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Field-Induced Ordering in Dipolar Spin Ice

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Department of Physics, National Taiwan University
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Lin and YJK, Phys. Rev. B 88, 220402(R) (2013)

Kao, Holdsworth, and YJK, Phys. Rev. B 93, 180410(R) (2016)

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Acknowledgement

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Wen-Han Kao
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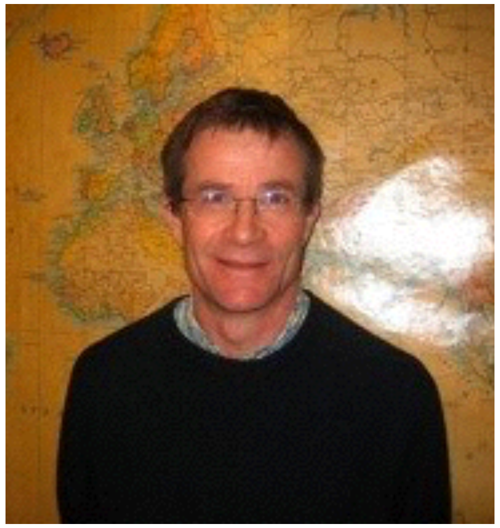
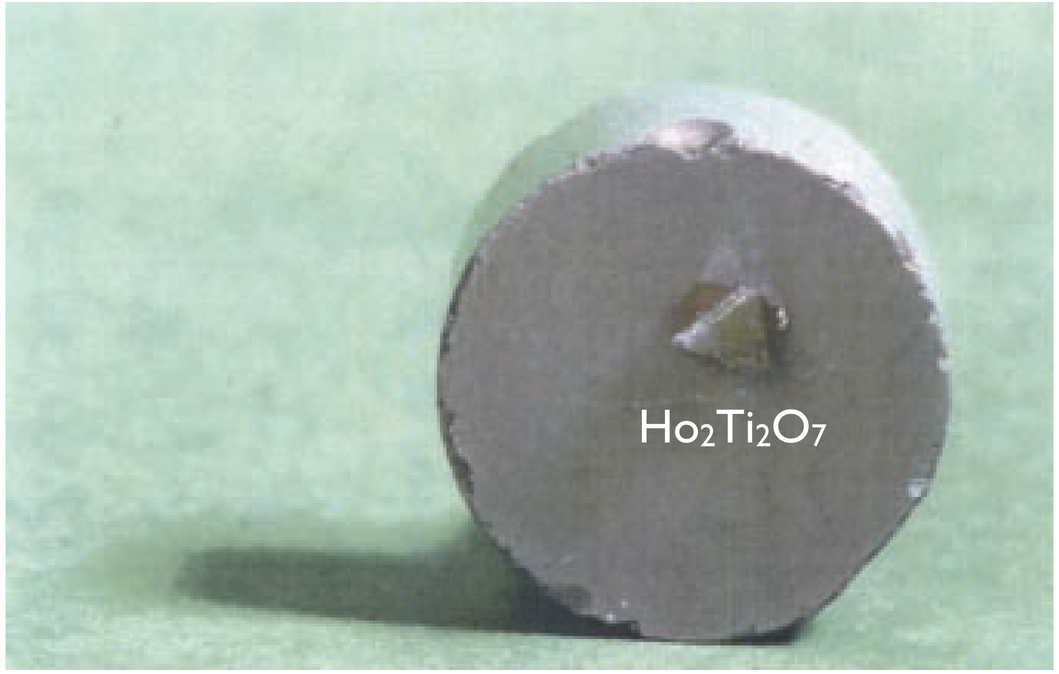


Peter Holdsworth
ENS, Lyon

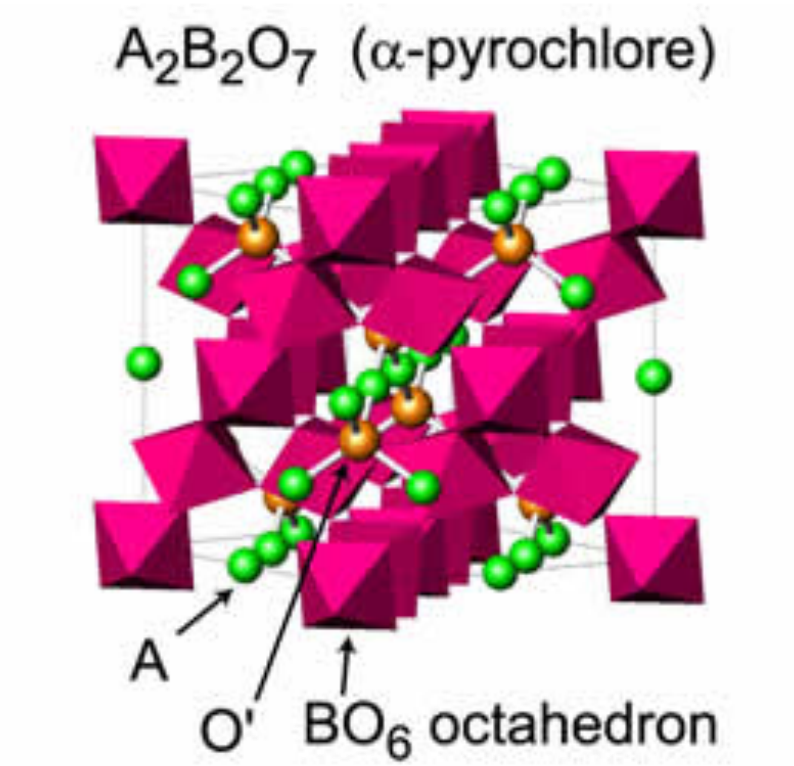
Outline

- Introduction
- Dipolar Spin ice in a [100] field
- Dipolar Spin ice in a tilted [111] field
- Conclusions

Spin Ice

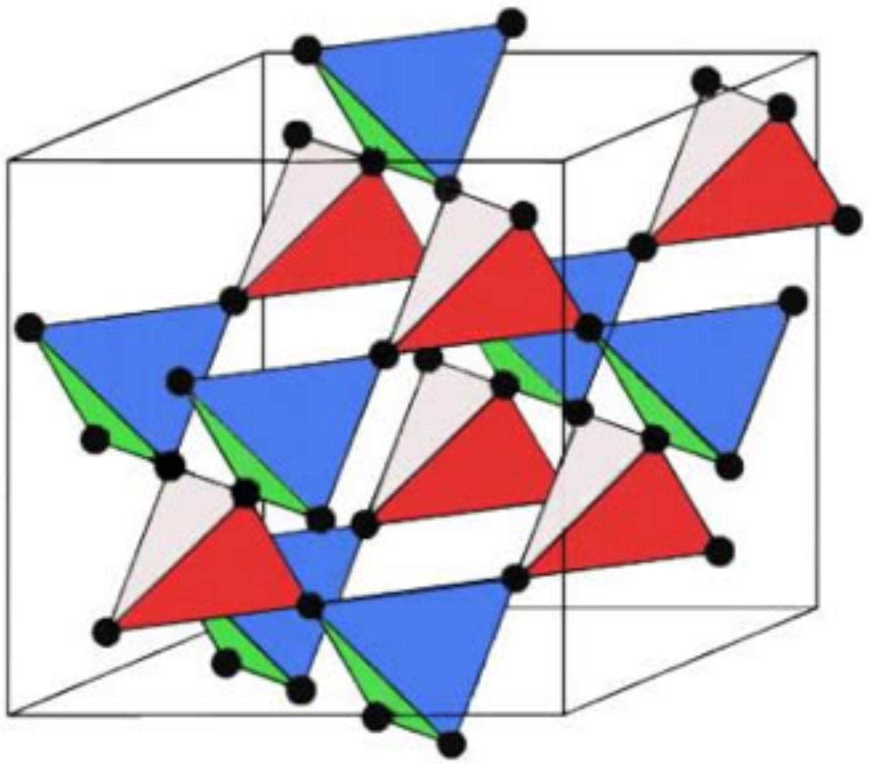


Science 294, 1495 (2001)



