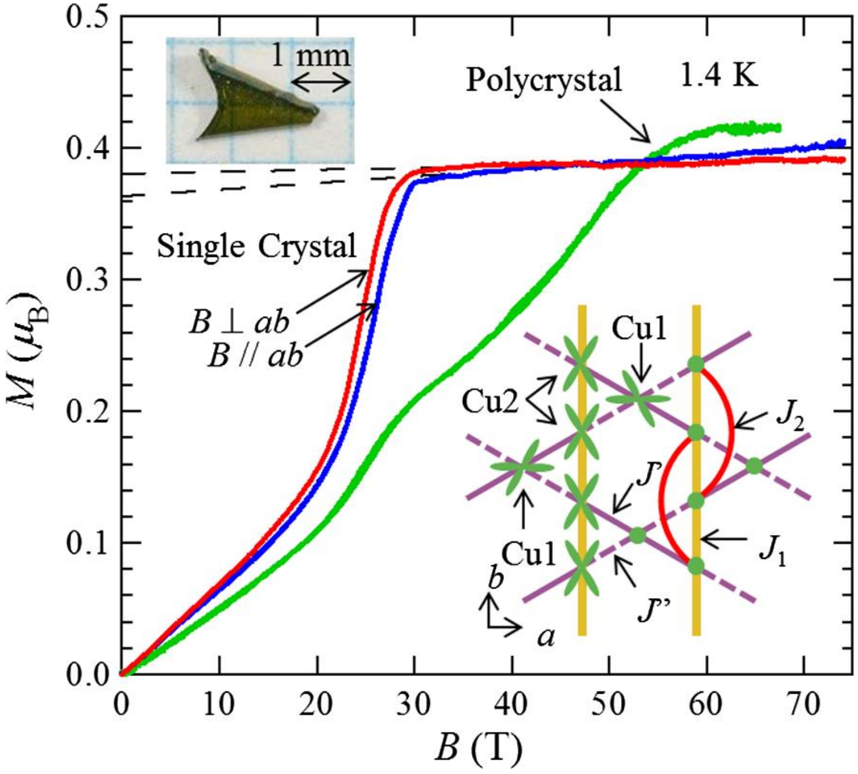
Y. Okamoto et al.,
JPSJ 78, 033701 (2009)

Volborthite: Deviation from kagome AFM? (-> less frustrated?)

Yet shows rich field-induced phenomena.

Magnetization process of a single crystal

Ishikawa *et al.*, PRL **114**, 227202 (2015)



Wide 1/3 plateau

26 T → 74 T → Over 100 T ?!

Faraday rotation exp.
by T. Yamashita *et al.*,
JPS Meeting, 2015.3

Inconsistent with a relatively short 1/3 plateau in kagome AFM

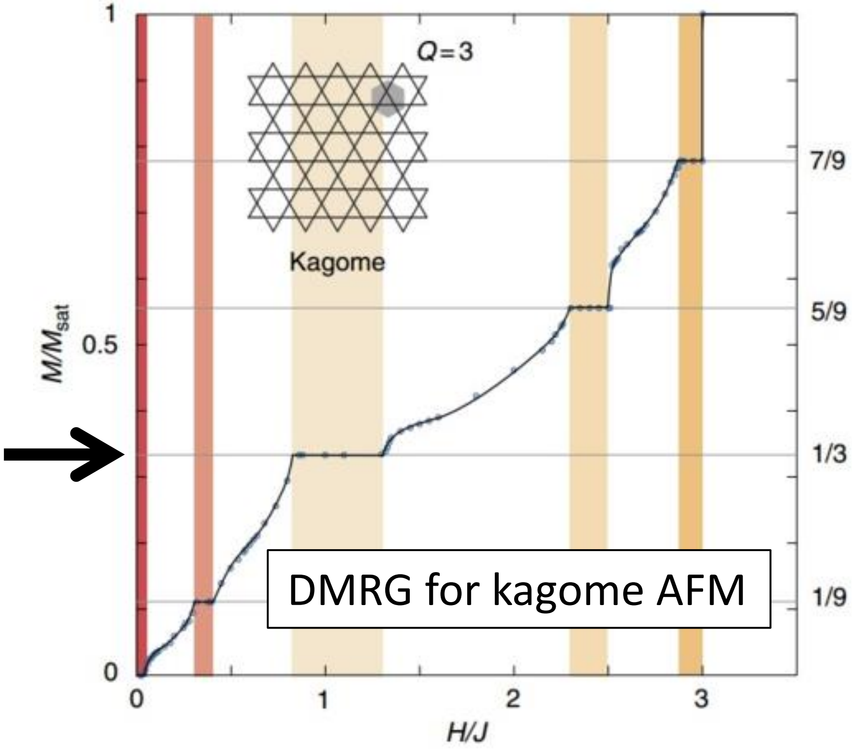
Theory

Nishimoto *et al*. Nat. Comm. **4**, 2287 (2013)

Capponi *et al*., PRB **88**, 144416 (2013)

Hida, JPSJ **70**, 3673 (2001)

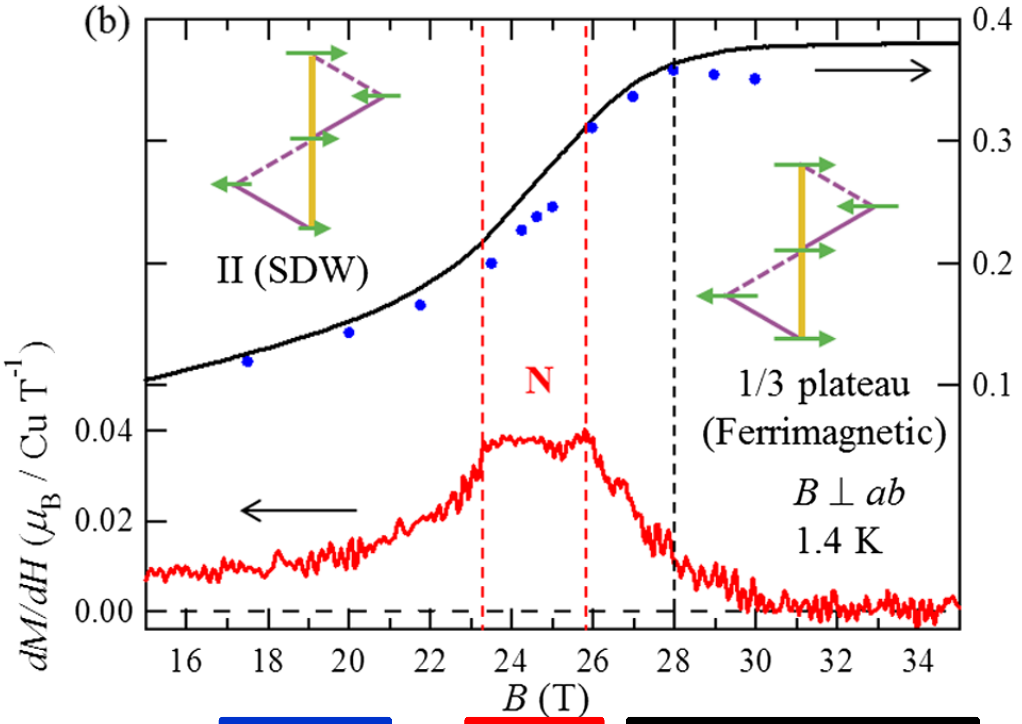
Nakano & Sakai, JPSJ **79**, 053707 (2010)



DMRG for kagome AFM

Field-induced phases of a single crystal - I

Ishikawa *et al.*, PRL **114**, 227202 (2015)



SDW

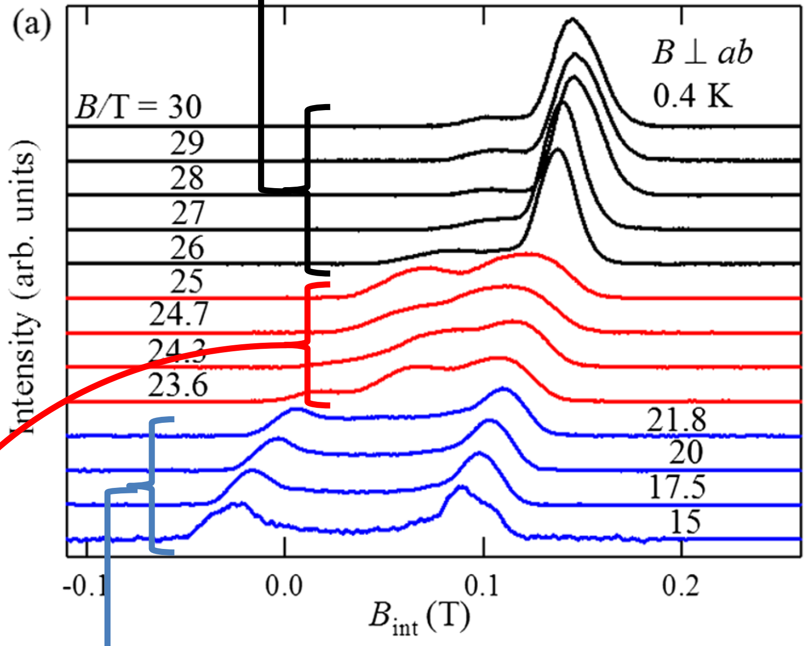
"N"

1/3-plateau

NMR spectra

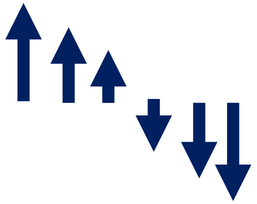
≈ Distribution of spin moments along the direction of B

1/3 plateau (ferrimagnetic)



Impossible to explain with conventional spin order

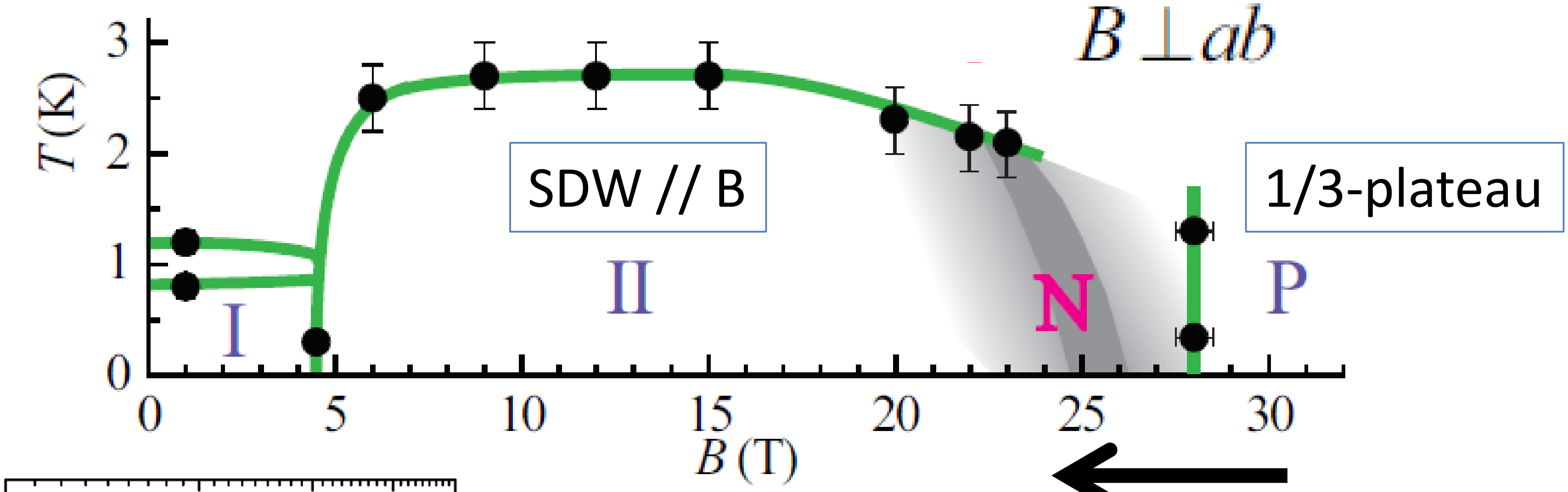
Double-horn: Typical of collinear SDW

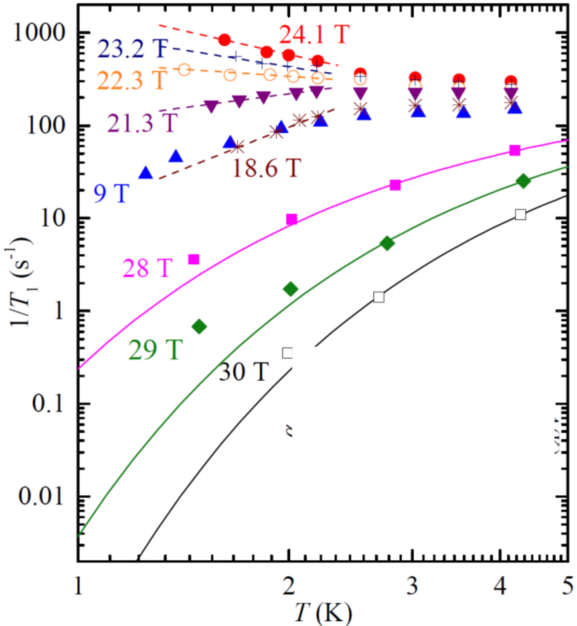


Field-induced phases of a single crystal - II

H. Ishikawa *et al.*, PRL **114**, 227202 (2015)

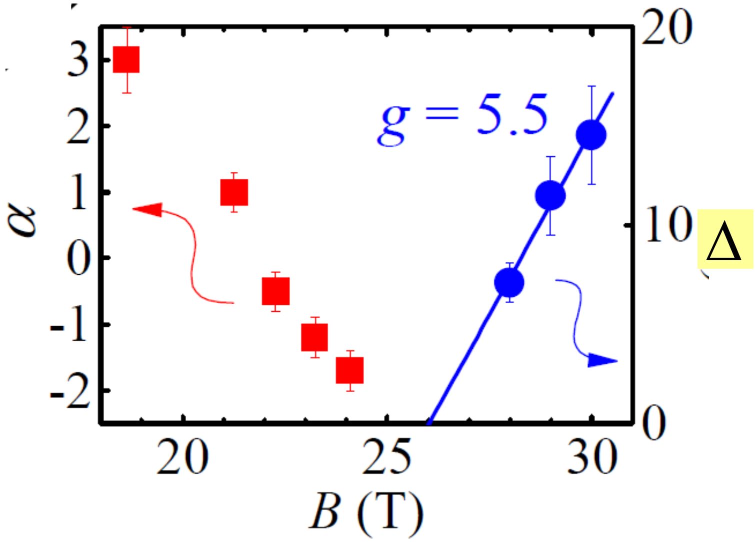
M. Yoshida *et al.*, arXiv:1602.04028





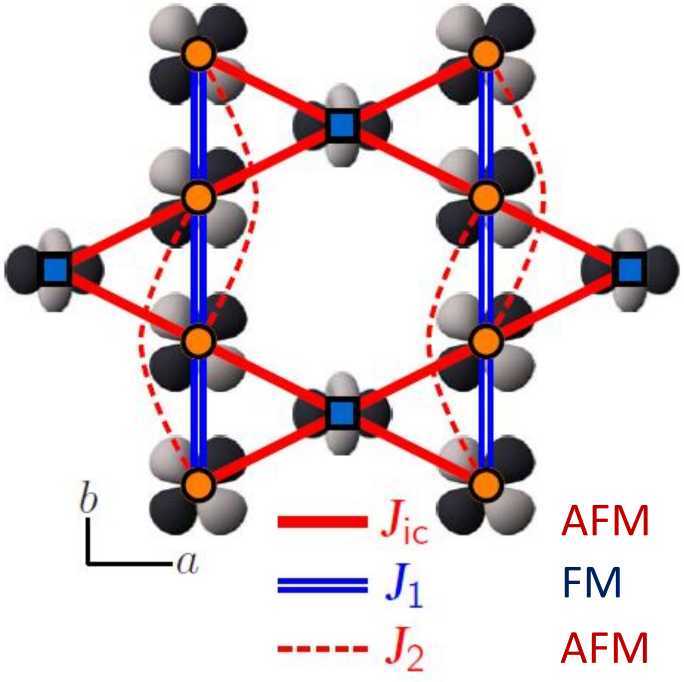
Estimation of a gap with $1/T_1 \propto \exp(-\Delta/T)$

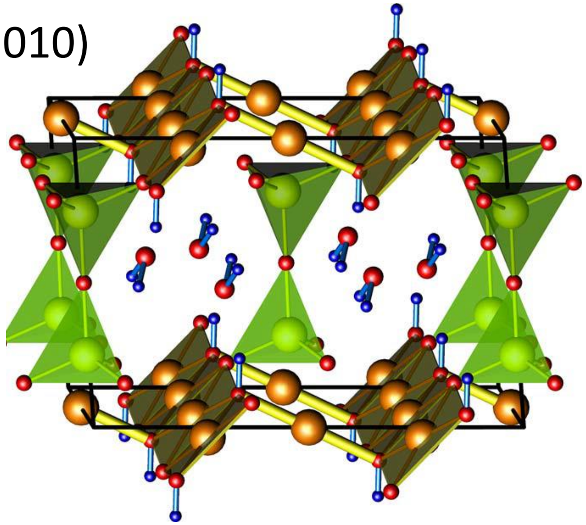
Condensation of multimagnon bound states?



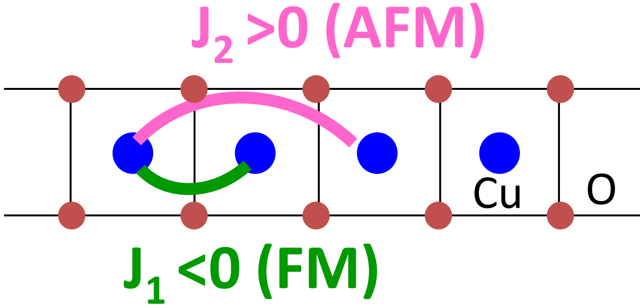
Previous model: coupled frustrated chains

O. Janson *et al.*, PRB **82**, 104434 (2010)

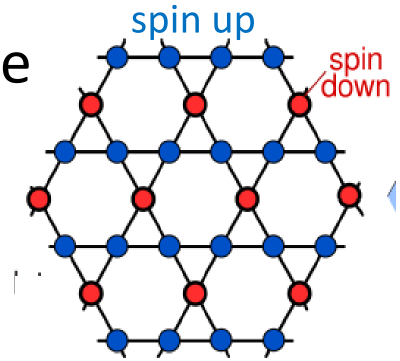




edge-shared CuO_2 chain as in $LiCuVO_4$

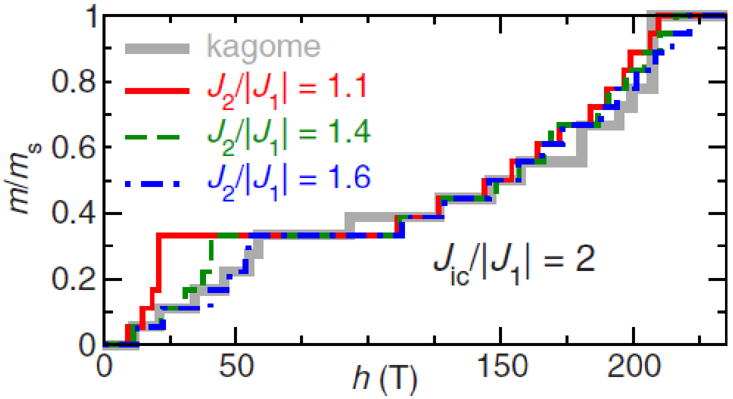


- ➢1/3-plateau: uud structure
- ➢Description of other field-induced phases: open problem



- ➢Based on powder structural data (closer to the high-T-phase structure of a single crystal)

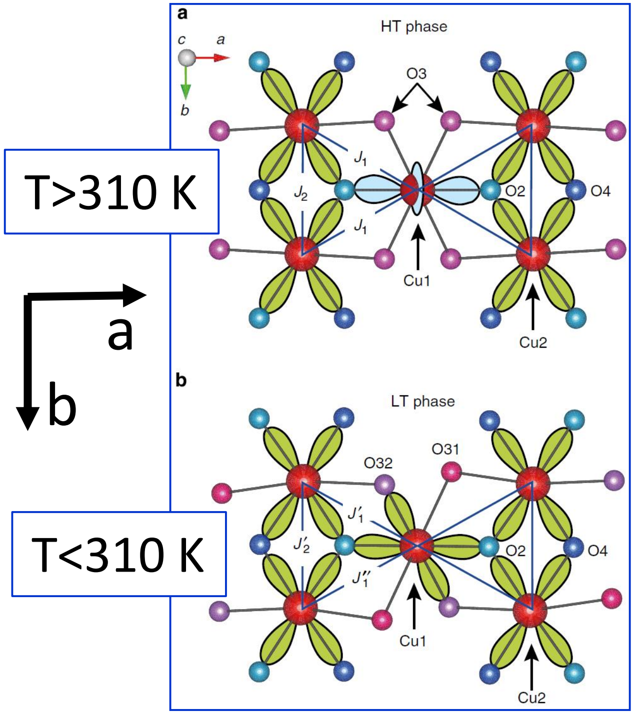
What is the model corresponding to the low-T-phase structure?

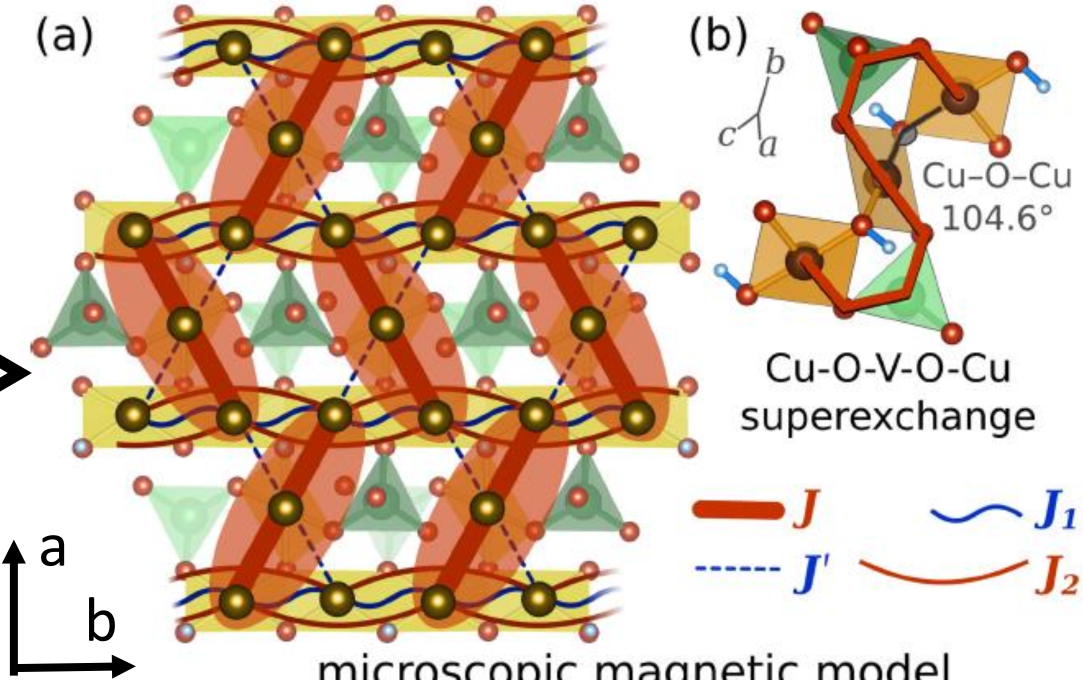


Modeling with single-crystal structural data

O. Janson *et al.*, arXiv:1509.07333

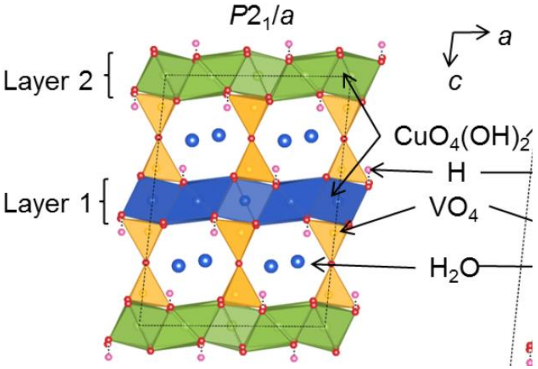
DFT+U calculation





microscopic magnetic model

Structural data:
Ishikawa *et al.*, Acta Crystallogr. **C68**, i41 (2012)



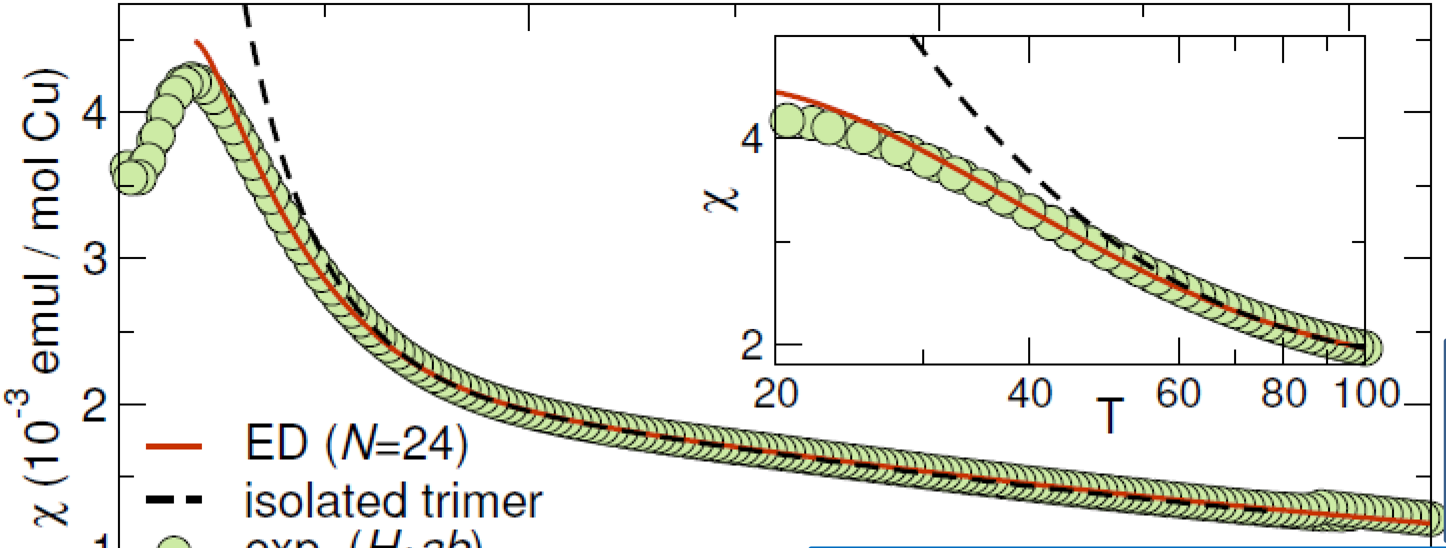
$J \simeq 200\ \mathrm{K}$ Coupled-trimer model!

$$J : J' : J_1 : J_2 = 1 : (-0.2) : (-0.5) : 0.2$$

Layer dependence is negligible.

Fit with susceptibility

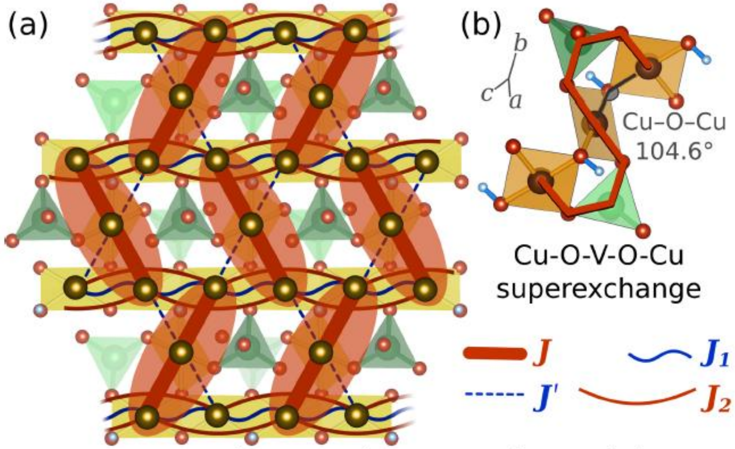
$$J \simeq 252\,\mathrm{K} \quad g = 2.151 \quad J : J' : J_1 : J_2 = 1 : (-0.2) : (-0.5) : 0.2$$



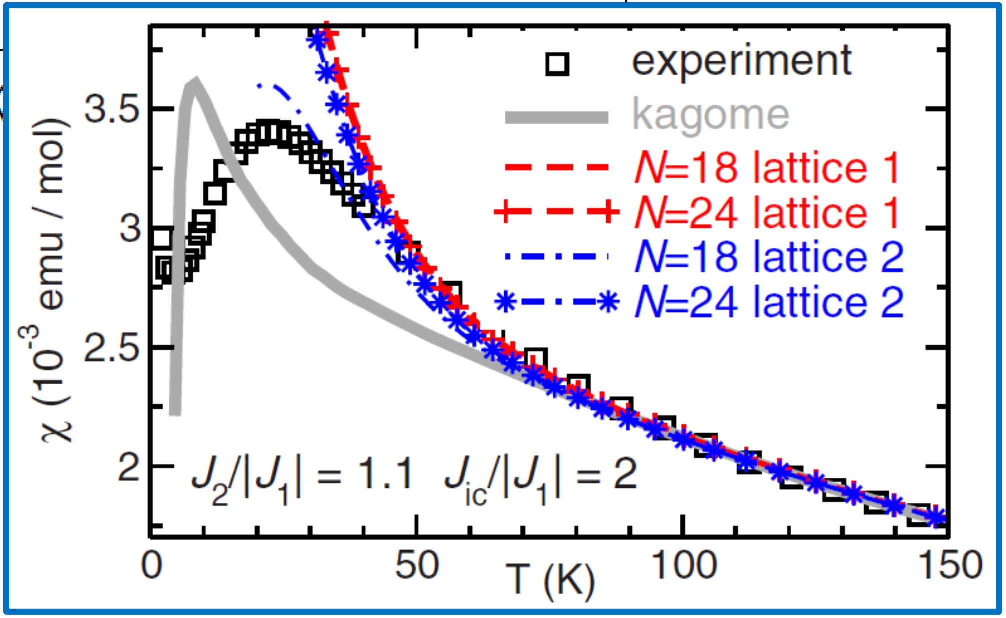
Agrees with exp. for

$T \gtrsim 30\,\mathrm{K}$

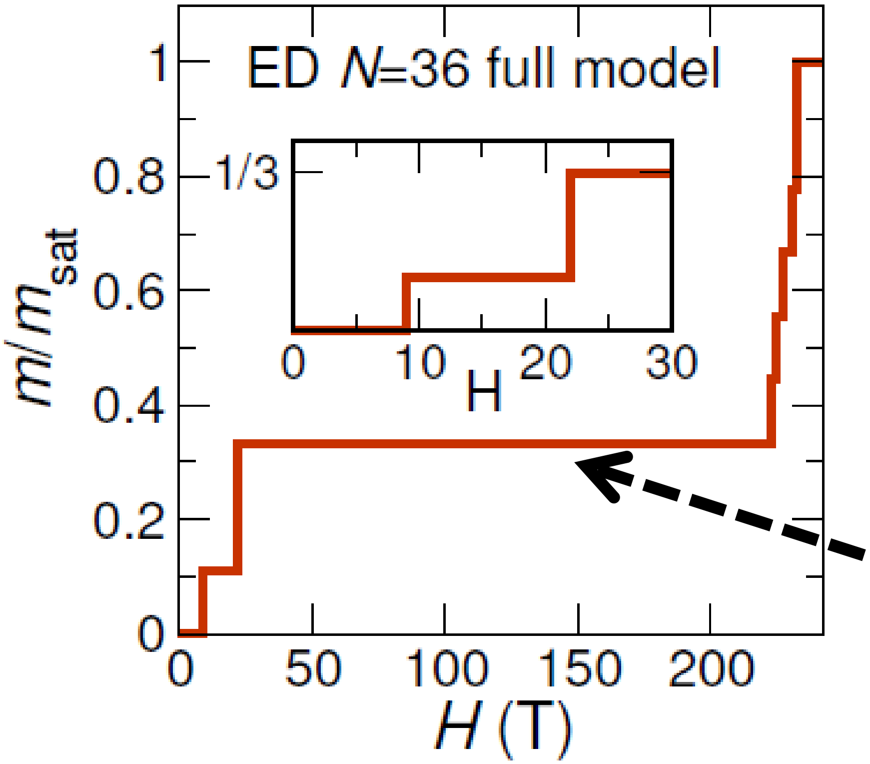
Previous model: agreement for $T \gtrsim 50\,\mathrm{K}$