http://hiroi.issp.u-tokyo.ac.jp/

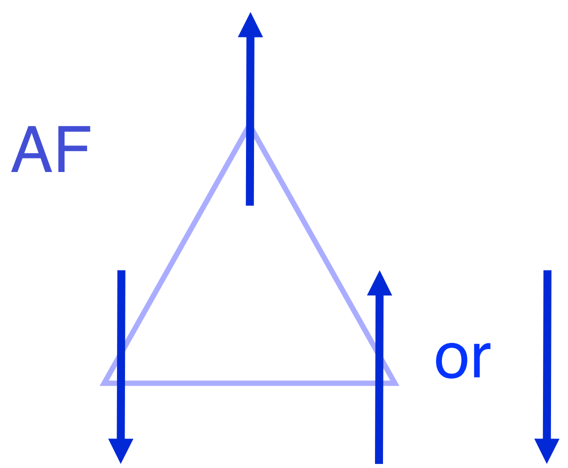
$Ho_2Ti_2O_7$
$Dy_2Ti_2O_7$

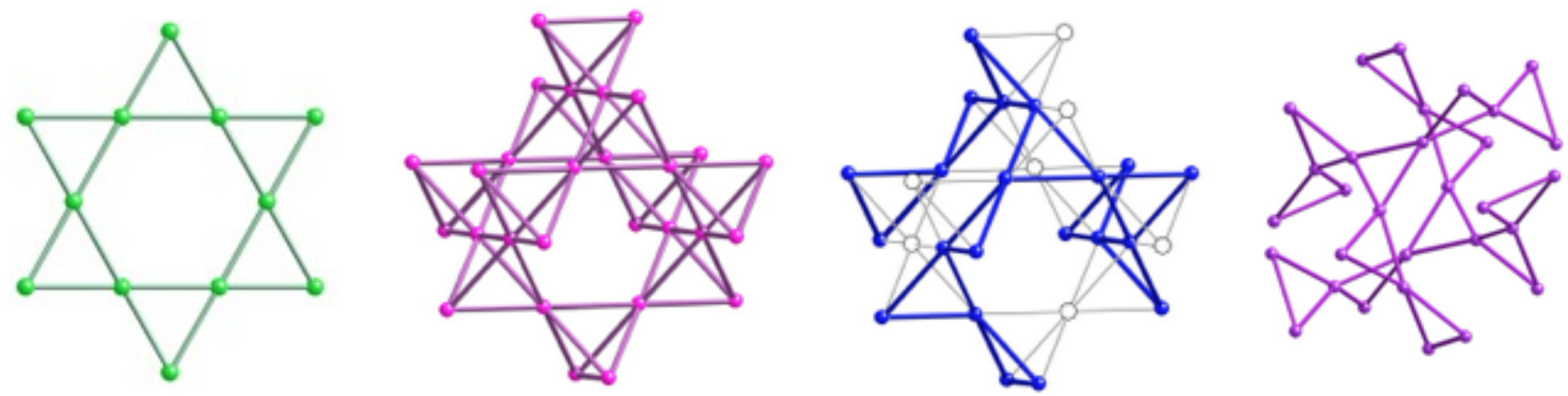


Pyrochlore

Geometrical Frustration

Geometrical Frustration



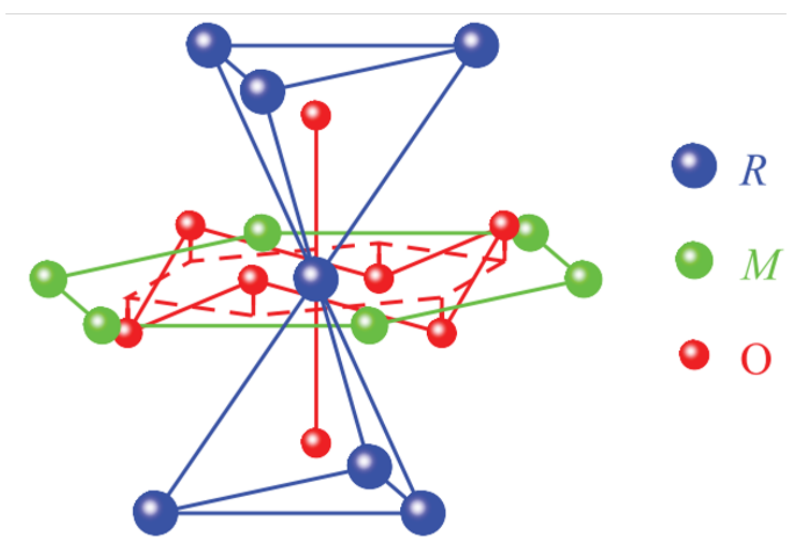


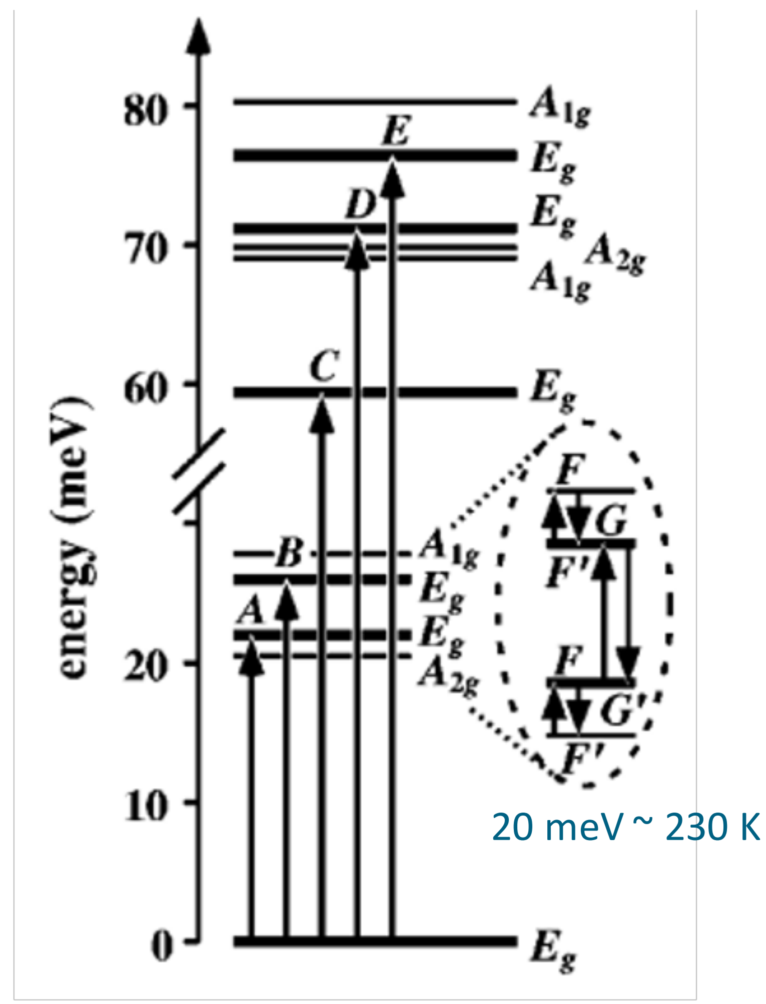
Kagome — $ZnCu_3(OH)_6Cl_2$ | Pyrochlore — $Tb_2Ti_2O_7$ | Hyperkagome — $Na_4Ir_3O_8$ | Garnet — $Gd_3Ga_5O_{12}$

- Large number of degenerate classical ground states
- Small perturbation leads to novel ground states

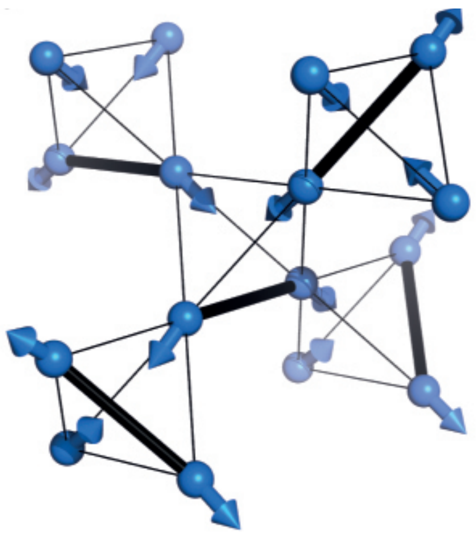
⟨111⟩ Ising Anisotropy

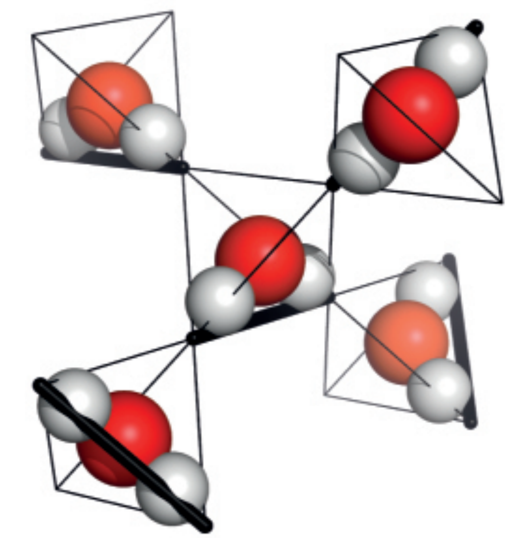
$Ho_2Ti_2O_7$
$Dy_2Ti_2O_7$





Crystal field levels of Ho^{3+} in $Ho_2Ti_2O_7$

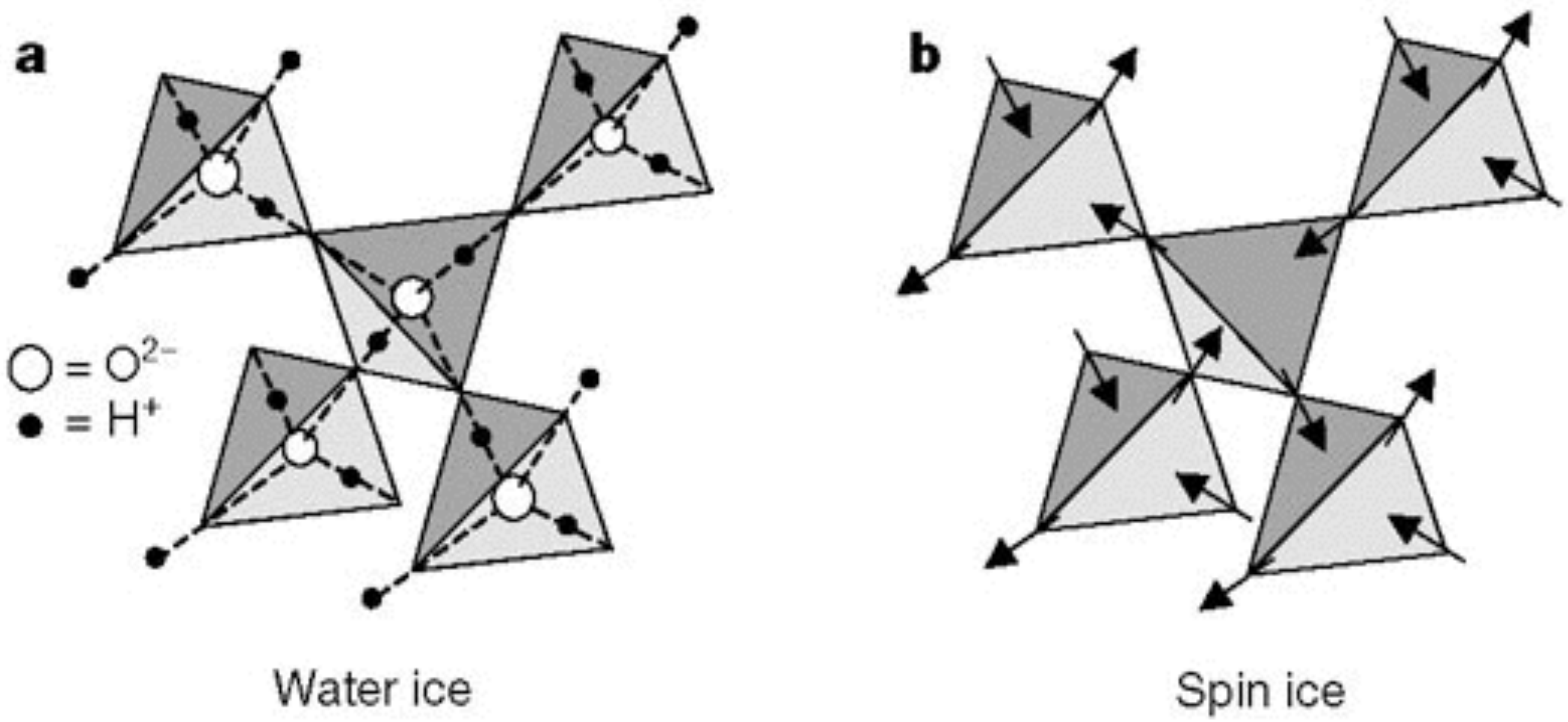




A. Yaouanc et al., Phys. Rev. B **84**, 172408 (2011)
S.Rosenkranz et al., J. Appl. Phys. **87**, 5914 (2000)
D. A. Keen and A. L. Goodwin, Nature **521**, 303 (2015)