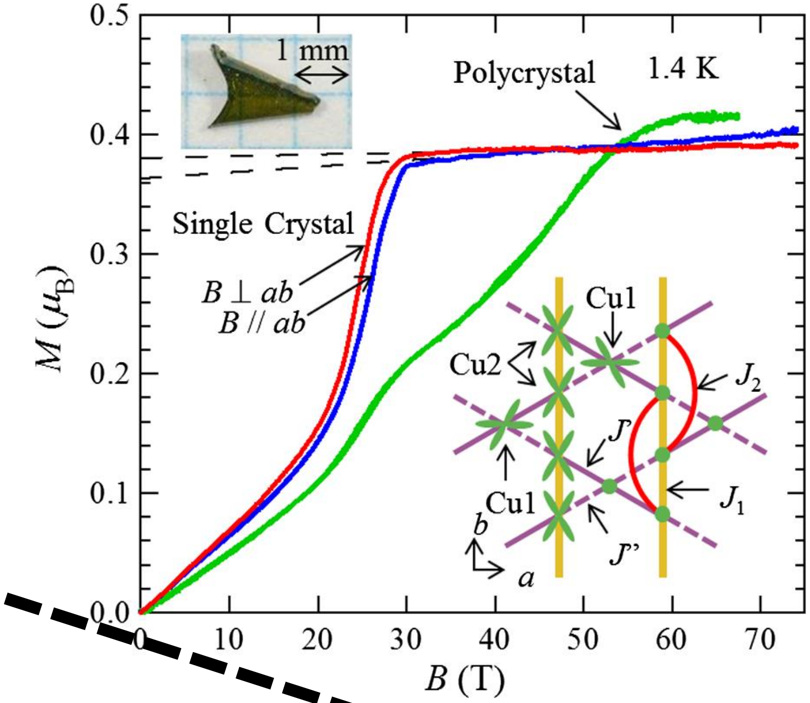


microscopic magnetic model



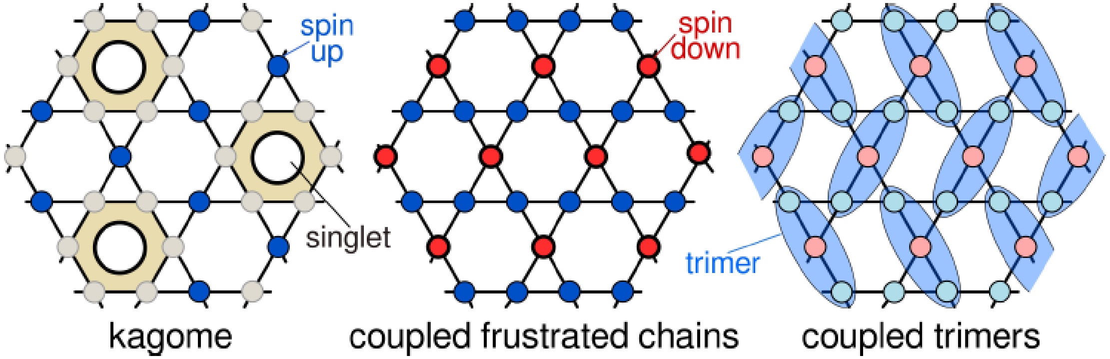
Magnetization process



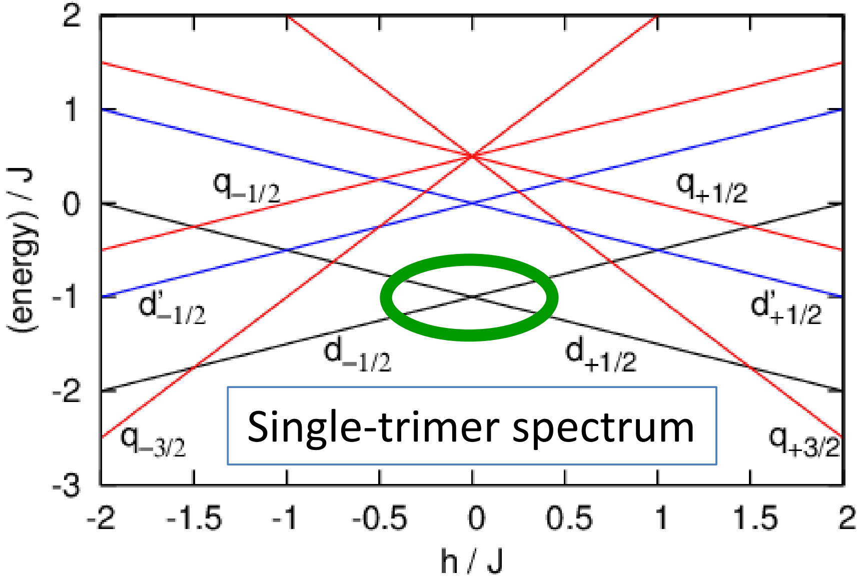


Product of polarized doublets

Wide 1/3-plateau between 22 T and 225 T, followed by a rapid tripling



Effective model for low fields $h = g\mu_B H/k_B \ll J$



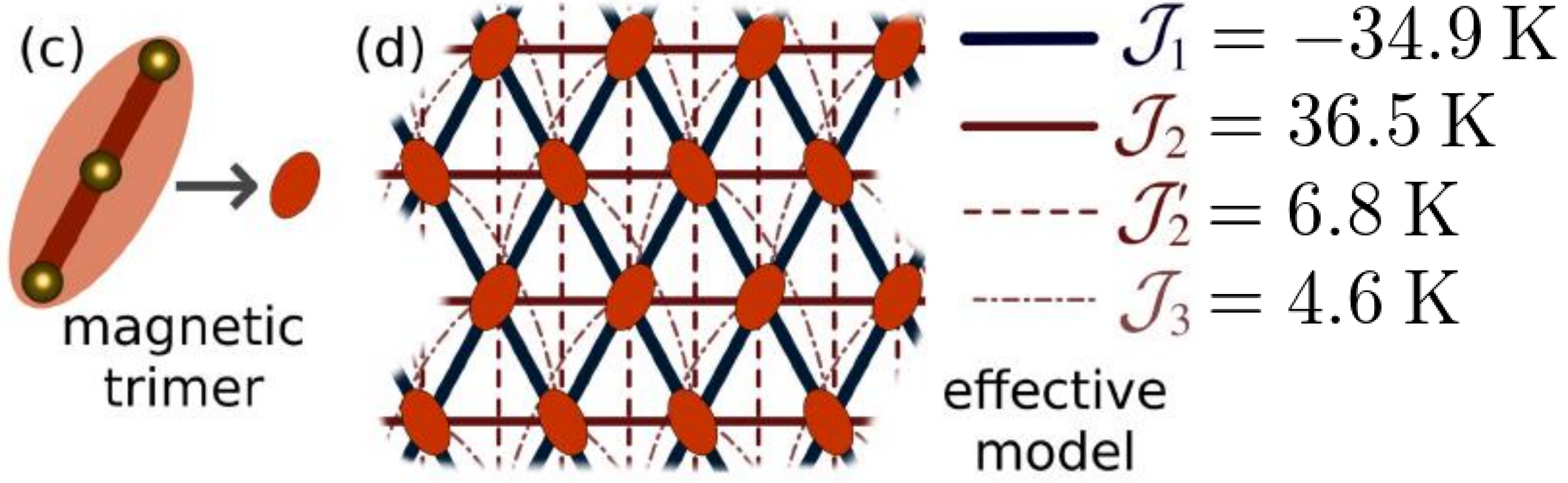
Pseudospin-1/2 moment living on each trimer

$$\boldsymbol{T}_{\boldsymbol{r}} = \left(|d_{+\frac{1}{2}}\rangle_{\boldsymbol{r}}, |d_{-\frac{1}{2}}\rangle_{\boldsymbol{r}}\right) \frac{\boldsymbol{\sigma}}{2} \begin{pmatrix} {}_{\boldsymbol{r}}\langle d_{+\frac{1}{2}}| \\ {}_{\boldsymbol{r}}\langle d_{-\frac{1}{2}}| \end{pmatrix}$$

$$|d_{+\frac{1}{2}}\rangle = \frac{1}{\sqrt{6}}\left(|\uparrow\uparrow\downarrow\rangle + |\downarrow\uparrow\uparrow\rangle - 2|\uparrow\downarrow\uparrow\rangle\right)$$

cf. distorted diamond chains
Tonegawa et al., 2000; Honecker & Laeuchli, 2001

2nd-order strong-coupling expansion



$$\mathcal{J}_1 = -34.9\ \mathrm{K}$$

$$\mathcal{J}_2 = 36.5\ \mathrm{K}$$

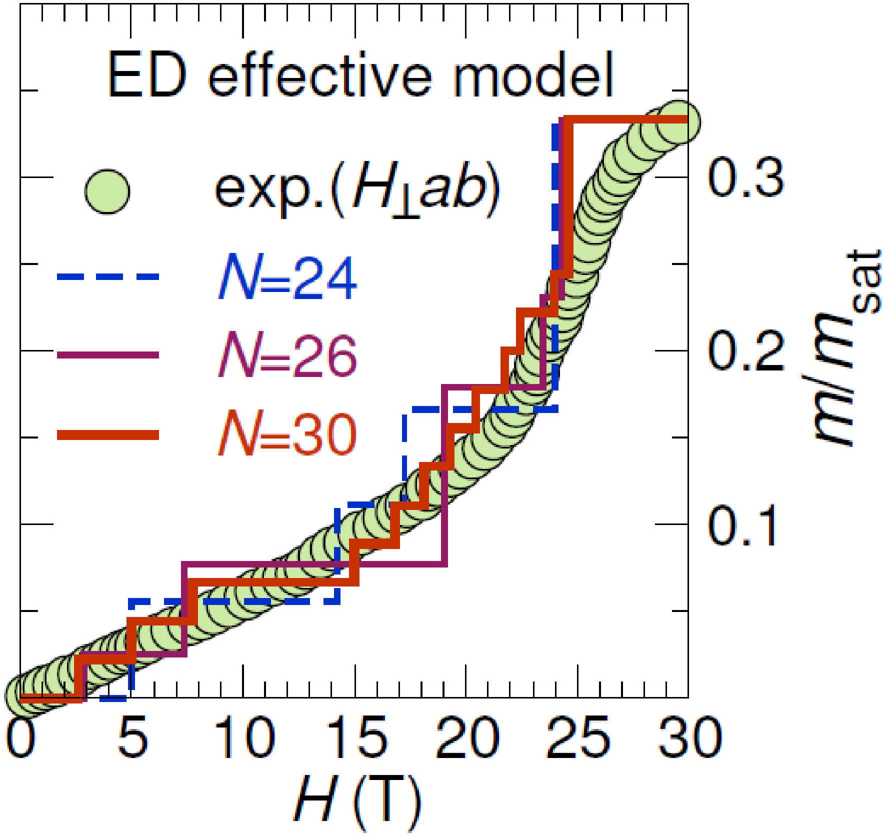
$$\mathcal{J}_2' = 6.8\ \mathrm{K}$$

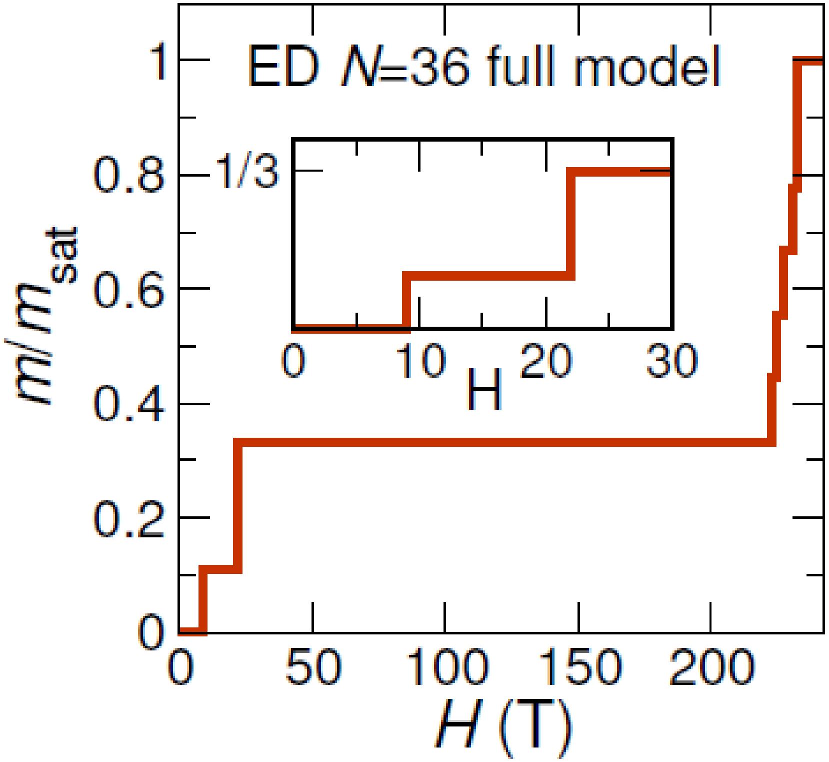
$$\mathcal{J}_3 = 4.6\ \mathrm{K}$$

Magnetization process for low fields

Exact diag. of the effective model: tripled size can be simulated.

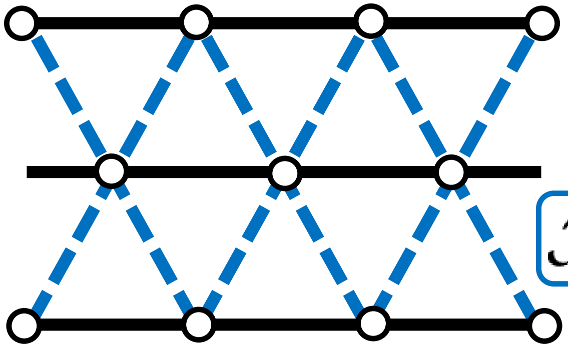
Saturation of pseudospins = 1/3 plateau of the original model





Reproduced the change of the slope!

Nature of field-induced phases I



$\mathcal{J}_1 = -34.9\,\mathrm{K}$

$\mathcal{J}_2 = 36.5\,\mathrm{K}$

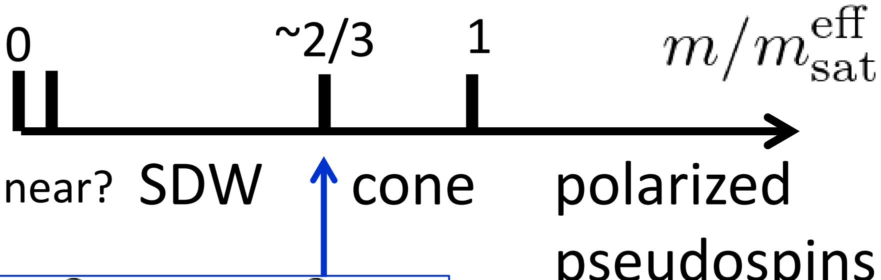
Two leading effective couplings

Spatially anisotropic triangular magnet

cf. Cs_2CuCl_4: $\mathcal{J}_1, \mathcal{J}_2 > 0$

Field theory for $|\mathcal{J}_1| \ll \mathcal{J}_2$

Starykh *et al.*, PRB **82**, 014421 (2010)



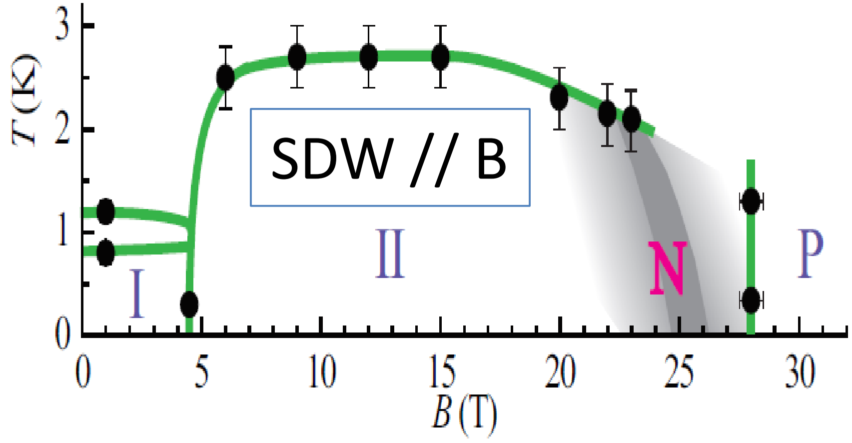
Collinear? SDW

cone

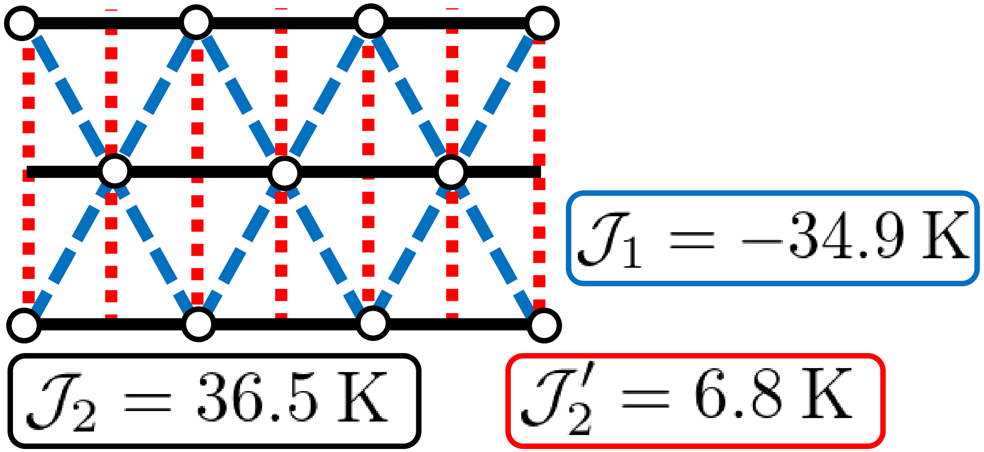
polarized pseudospins (=1/3 plateau)

$$m \simeq \frac{2}{3} m_{\mathrm{sat}}^{\mathrm{eff}} = \frac{2}{9} m_{\mathrm{sat}}$$