Spin Ice

Magnetic Equivalent of Water Ice



Nature 413, 48-51 (2001)

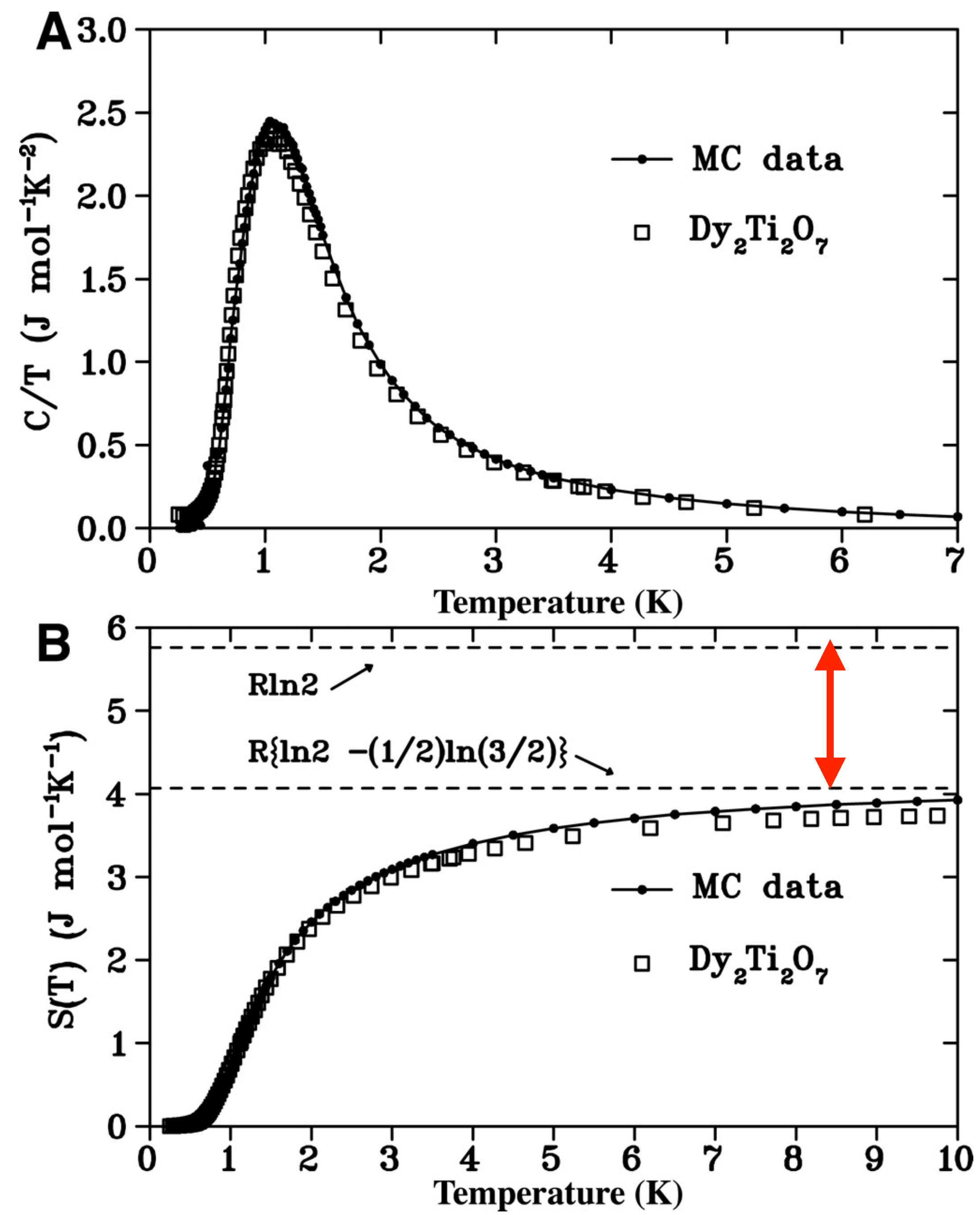
Residual Entropy

- $\langle 111 \rangle$ Ising spins with ferromagnetic interaction on a pyrochlore lattice
- Number of ground states:

$$\Omega \approx 6^{N/4}(6/16)^{N/4}$$

- Residual Entropy per spin:

$$S_{T=0} = k_B \frac{1}{2} \ln \frac{3}{2}$$



Same as Water Ice! (Pauling, 1935)

A.P. Ramirez et al., Nature 399, 333, 1999
B.C. den Hertog et al., PRL 84, 3430, 2000

Wrong Model? Correct Physics!

- Ice Rules: Nearest Neighbor Spin Ice (NNSI) Model

$$H_{\rm nn} = -\frac{J}{3}\sum_{\langle ij\rangle}\sigma_i\cdot\sigma_j$$

- Real Material: Ho^{3+}, Dy^{3+} have large moments, dipolar interaction is dominant!
- Dipolar Spin Ice (DSI) Model

$$H_{\rm dip} = -\frac{J}{3}\sum_{\langle ij\rangle}\sigma_i\cdot\sigma_j + Da^3\sum_{i<j}\left(\frac{3\hat{\mathbf{e}}_i\cdot\hat{\mathbf{e}}_j - (\hat{\mathbf{e}}_i\cdot\hat{\mathbf{r}}_{ij})(\hat{\mathbf{e}}_j\cdot\hat{\mathbf{r}}_{ij})}{r_{ij}^3}\right)\sigma_i\sigma_j$$

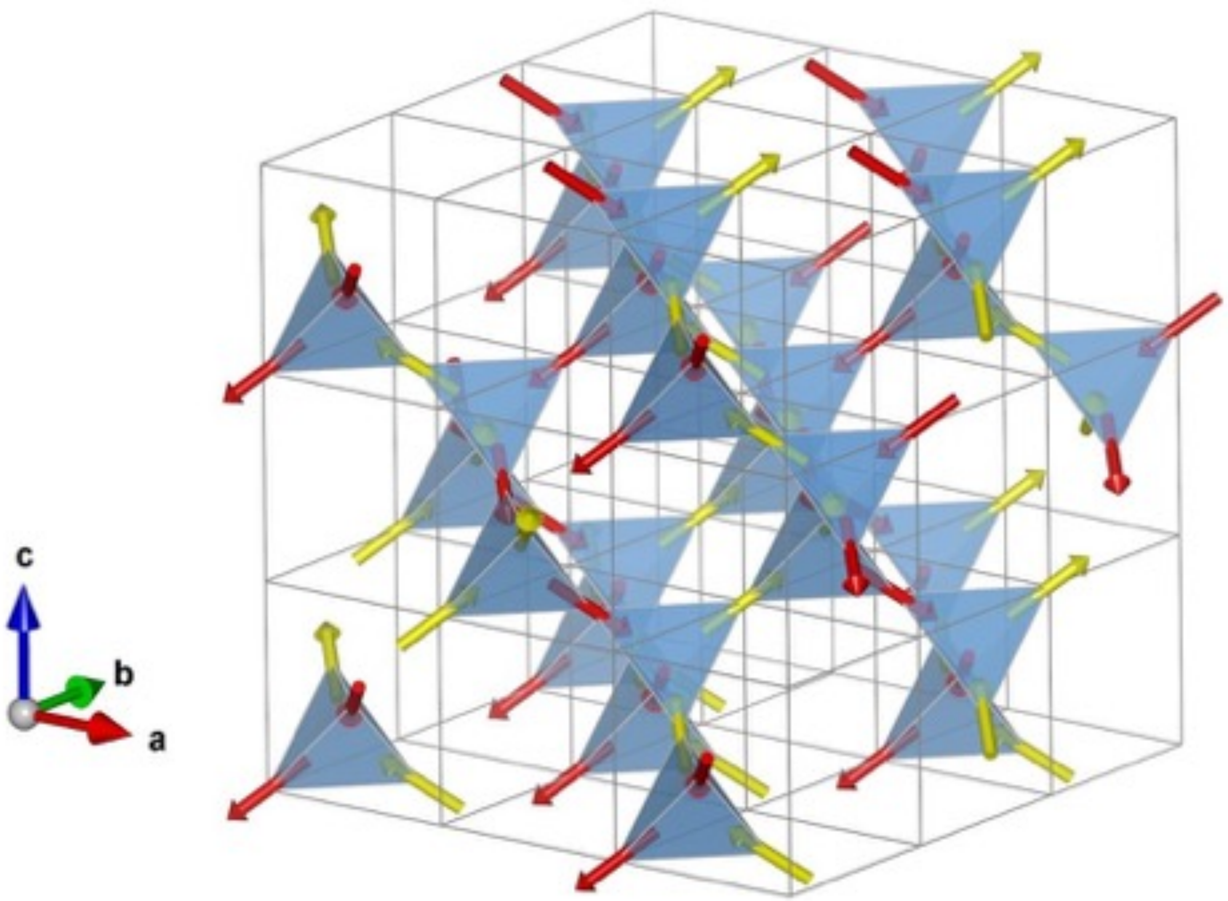
- Why does the spin ice materials obey ice rules?
- Self-screening of the dipolar interaction in the pyrochlore lattice: Projective Equivalence

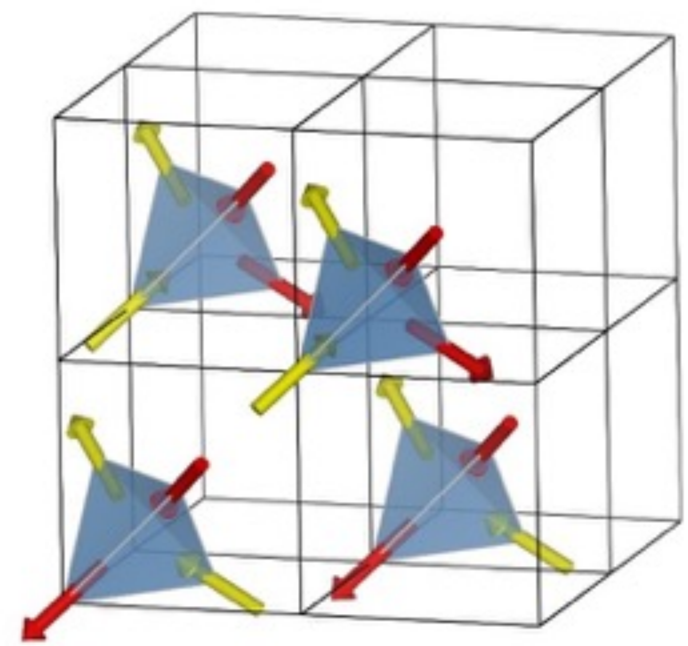
M. J. P. Gingras and B. C. den Hertog, Can. J. Phys. 79, 1339 (2001),
M. Enjalran and M. J. P. Gingras, Phys. Rev. B 70, 174426 (2004),
R. G.Melko and M. J. P. Gingras, J. Phys. Condens. Matter 16, R1277 (2004),
S.V. Isakov, R. Moessner, and S. L. Sondhi, Phys. Rev. Lett., 95 217201 (2005).