H. Ishikawa *et al.*, PRL **114**, 227202 (2015)
M. Yoshida *et al.*, arXiv:1602.04028



Nature of field-induced phases II



Three leading effective couplings

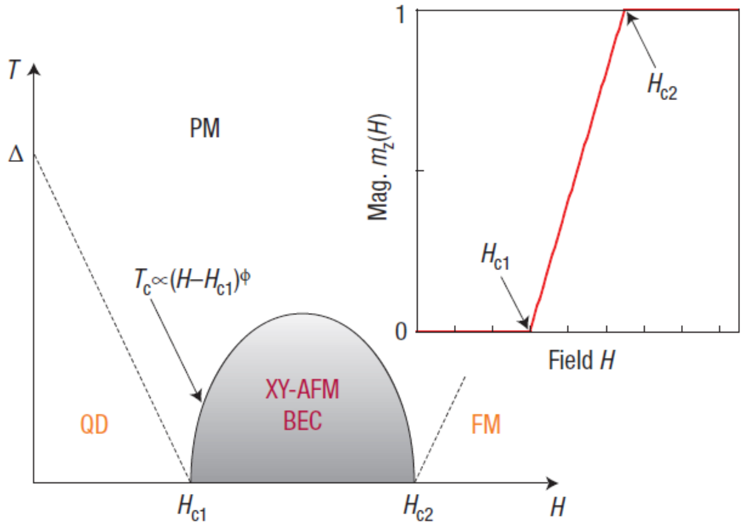
Anisotropic version of J_1-J_2 model on square lattice

Nematic order due to condensation
of two-magnon bound states

Shannon, Momoi, & Sindzingre, PRL **96**, 027213 (2006)

Mechanism for a bond nematic state

- cf. Bose-Einstein condensation of single magnons



$$\langle b_j \rangle = \langle S_j^+ \rangle \neq 0$$

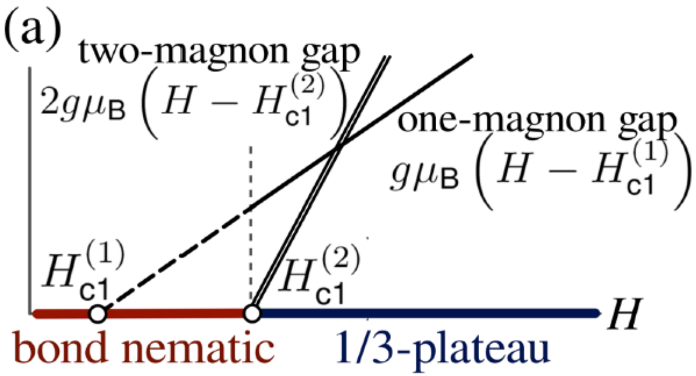
Transverse magnetic order

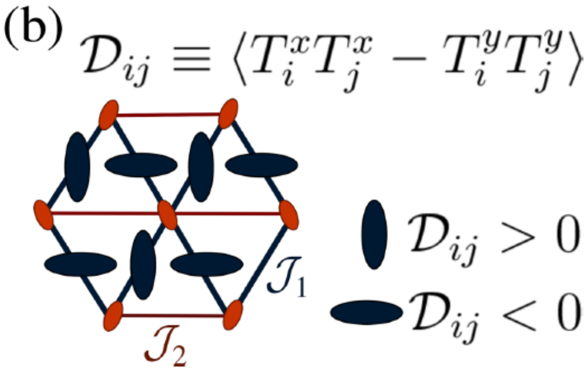
Review article: T. Giamarchi et al., Nat. Phys. 4, 198 (2008)
$TlCuCl_3$ (Exp. & Theory): Nikuni et al., PRL 84, 5868 (2000)

- Bimagnon condensation

Shannon, Momoi, & Sindzingre, PRL **96**, 027213 (2006)

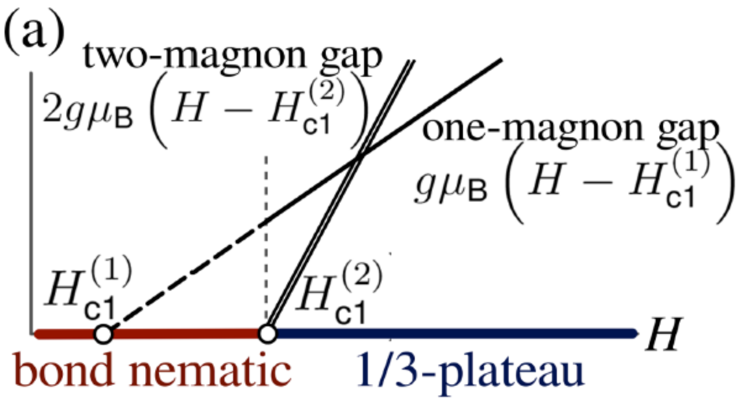
$$\langle b_j b_{j'} \rangle = \langle T_j^+ T_{j'}^+ \rangle \neq 0$$



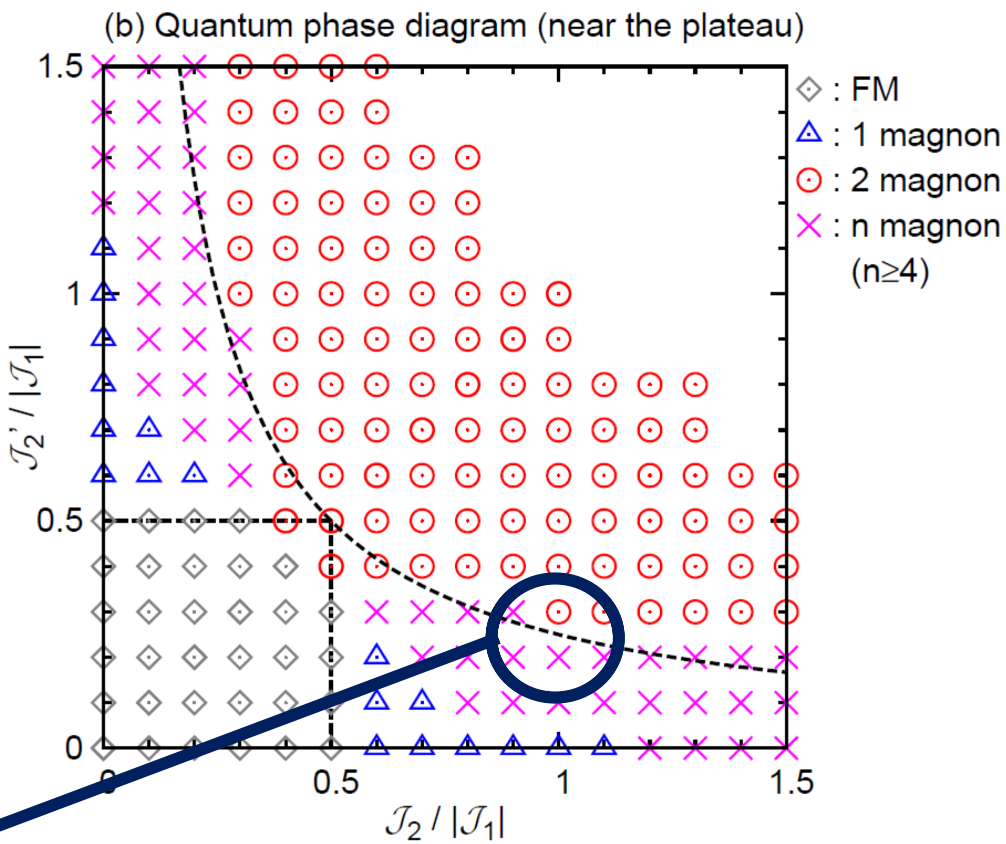


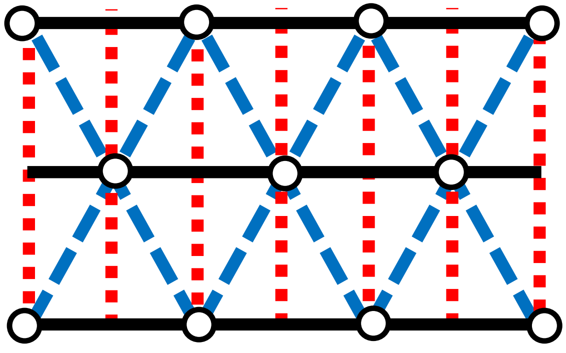
Bond nematic order

Leading magnon instability from the plateau



Exact diag. calculation of n-magnon states (n=1,2,3,4) In the $\mathcal{J}_1-\mathcal{J}_2-\mathcal{J}_2'$ model





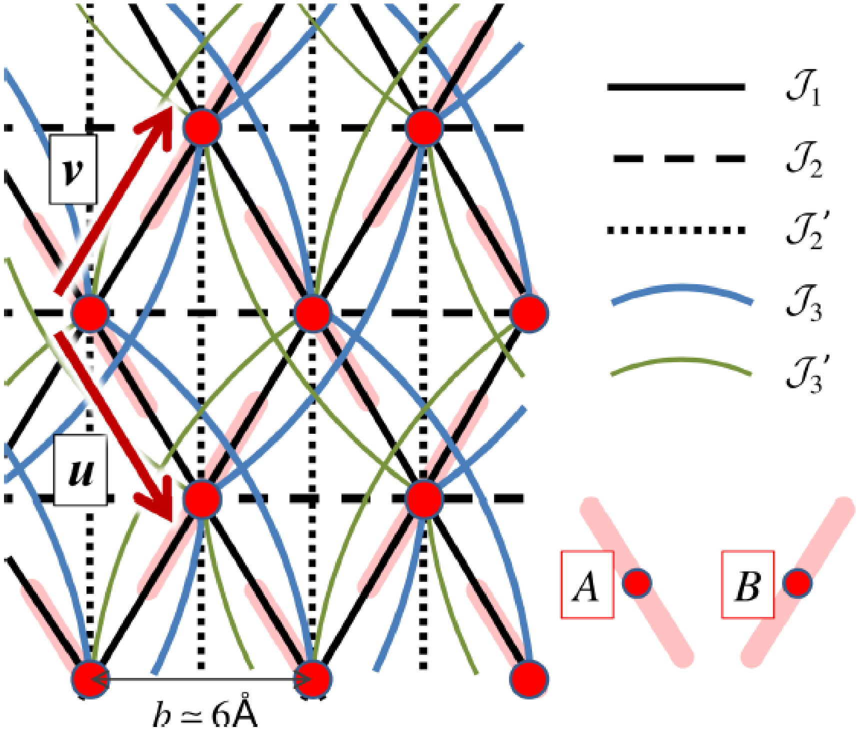
$\mathcal{J}_1 = -34.9\ \mathrm{K}$

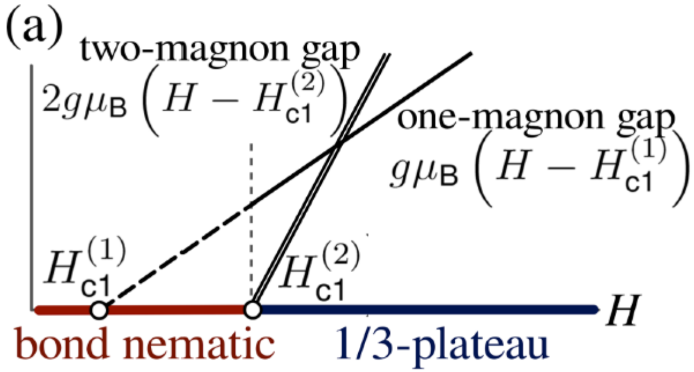
$\mathcal{J}_2 = 36.5\ \mathrm{K}$

$\mathcal{J}_2' = 6.8\ \mathrm{K}$

Nematic order
In an extended region
of the parameter space

Full effective model





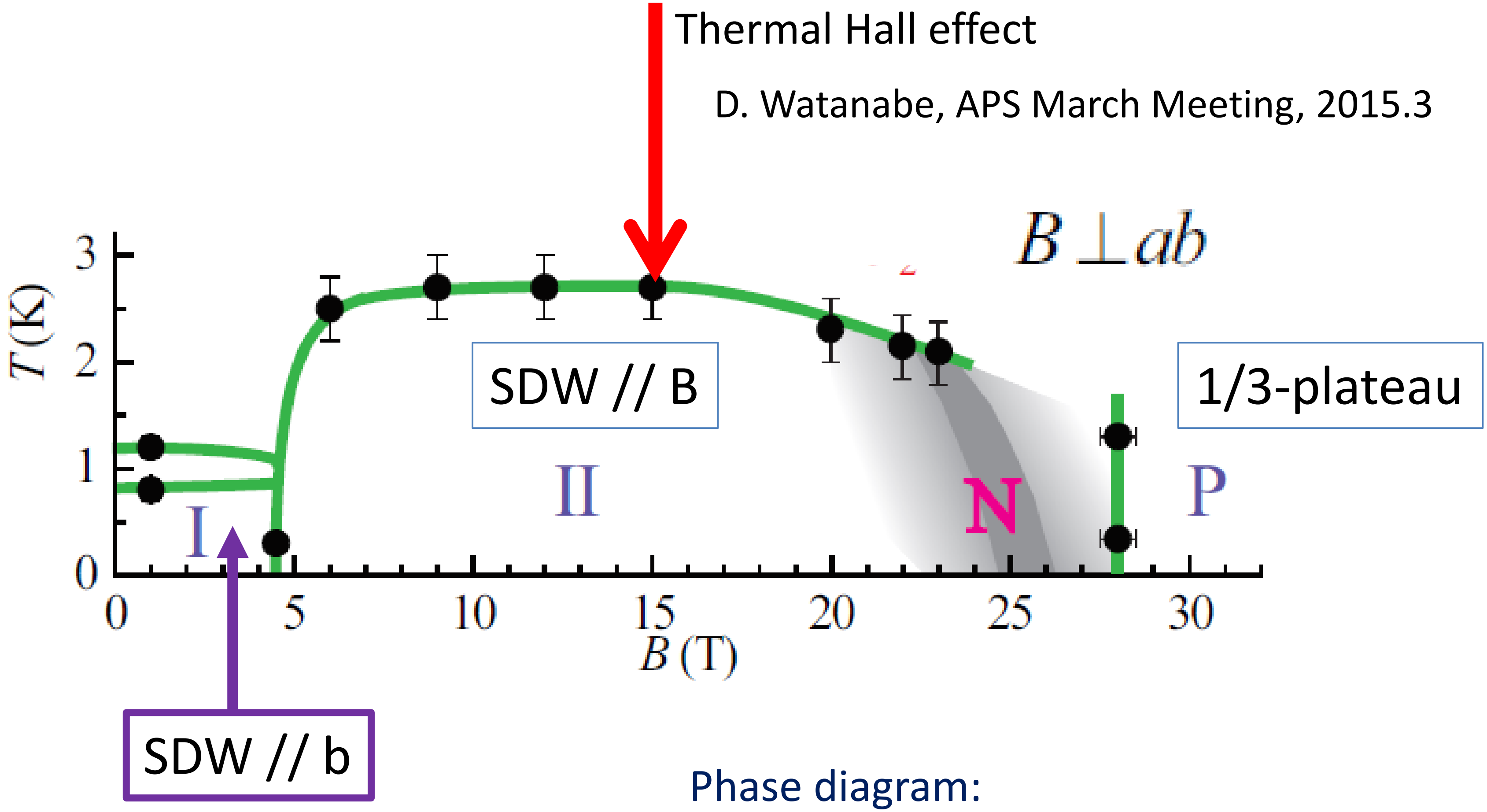
- Longer-range interactions such as $J_3 = 4.6\ \mathrm{K}, J_3' = 1.7\ \mathrm{K}$ tend to destabilize bimagnons, leading to a confentional single-magnon condensation.
- Slight tuning of the original model (e.g., increased J'=-0.25J) recovers bimagnons.

Consistent with experiment!

Yet, the fit with χ is disproved.

The best parameter set for describing the system is still under investigation.

Open issue: Dzyaloshinskii-Moriya interactions



Thermal Hall effect

D. Watanabe, APS March Meeting, 2015.3

SDW // B

1/3-plateau

SDW // b

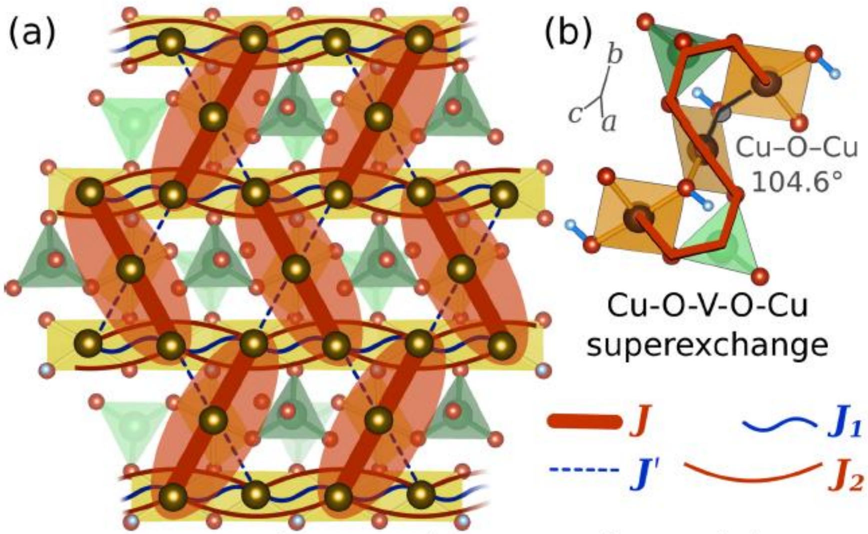
M. Yoshida, JPS Meeting, 2015.3

Phase diagram:
H. Ishikawa *et al.*, PRL **114**, 227202 (2015)
M. Yoshida *et al.*, arXiv:1602.04028

Summary

O. Janson *et al.*, arXiv:1509.07333
(to appear in Phys. Rev. Lett.)

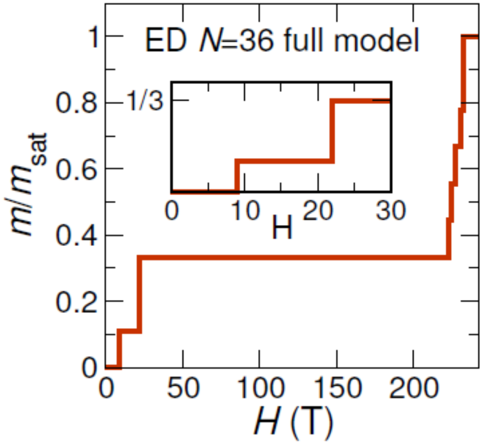
Coupled-trimer model

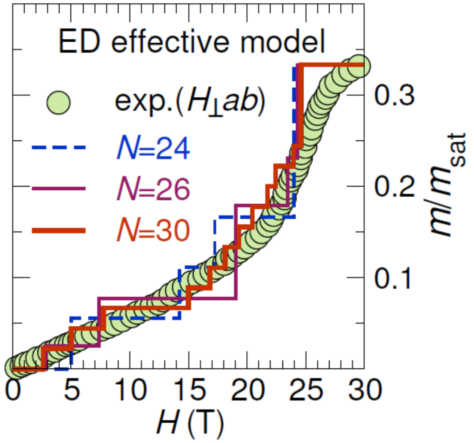


microscopic magnetic model

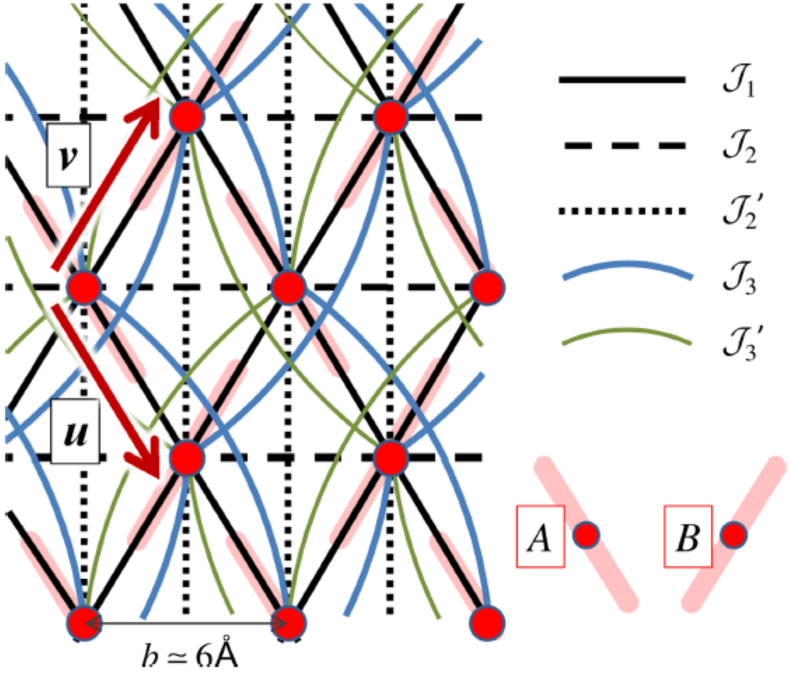
$$J \simeq 252\,\mathrm{K}$$

$$J : J' : J_1 : J_2 = 1 : (-0.2) : (-0.5) : 0.2$$

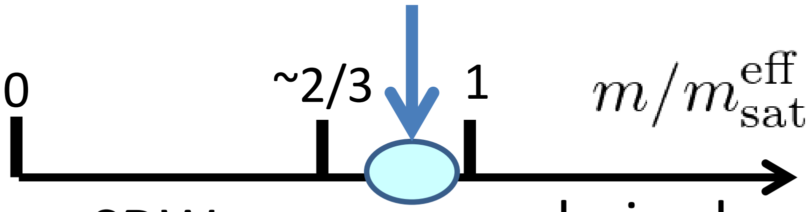




Effective model



Nematic

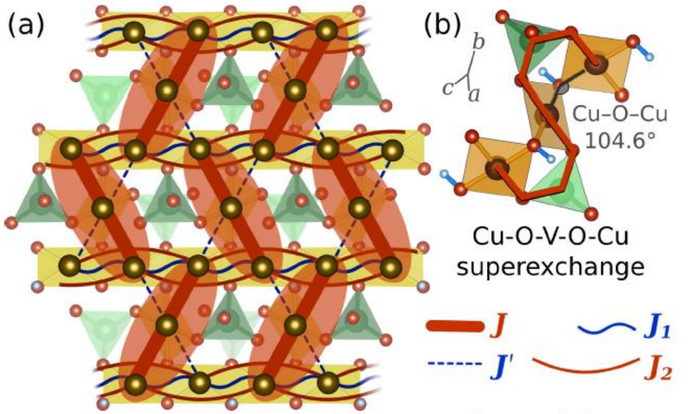


SDW

cone

polarized
pseudospins
=1/3 plateau

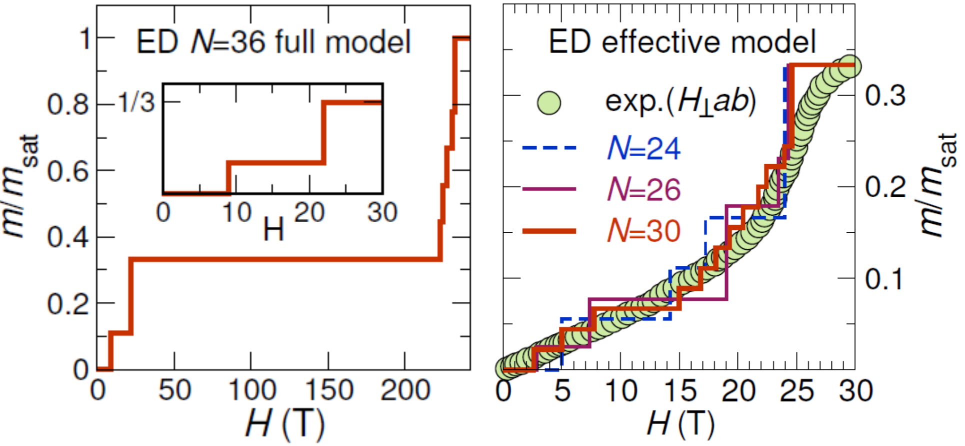
Coupled trimers vs. coupled frustrated chains



microscopic magnetic model

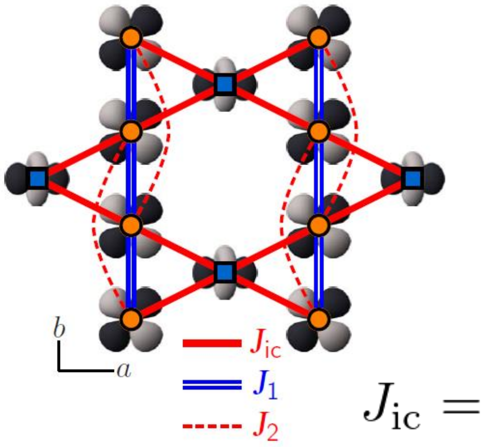
$$J \simeq 252\ \mathrm{K}$$

$$J : J' : J_1 : J_2 = 1 : (-0.2) : (-0.5) : 0.2$$

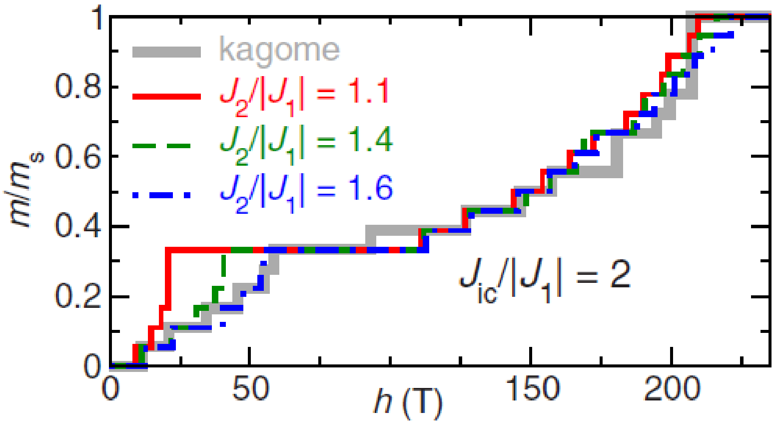


Agreement with mag. susceptibility $T \gtrsim 30\ \mathrm{K}$

Somewhere inbetween?



$$J_{\mathrm{ic}} = 100\ \mathrm{K}$$



Faraday rotation measurement agrees better with coupled frustrated chain model. (Takeyama group @ ISSP, March, 2016)

Agreement with mag. susceptibility $T \gtrsim 50\ \mathrm{K}$

Field-induced phenomena in a single crystal

H. Ishikawa *et al.*, PRL **114**, 227202 (2015)
M. Yoshida *et al.*, arXiv:1602.04028

- Wide 1/3 plateau

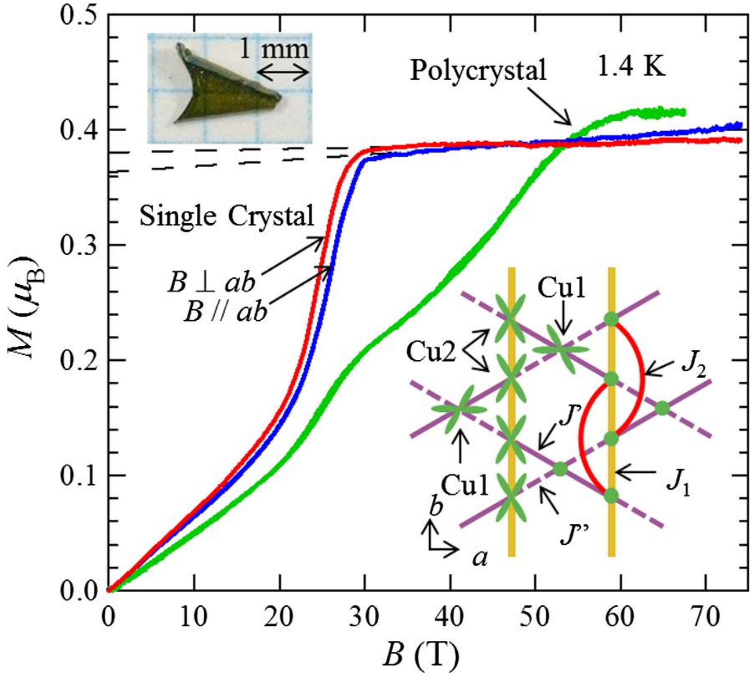
26 T ➔ 74 T ➔ Over 100 T ?!

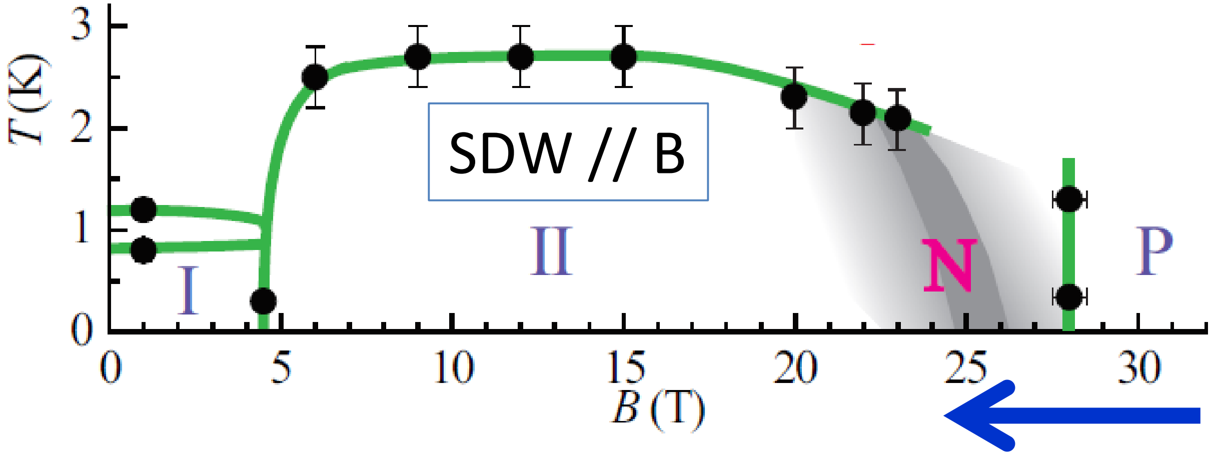
Faraday rotation exp.,
T. Yamashita *et al.*, JPS Meeting, 2015.3

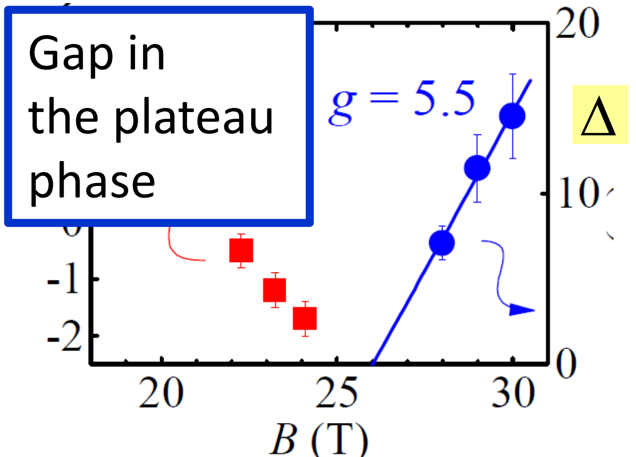
Much larger than the kagome AFM case!!