Ground State of DSI

$$H = -J \sum_{<(i,a),(j,b)>} \mathbf{S}_\mathrm{i}^\mathrm{a} \cdot \mathbf{S}_\mathrm{j}^\mathrm{b} + Dr_{nn}^3 \sum_{i<j,a,b} \left(\frac{\mathbf{S}_\mathrm{i}^\mathrm{a} \cdot \mathbf{S}_\mathrm{j}^\mathrm{b}}{|\mathbf{R}_\mathrm{ij}^\mathrm{ab}|^3} - \frac{3(\mathbf{S}_\mathrm{i}^\mathrm{a} \cdot \mathbf{R}_\mathrm{ij}^\mathrm{ab})(\mathbf{S}_\mathrm{j}^\mathrm{b} \cdot \mathbf{R}_\mathrm{ij}^\mathrm{ab})}{|\mathbf{R}_\mathrm{ij}^\mathrm{ab}|^5} \right) + \mathbf{H} \cdot \sum_i \mathbf{S}_\mathrm{i}$$

- LRO ground state: MDG state
- Degeneracy lifted by the residual dipolar interaction

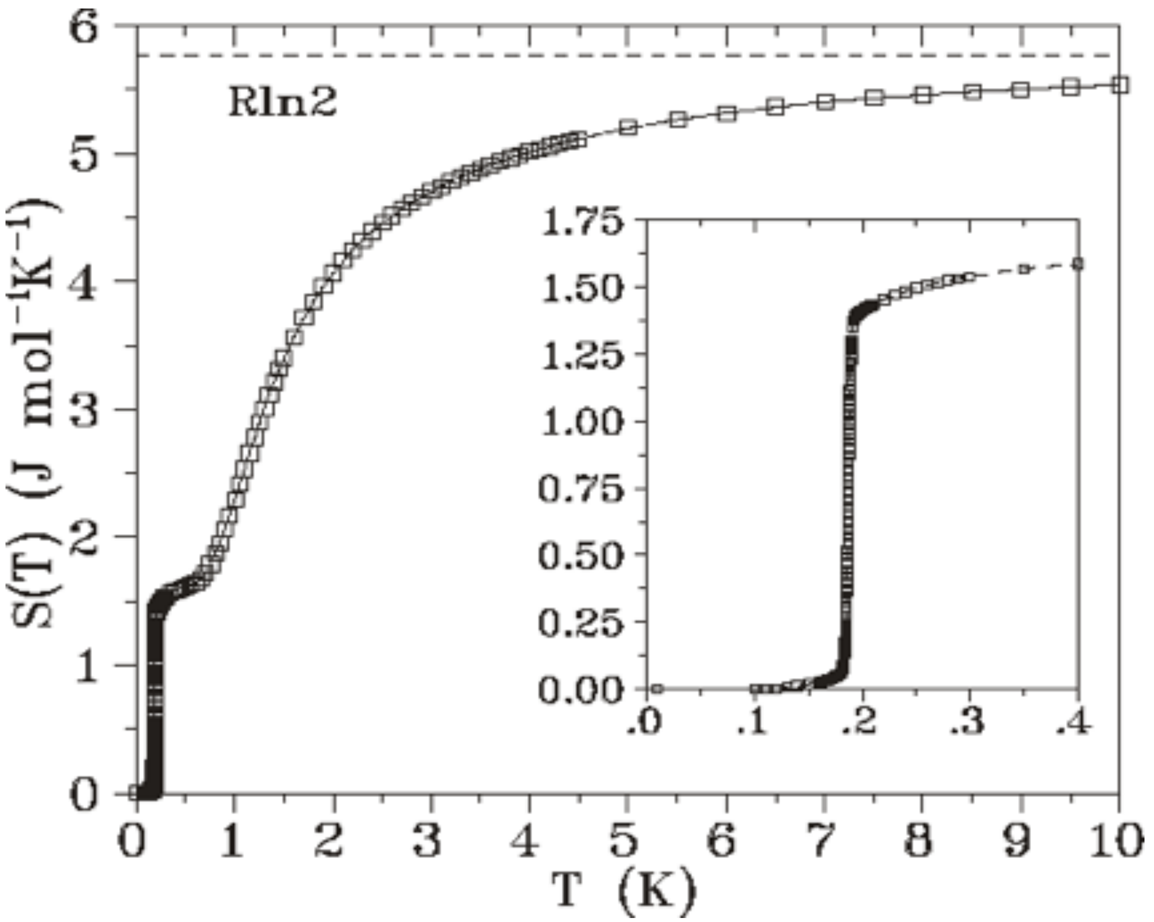


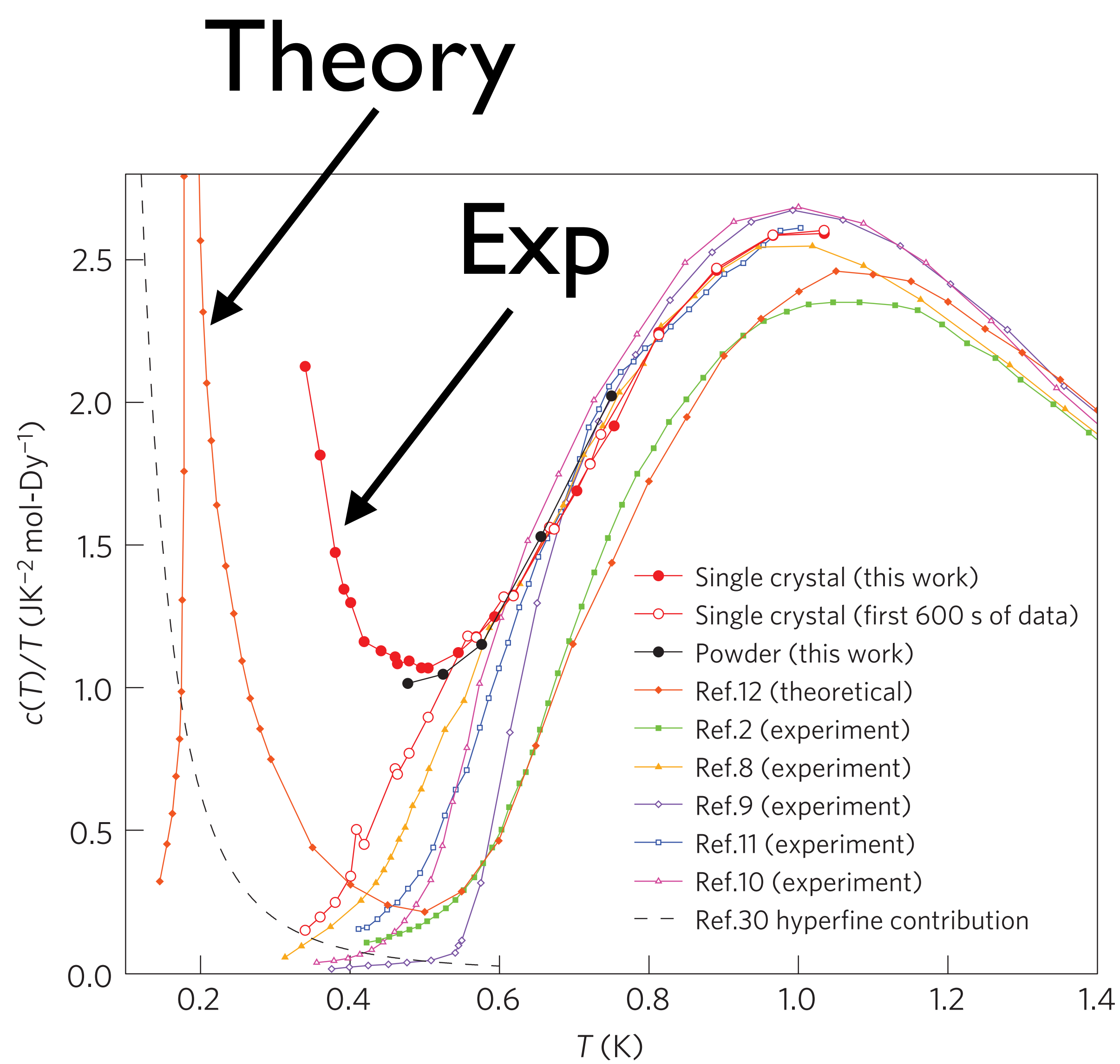


R. G. Melko, B. C. den Hertog, M. J. P. Gingras, Phys. Rev. Lett. **87**, 67203 (2001)

Ground State of DSI

- First order transition to the MDG state
    - ✦ symmetry breaking long-range order, q=(0,0,1).
- Pauling's residual entropy is recovered.
- Impurity effects?