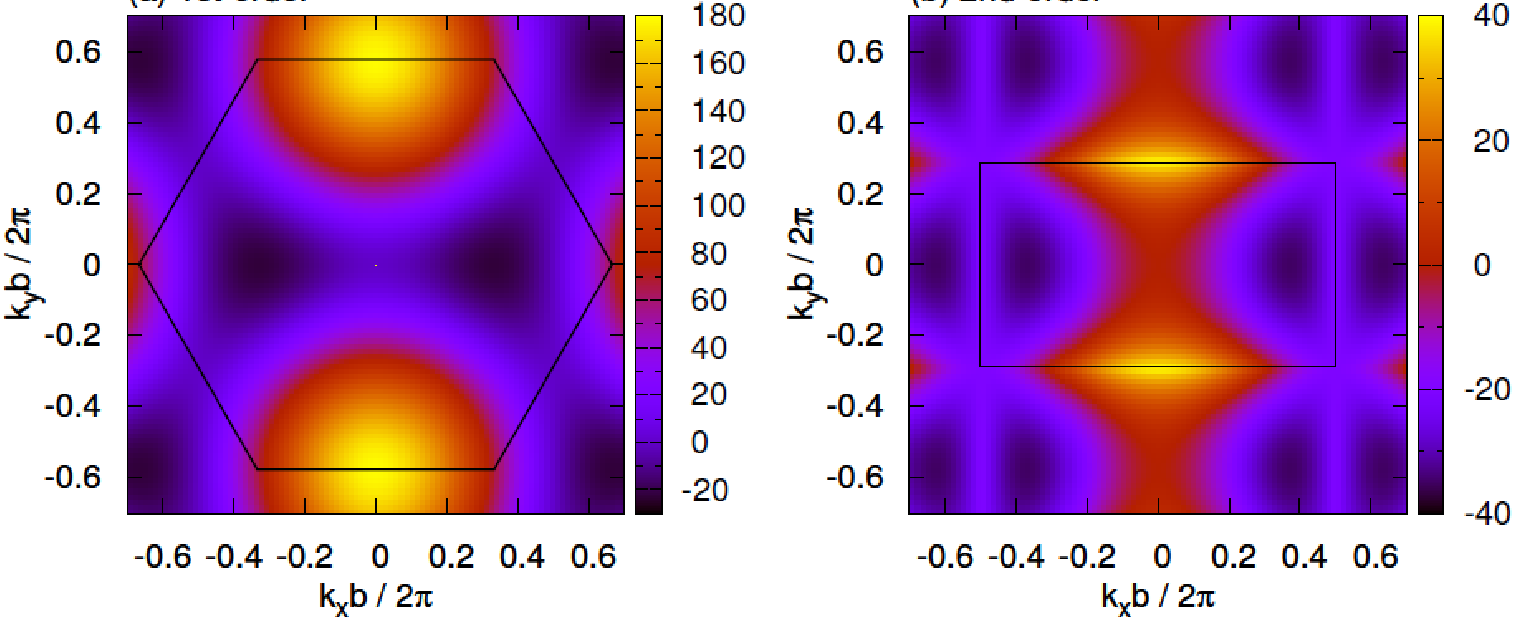
- "N" phase between SDW and plateau:

Originates from condensation of multimagnon bound states?



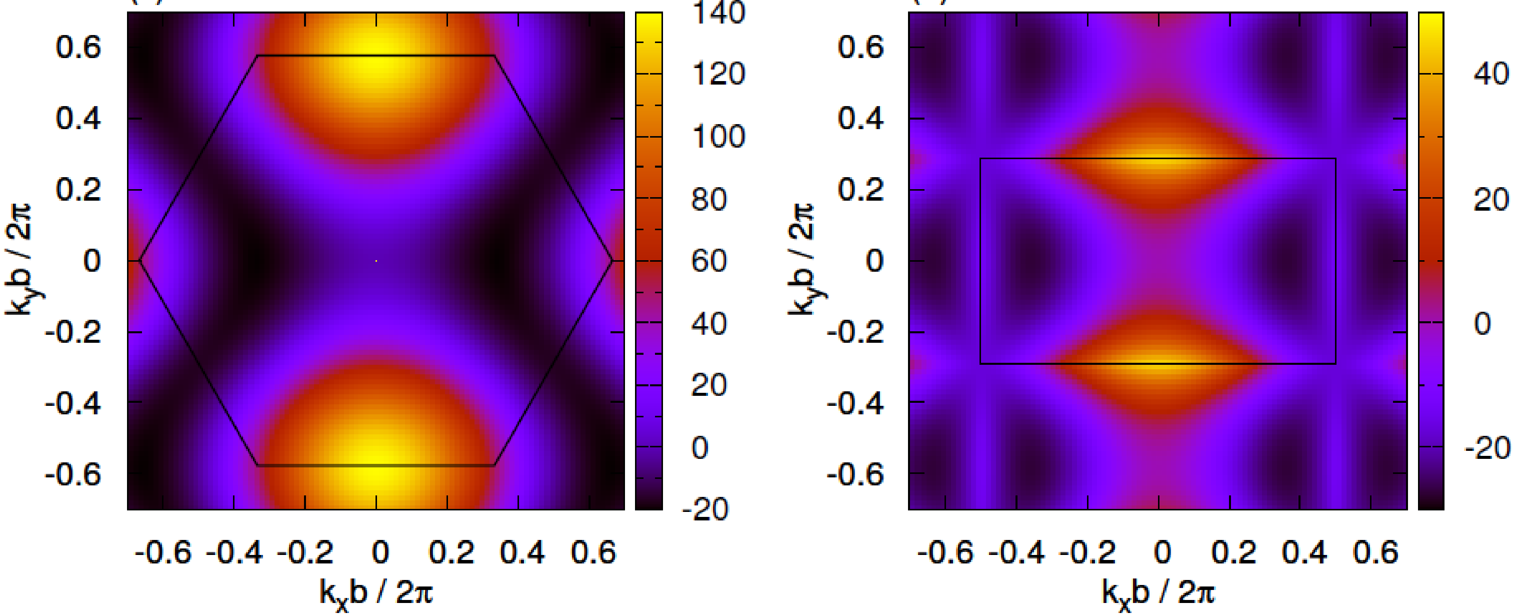



Gap in the plateau phase



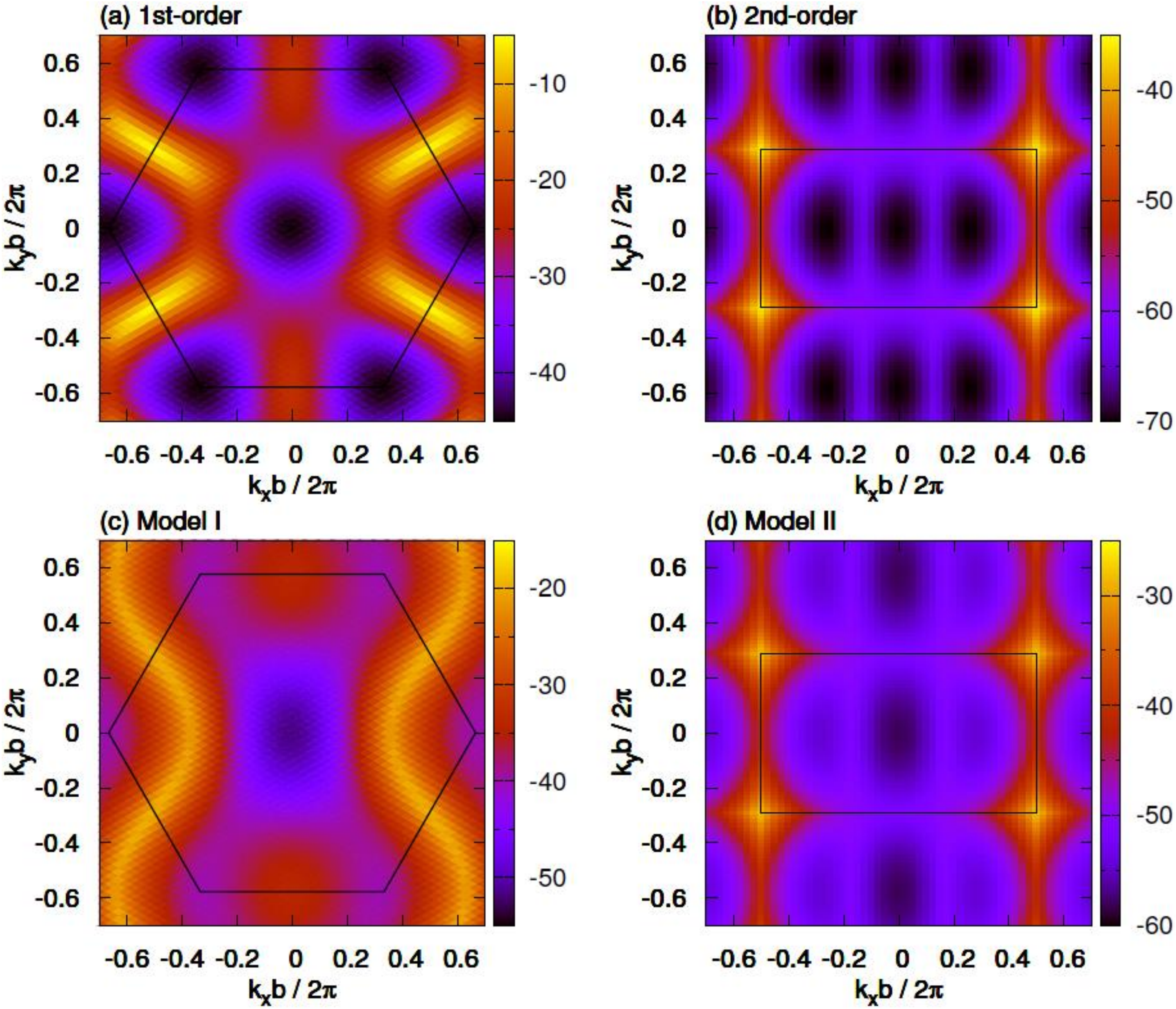
(a) 1st-order

(b) 2nd-order



(c) Model I

(d) Model II


(a) 1st-order

(b) 2nd-order

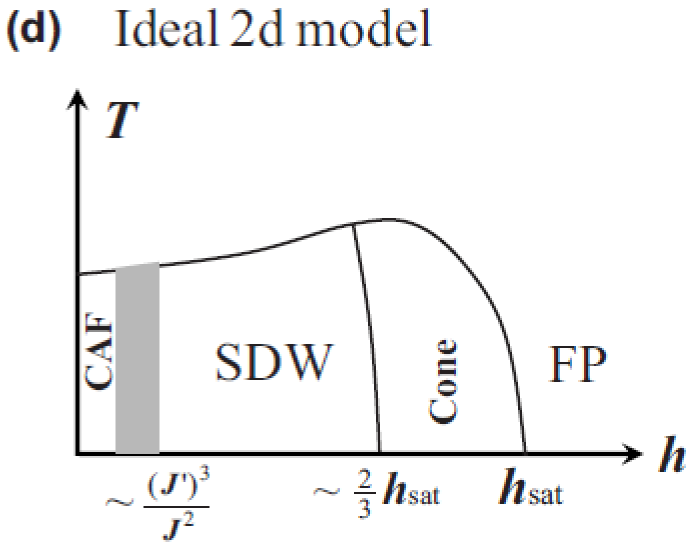
(c) Model I

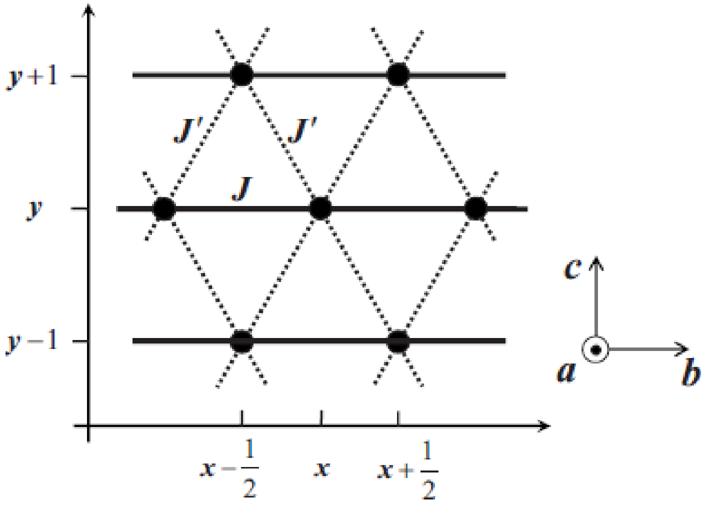
(d) Model II

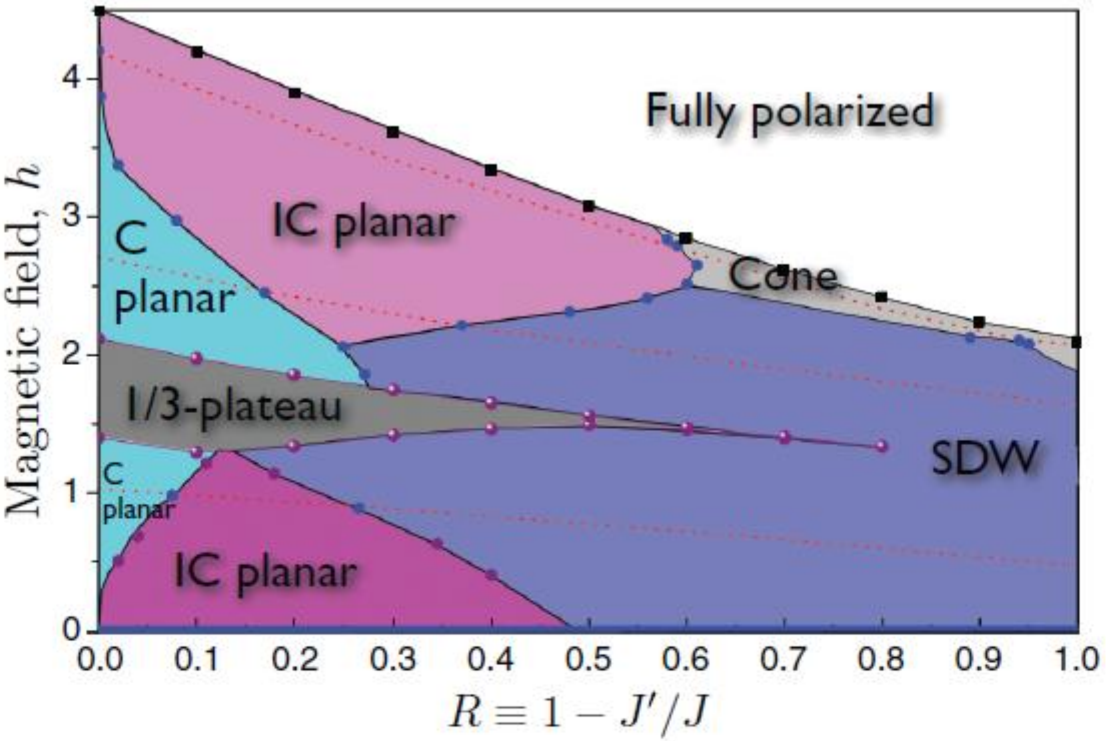
Spatially anisotropic triangular antiferromagnet (case of J2, J1>0, J2'=0)

Starykh, Katsura, and Balents,
PRB 82, 014421 (2010)
Bosonization analysis for J'/J<<1

Chen, Ju, Jiang, Starykh, and Balents,
PRB 87, 165123 (2013)
DMRG calculation

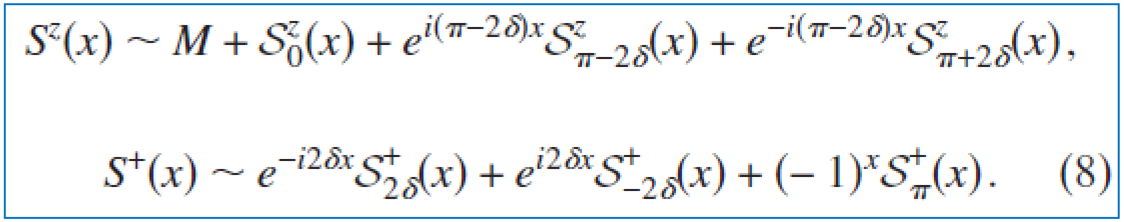






Bosonization analysis

Starykh, Katsura, and Balents,
PRB 82, 014421 (2010)



$$S^z(x) \sim M + \mathcal{S}^z_0(x) + e^{i(\pi-2\delta)x}\mathcal{S}^z_{\pi-2\delta}(x) + e^{-i(\pi-2\delta)x}\mathcal{S}^z_{\pi+2\delta}(x),$$

$$S^+(x) \sim e^{-i2\delta x}\mathcal{S}^+_{2\delta}(x) + e^{i2\delta x}\mathcal{S}^+_{-2\delta}(x) + (-1)^x\mathcal{S}^+_\pi(x). \quad (8)$$

$$\begin{aligned} H_1 \approx J'\sum_{y,z}\int dx\Big\{ & 2M^2 + 2\mathcal{S}^z_{y,z;0}\mathcal{S}^z_{y+1,z;0} \\ & + 2\sin\delta[\mathcal{S}^z_{y,z;\pi-2\delta}\mathcal{S}^z_{y+1,z;\pi+2\delta} + \text{H.c.}] \\ & + \frac{1}{2}[-i\mathcal{S}^+_{y,z;\pi}\partial_x\mathcal{S}^-_{y+1,z;\pi} + \text{H.c.}] \\ & + \cos\delta[\mathcal{S}^+_{y,z;2\delta}\mathcal{S}^-_{y+1,z;2\delta} + \mathcal{S}^+_{y,z;-2\delta}\mathcal{S}^-_{y+1,z;-2\delta} + \text{H.c.}]\Big\} \end{aligned}$$

scaling dimensions

$\boxed{2}$ marginal

$\boxed{1/2\pi R^2 = 2K : 1 \to 2}$ SDW at low and middle h

$\boxed{1 + 2\pi R^2 = 1 + 1/2K : 2 \to 3/2}$ cone at high h

Consider only the 2nd and 3rd terms

$$H_1' = \sum_{y,z}\int dx\{\tilde{\gamma}_{\mathrm{SDW}}\cos[2\pi(\phi_{y,z} - \phi_{y+1,z})/\beta] - \tilde{\gamma}_{\mathrm{cone}}(\partial_x\theta_{y,z} + \partial_x\theta_{y+1,z})\cos[\beta(\theta_{y,z} - \theta_{y+1,z})]\}, \quad (27)$$

$$\tilde{\gamma}_{\mathrm{SDW}} = J'A_1^2\sin\delta \text{ and } \tilde{\gamma}_{\mathrm{cone}} = J'A_3^2\beta/2.$$