R.G. Melko *et al.* PRL **87**, 067304; D. Pomaranski *et al.* Nat. Phys., **9**, 353; P. Henelius *et al.* Phys. Rev. B 93, 024402 (2016)

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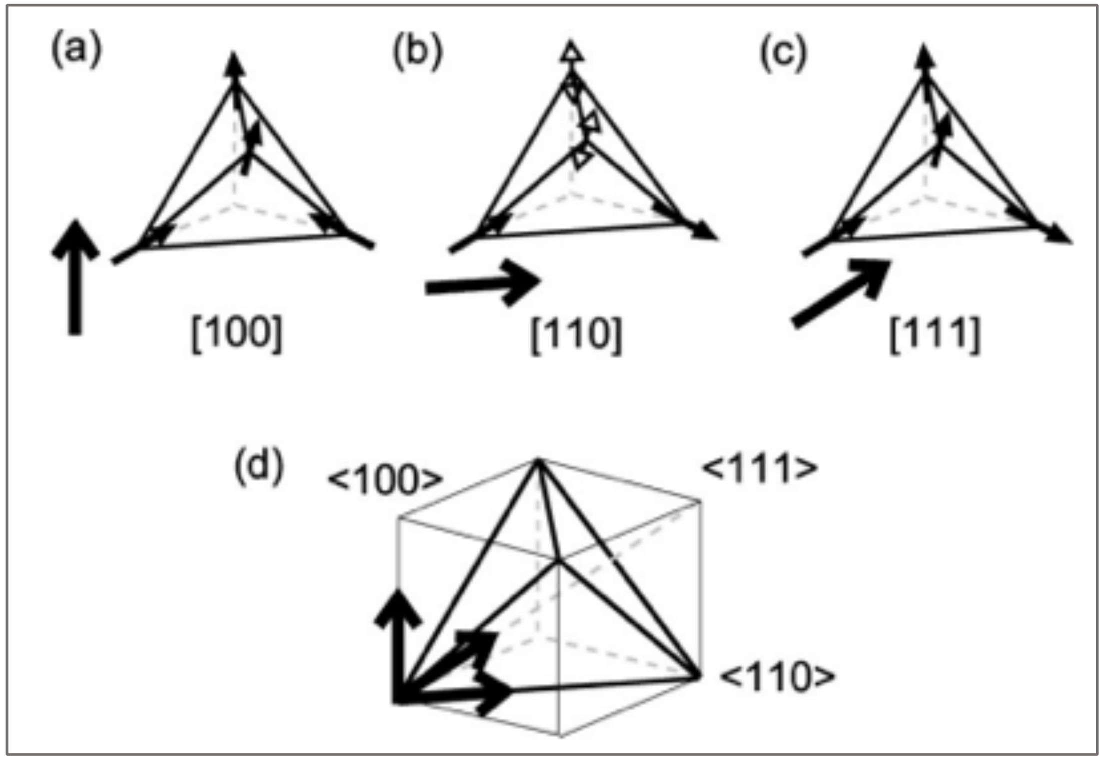
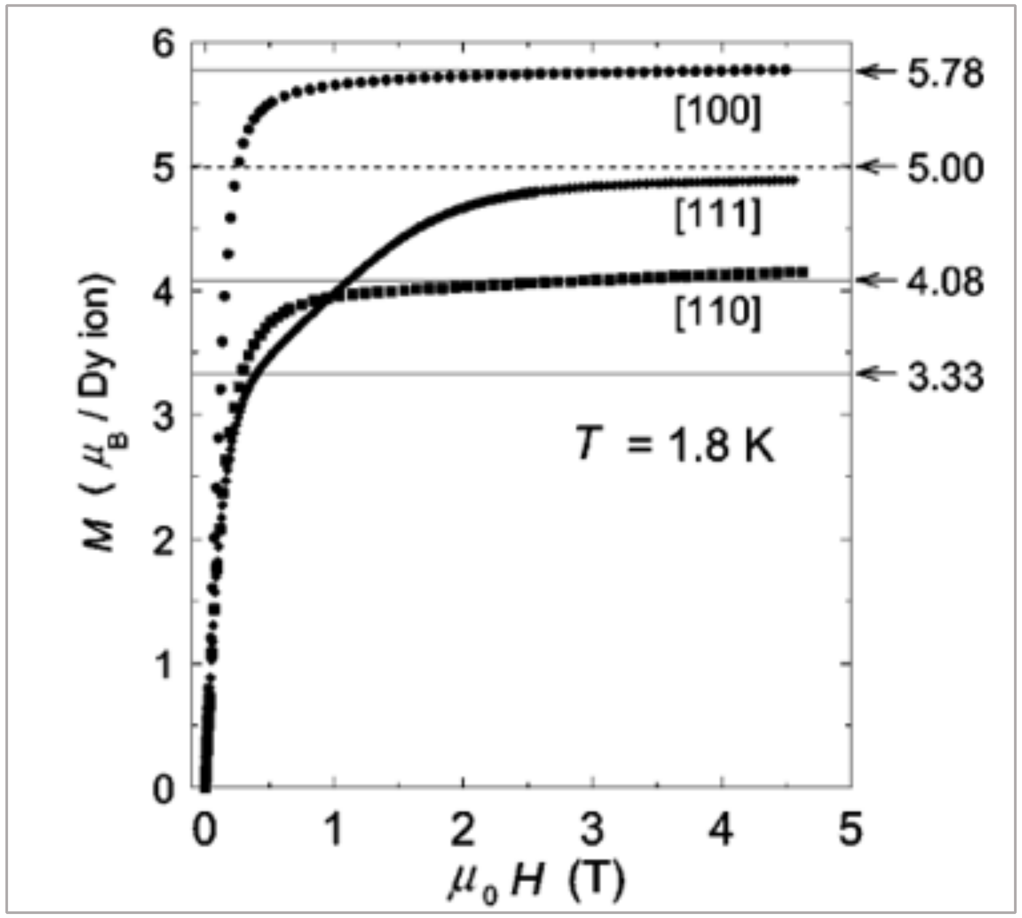
Magnetic Anisotropy

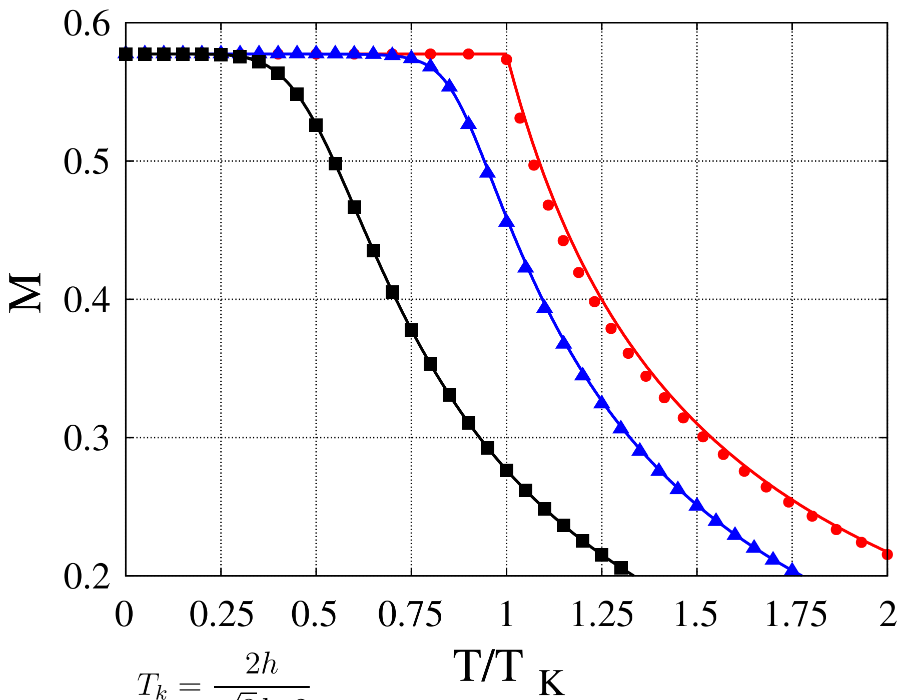
- [100] field: $10\mu_B/\sqrt{3} = 5.77\mu_B$
- [111] field: $10 \times (1/3 \times 3 + 1)/4 = 5.00$
- [110] field: $10 \times \left(2 \times \sqrt{2/3}\right)/4 = 4.08$





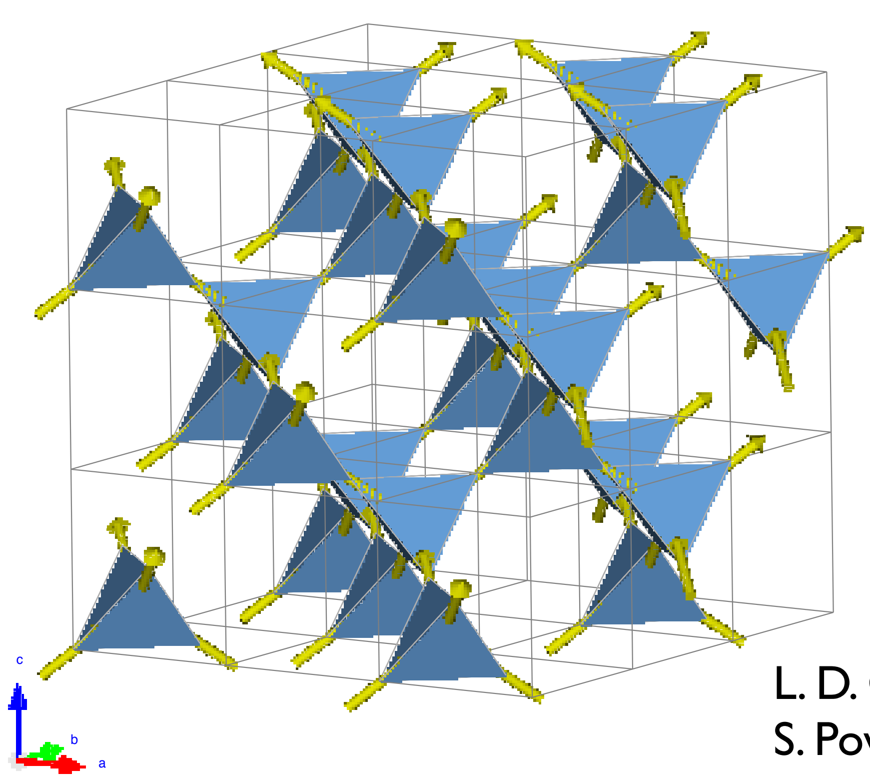
H. Fukazawa *et al.*,
Phys. Rev. B **65**, 54410 (2002)

NNSI in a [100] Field



$T_k = \frac{2h}{\sqrt{3}\ln 2}$

$h/J = 10^{-3}$ (●), 0.13 (▲), and 0.58 (■)



- Kasteleyn transition
  - ★ From the Coulomb phase to a fully polarized (FP) state
  - ★ Proliferation of `
  - ★ Classical to quantum mapping 2D bosonic model Bose condensate to vacuum transition (field $\leftrightarrow \mu$)
- FP state: q=(000)

L. D. C. Jaubert, J. T. Chalker, P. C. W. Holdsworth, and R. Moessner, PRL 100, 067207 (2008)
S. Powell and J. T. Chalker, Phys. Rev. B 78, 024422 (2008).

Ground States

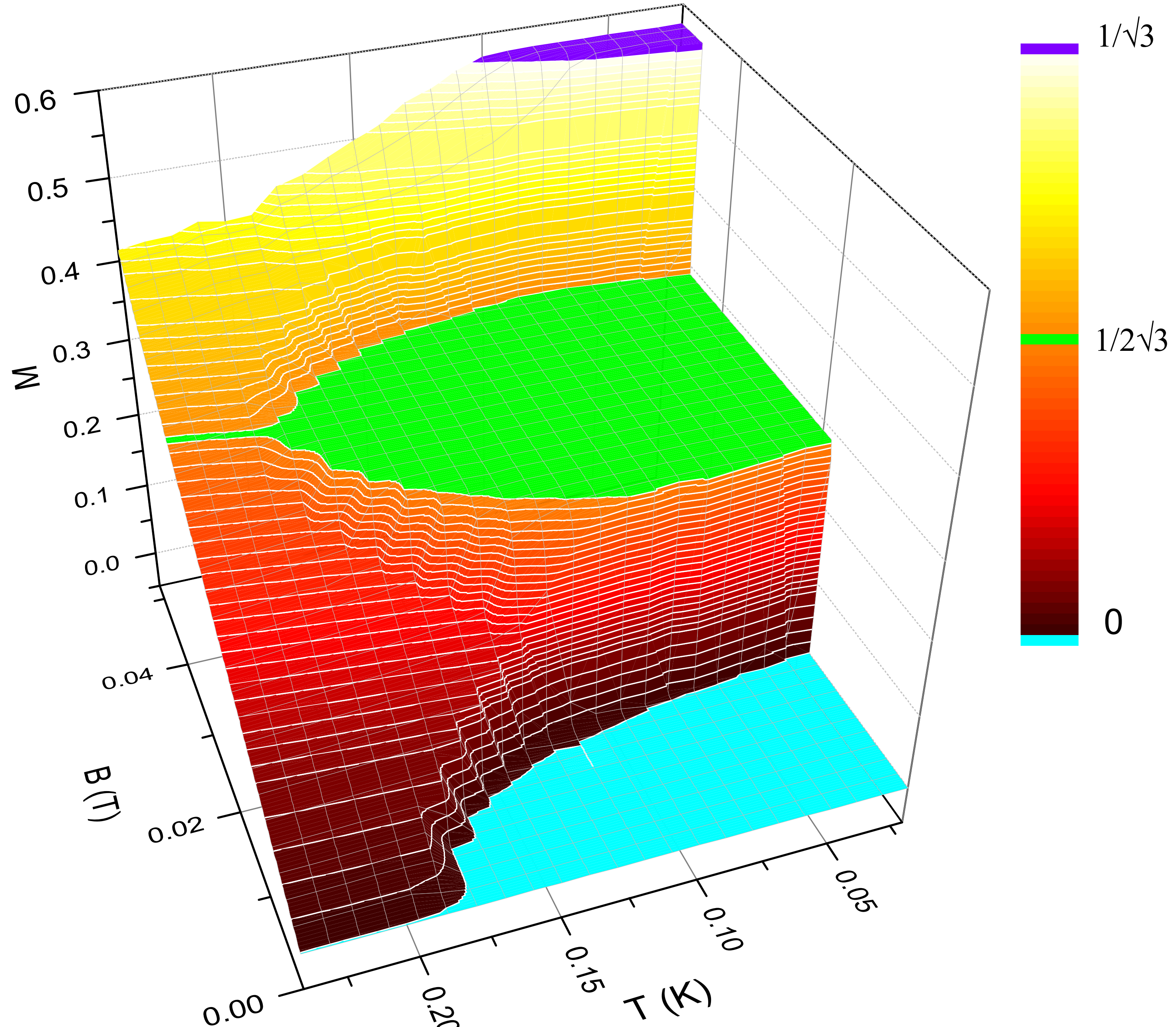
	NN Spin Ice	Dipolar Spin Ice
B=0	Coulomb Phase	MDG state q=(001)
B ll [100]	Fully Polarized State q=(000)	???

What is the ground state of a DSI in a [100] field?

High T: Coulomb Phase

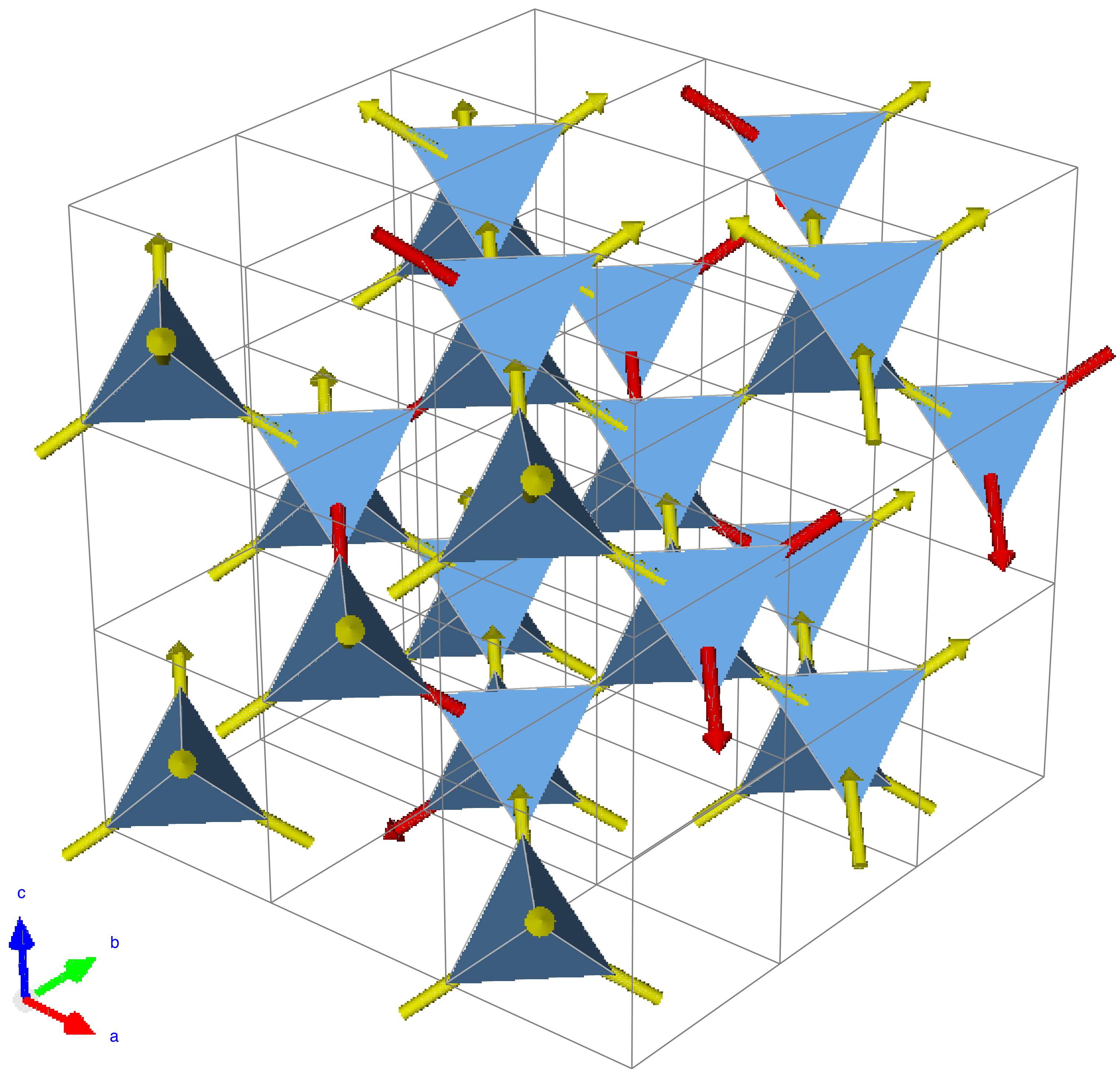
Low B: MDG, High B: FP

Magnetization Plateau



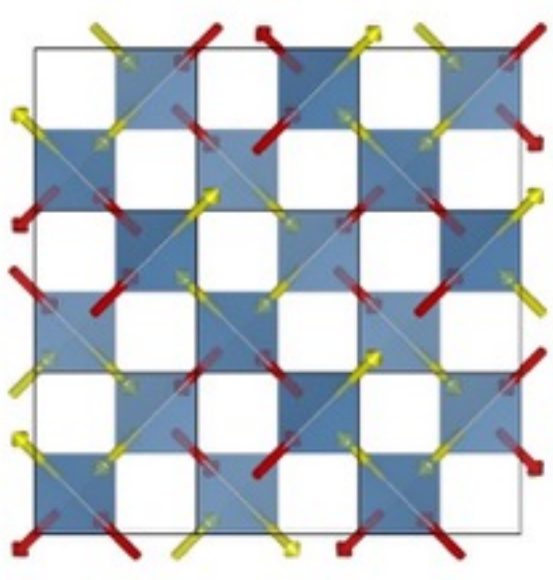
Lin and YJK, Phys. Rev. B 88, 220402(R) (2013)