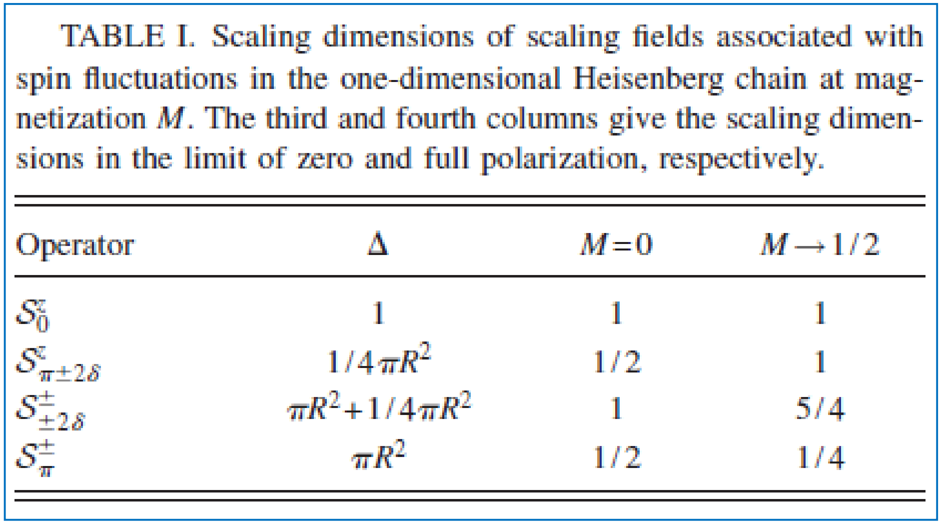
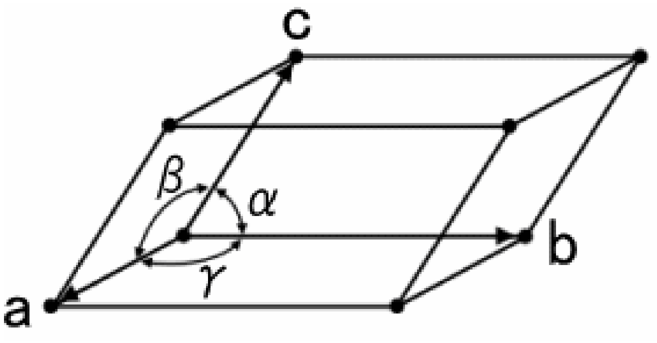
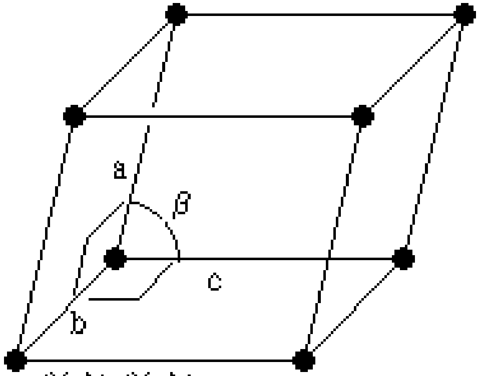


TABLE I. Scaling dimensions of scaling fields associated with spin fluctuations in the one-dimensional Heisenberg chain at magnetization M. The third and fourth columns give the scaling dimensions in the limit of zero and full polarization, respectively.

Operator	Δ	$M=0$	$M\to 1/2$
$\mathcal{S}^z_0$	1	1	1
$\mathcal{S}^z_{\pi\pm 2\delta}$	$1/4\pi R^2$	$1/2$	1
$\mathcal{S}^\pm_{\pm 2\delta}$	$\pi R^2 + 1/4\pi R^2$	1	$5/4$
$\mathcal{S}^\pm_\pi$	πR^2	$1/2$	$1/4$

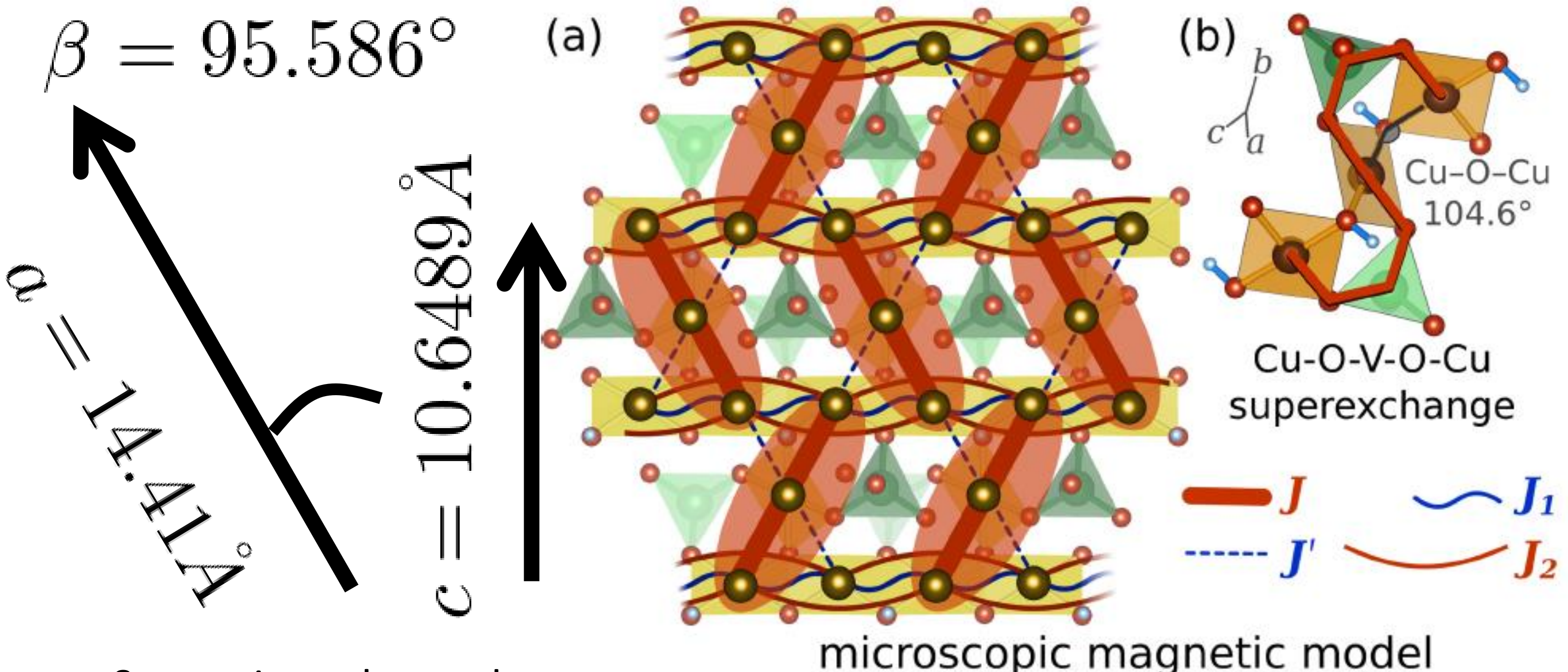
monoclinic : $a \neq b \neq c,\ \alpha = \gamma = 90^\circ \neq \beta$





単純単斜

© K.Kakegawa, Chiba Univesity



$\beta = 95.586^\circ$

$a = 14.41\mathring{A}$

$c = 10.6489\mathring{A}$

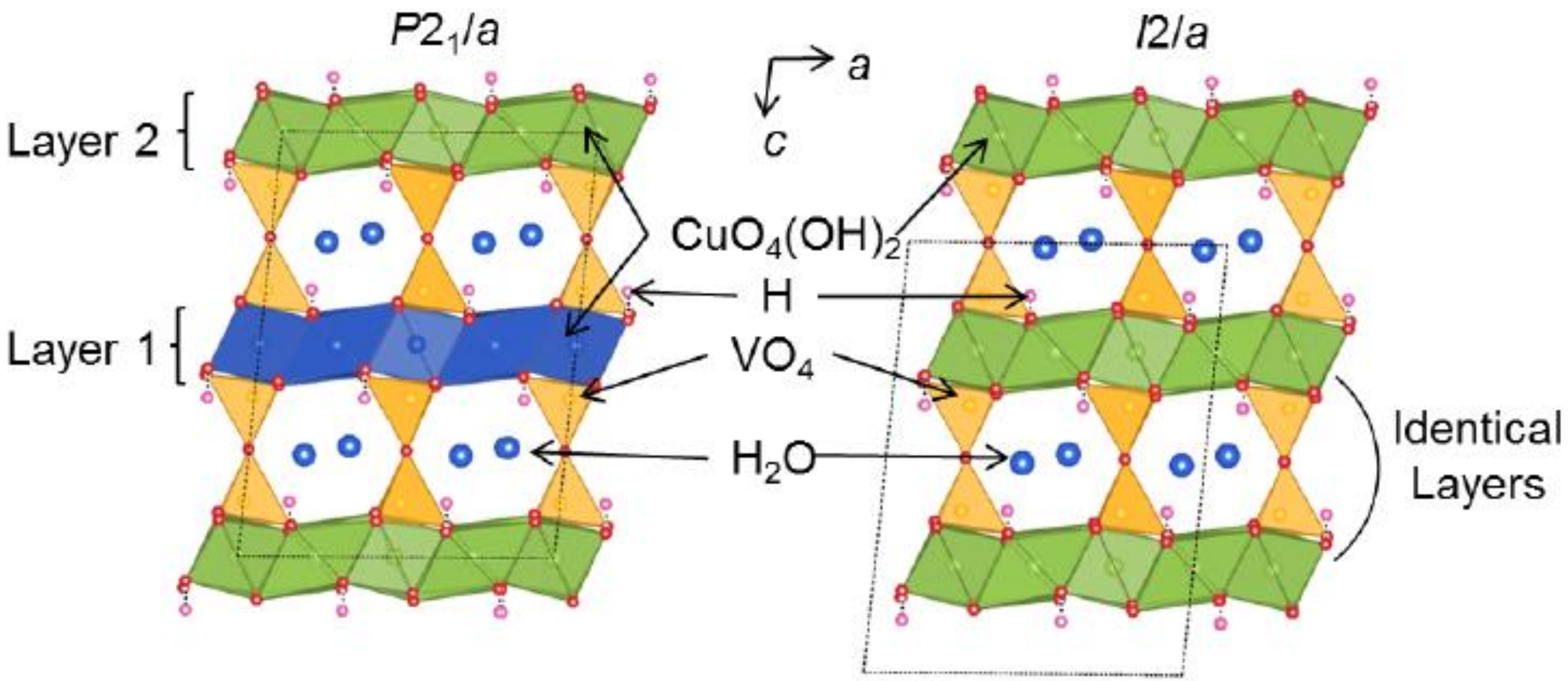
microscopic magnetic model

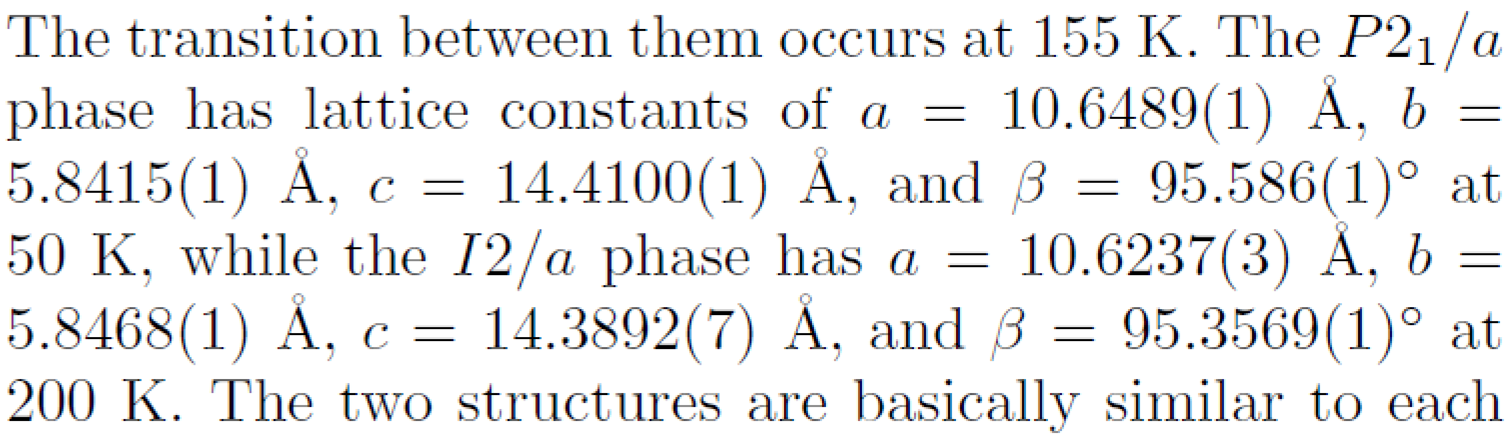
$b = 5.8415\mathring{A}$

Note: a & c are interchanged
in comparison with Ishikawa et al.