Half-polarized state

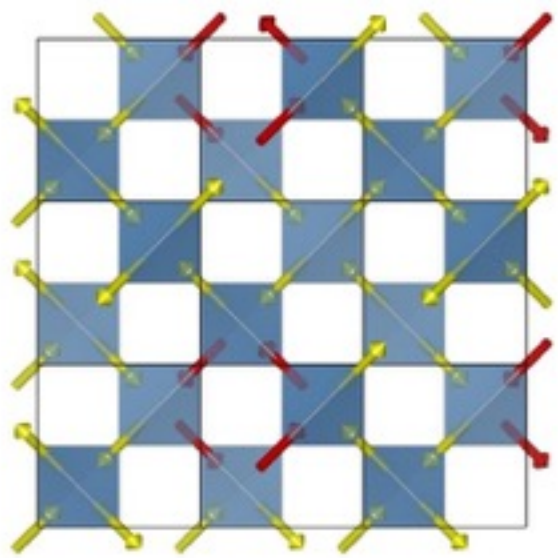


Lin and YJK, Phys. Rev. B 88, 220402(R) (2013)

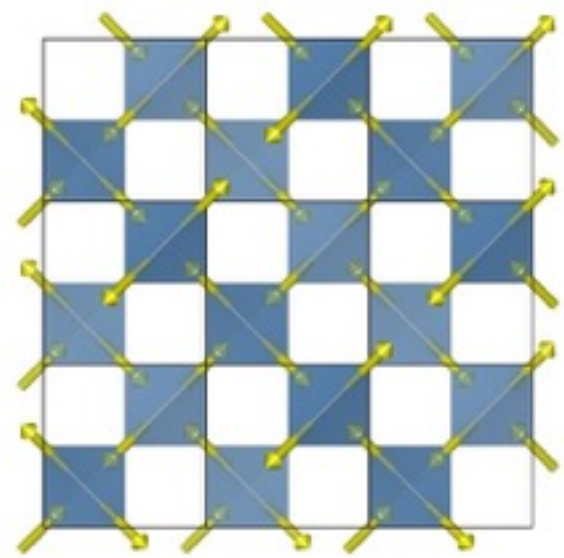
Dipolar Spin Ice in [100] Field



MDG

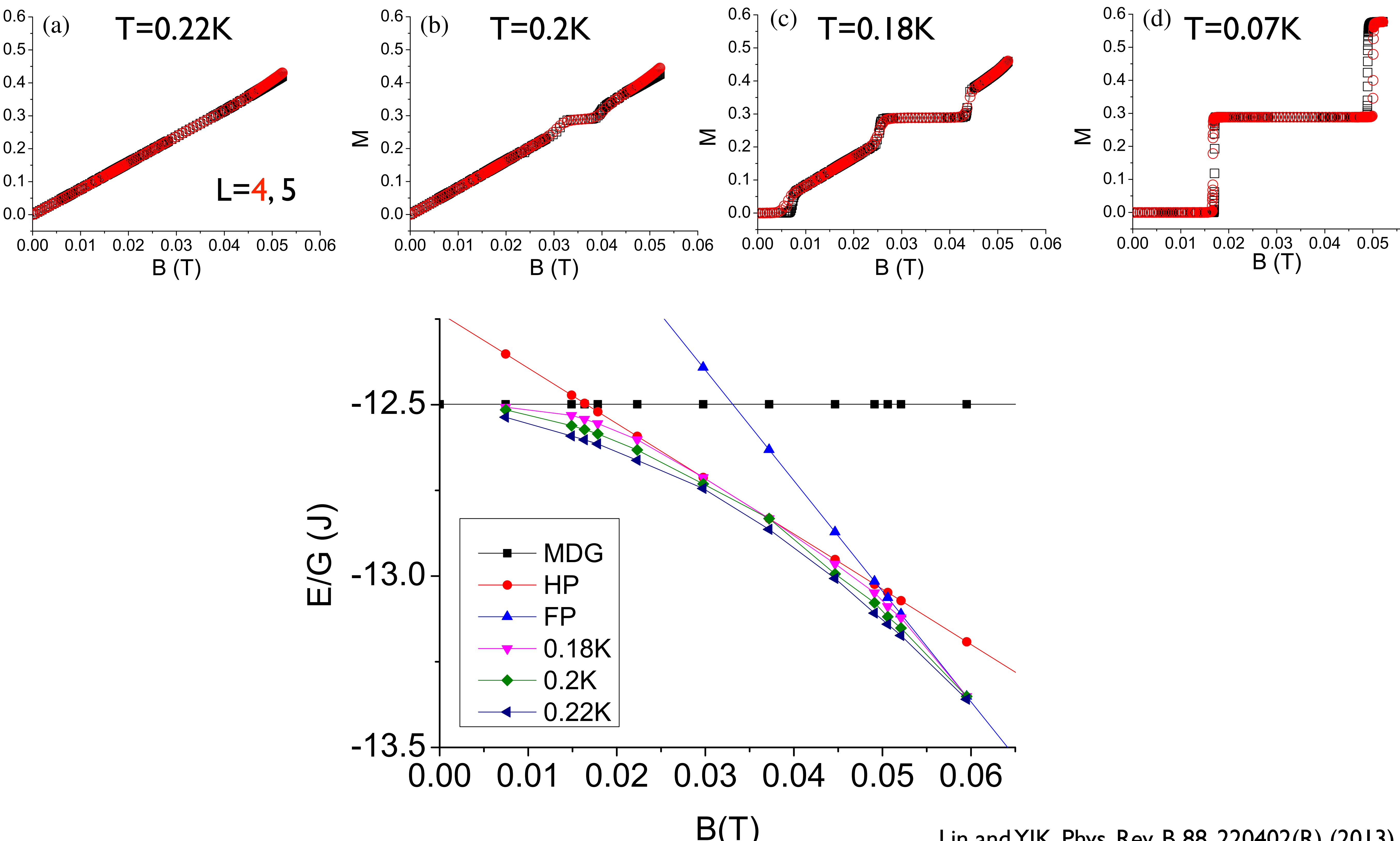


HP



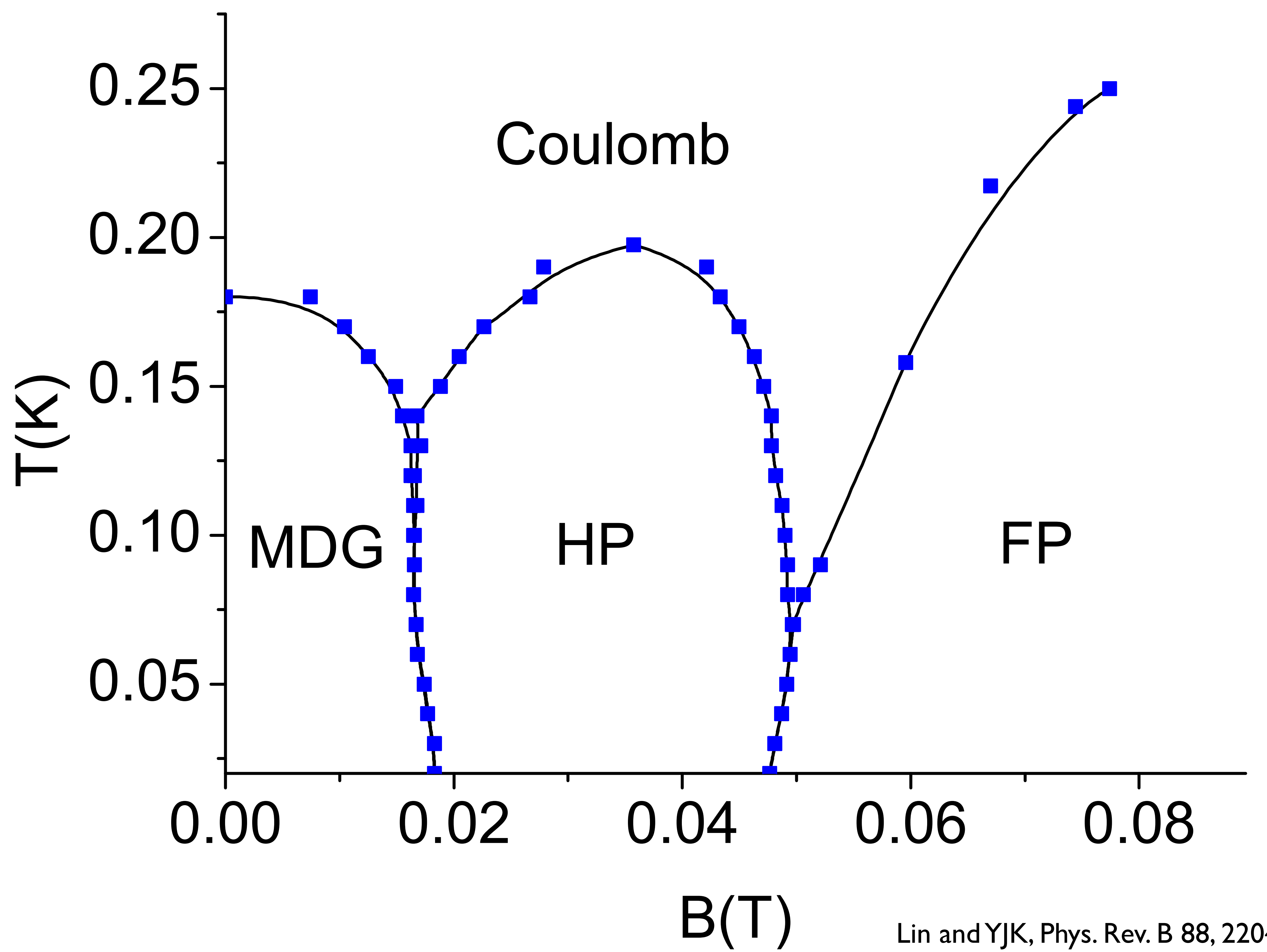
FP

Magnetization Plateau