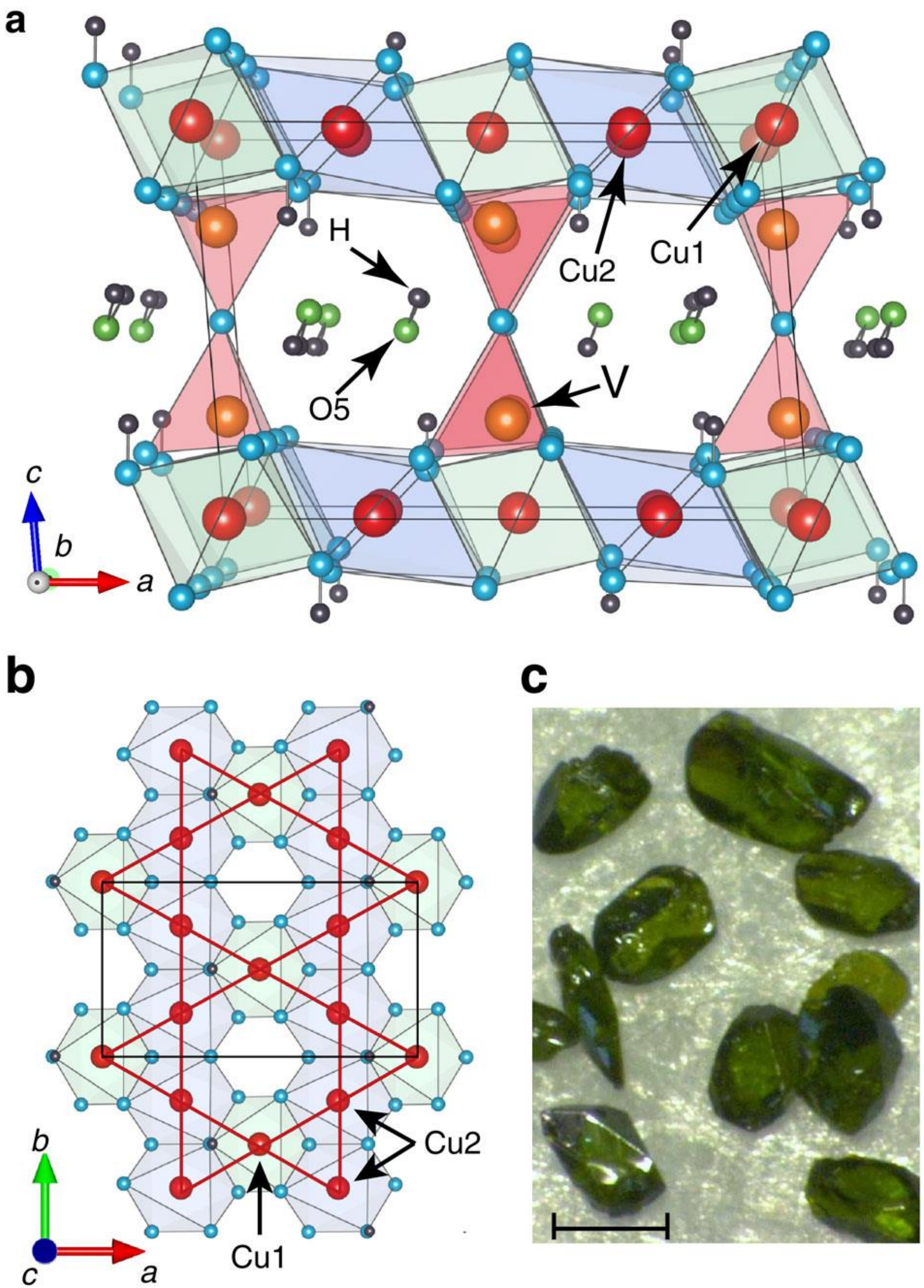
T=50 K

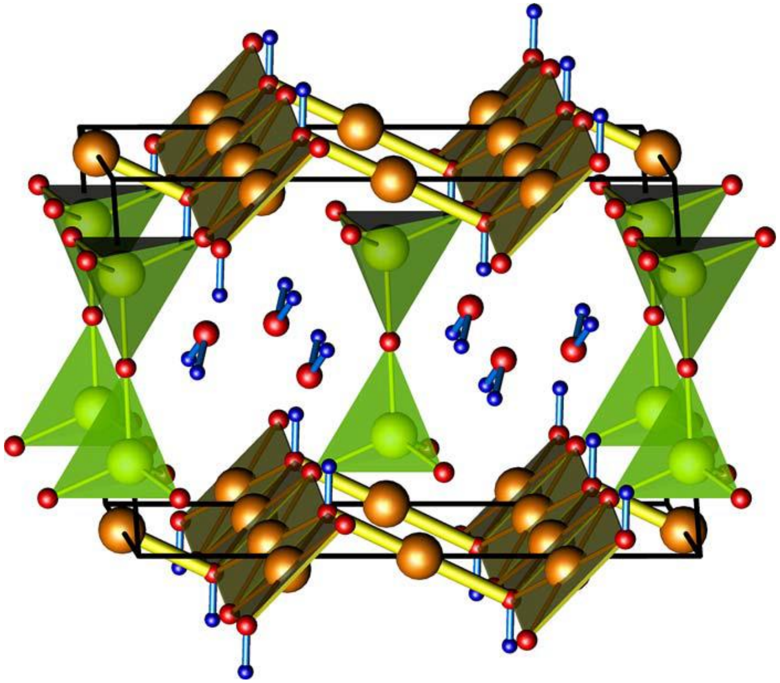
T=200 K



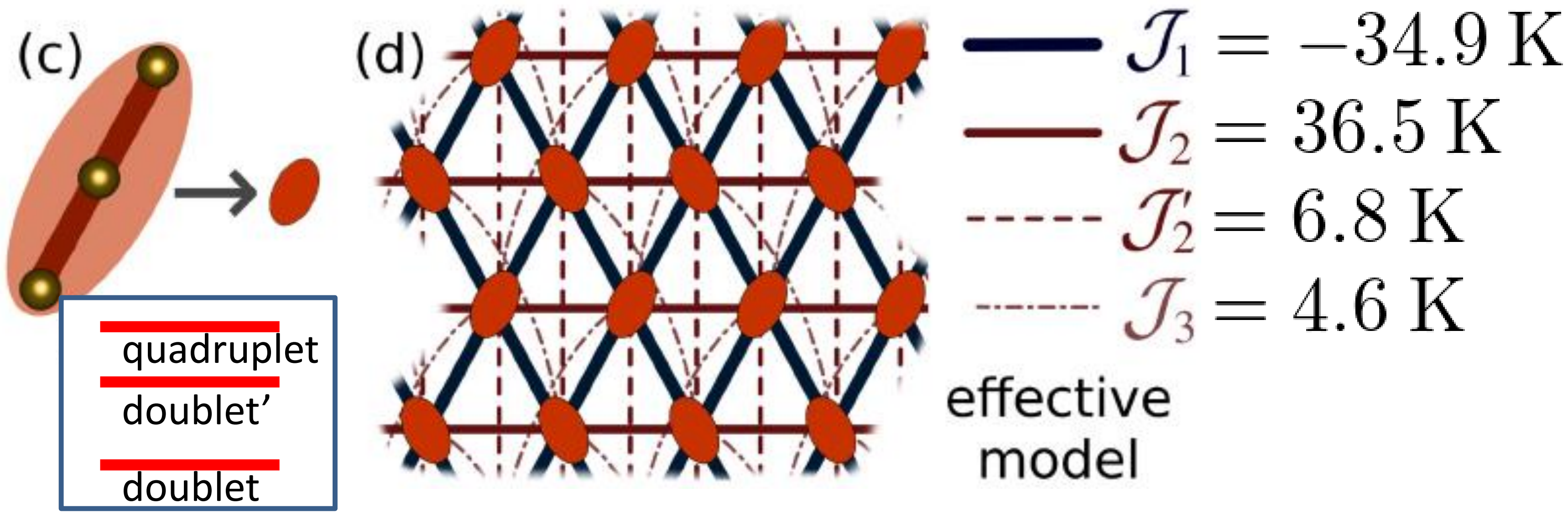


The transition between them occurs at 155 K. The $P2_1/a$ phase has lattice constants of a = 10.6489(1) Å, b = 5.8415(1) Å, c = 14.4100(1) Å, and β = 95.586(1)° at 50 K, while the $I2/a$ phase has a = 10.6237(3) Å, b = 5.8468(1) Å, c = 14.3892(7) Å, and β = 95.3569(1)° at 200 K. The two structures are basically similar to each


a
H
Cu2
Cu1
O5
V
c
b
a
b
Cu2
Cu1
b
c
a
c



Effective model for low fields $h = g\mu_B H/k_B \ll J$

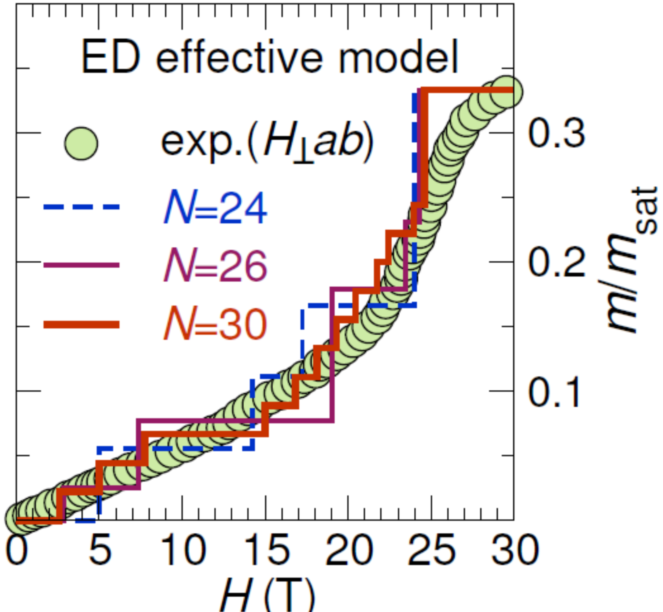


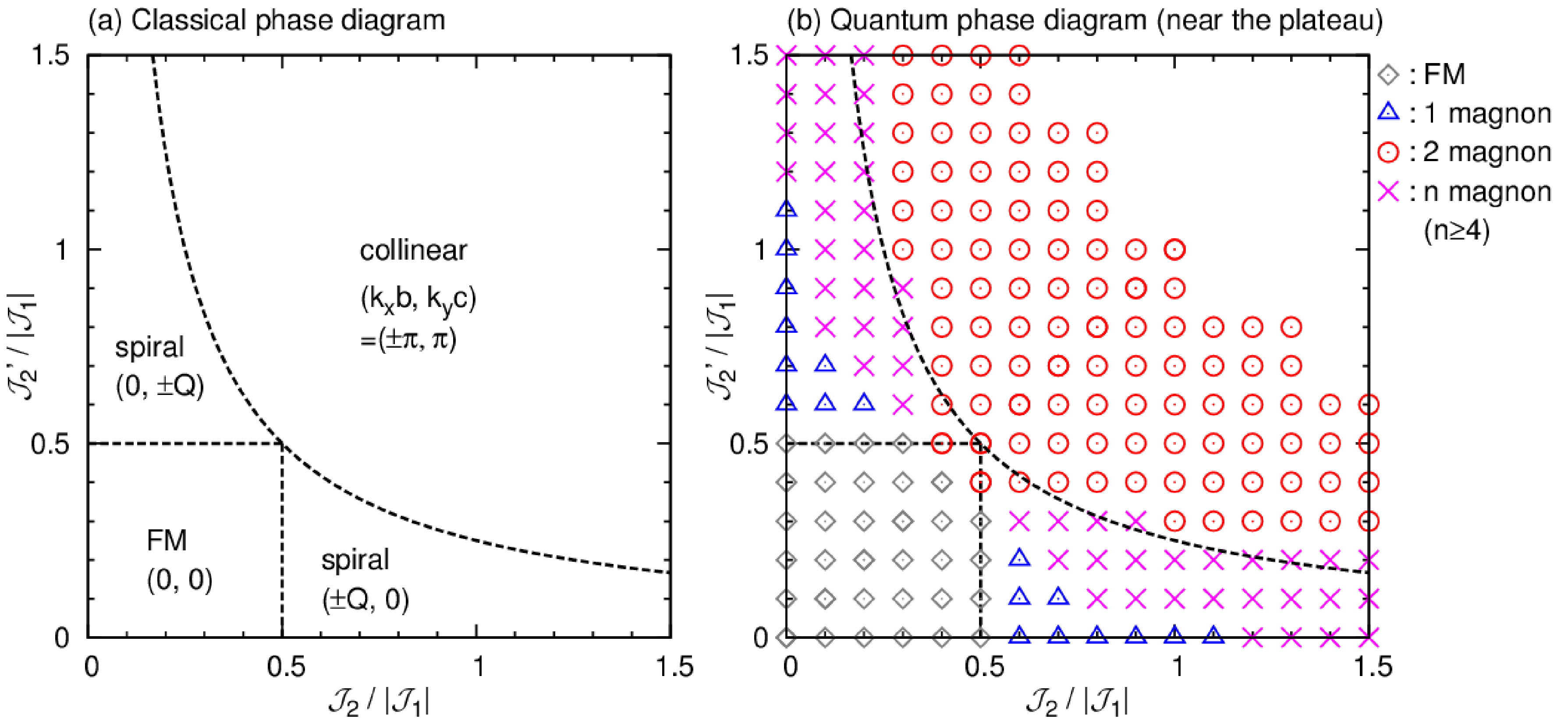
- Take the lowest-energy doublet in each trimer, perform 2nd-order strong-coupling expansion

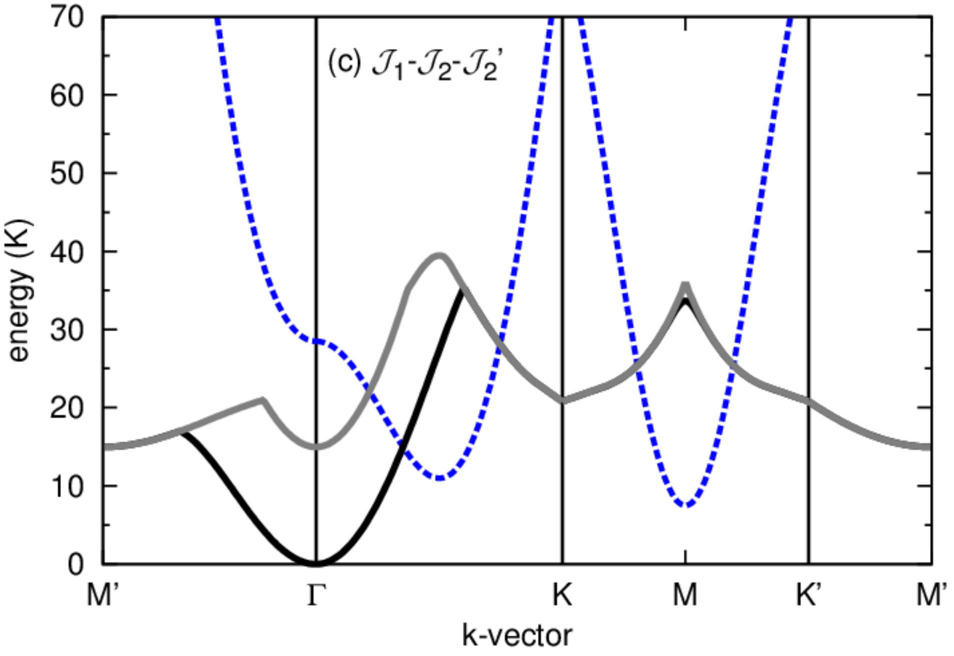
  cf. distorted diamond chains
  Tonegawa et al., 2000; Honecker & Laeuchli, 2001

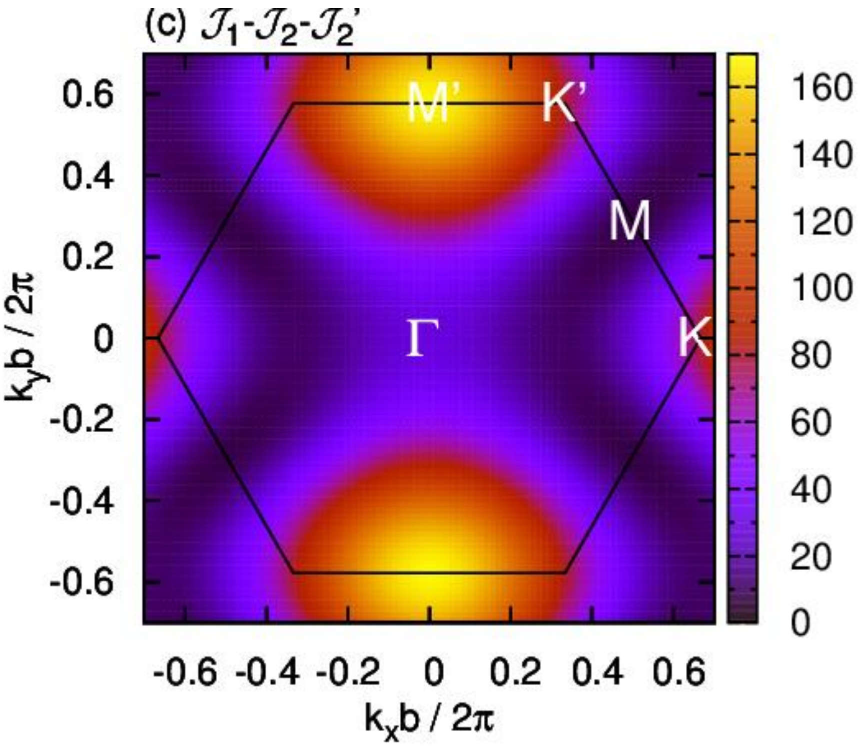
- Saturation of pseudospins = 1/3 plateau of the original model
- Exact diag. of the effective model: tripled size can be simulated.

  Reproduced the change of the slope!




(a) Classical phase diagram
(b) Quantum phase diagram (near the plateau)
collinear
$(k_x b, k_y c) = (\pm\pi, \pi)$
spiral
$(0, \pm Q)$
FM
$(0, 0)$
spiral
$(\pm Q, 0)$
◇ : FM
△ : 1 magnon
○ : 2 magnon
× : n magnon ($n \geq 4$)
$\mathcal{J}_2' / |\mathcal{J}_1|$
$\mathcal{J}_2 / |\mathcal{J}_1|$
0
0.5
1
1.5


(c) $\mathcal{J}_1$-$\mathcal{J}_2$-$\mathcal{J}_2'$
energy (K)
70
60
50
40
30
20
10
0
M'
Γ
K
M
K'
M'
k-vector


(c) $\mathcal{J}_1$-$\mathcal{J}_2$-$\mathcal{J}_2'$
M'
K'
M
Γ
K
$k_y b / 2\pi$
0.6
0.4
0.2
0
-0.2
-0.4
-0.6
-0.6 -0.4 -0.2 0 0.2 0.4 0.6
$k_x b / 2\pi$
160
140
120
100
80
60
40
20
0