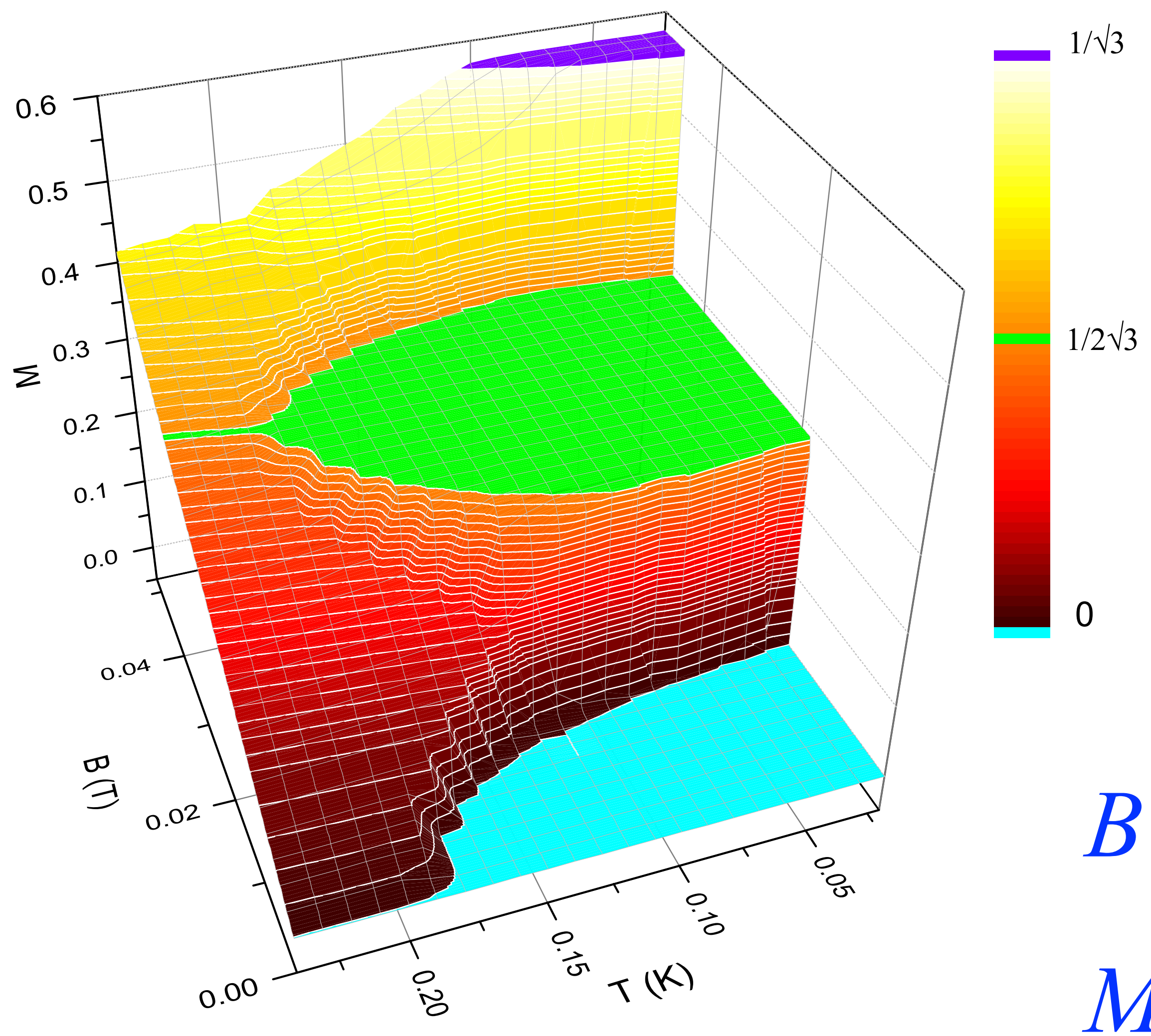
Lin and YJK, Phys. Rev. B 88, 220402(R) (2013)

Phase Diagram



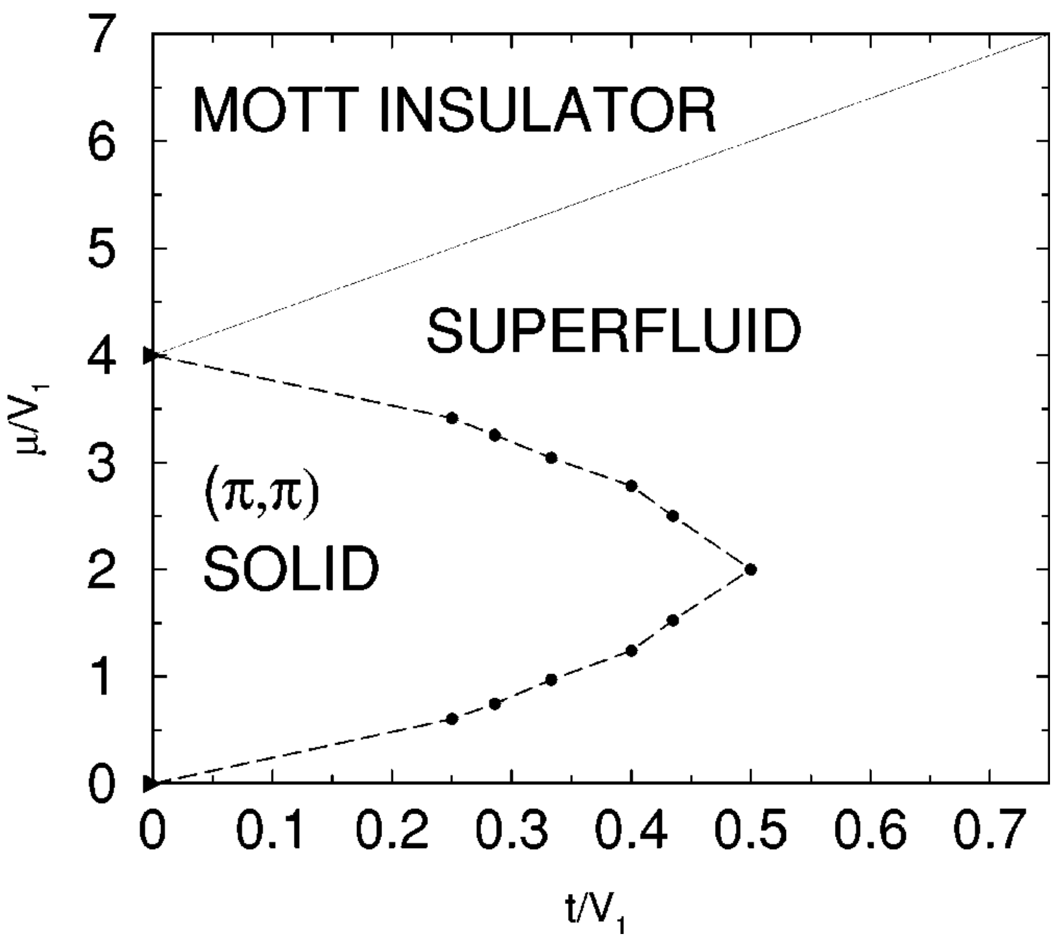
Lin and YJK, Phys. Rev. B 88, 220402(R) (2013)

2D Bosonic model



$B \leftrightarrow \mu$

$M \leftrightarrow n$



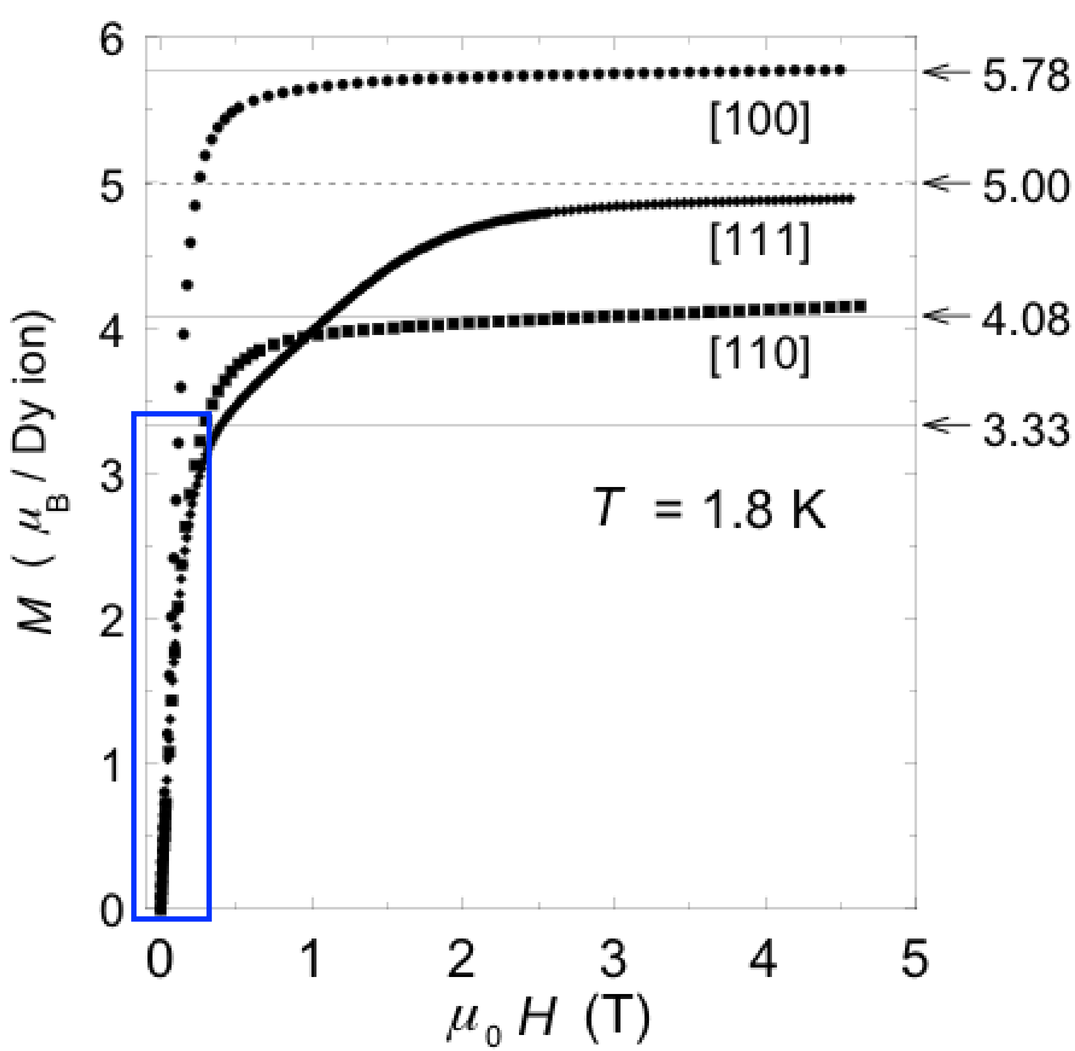
Long-range dipolar interaction

NN repulsion

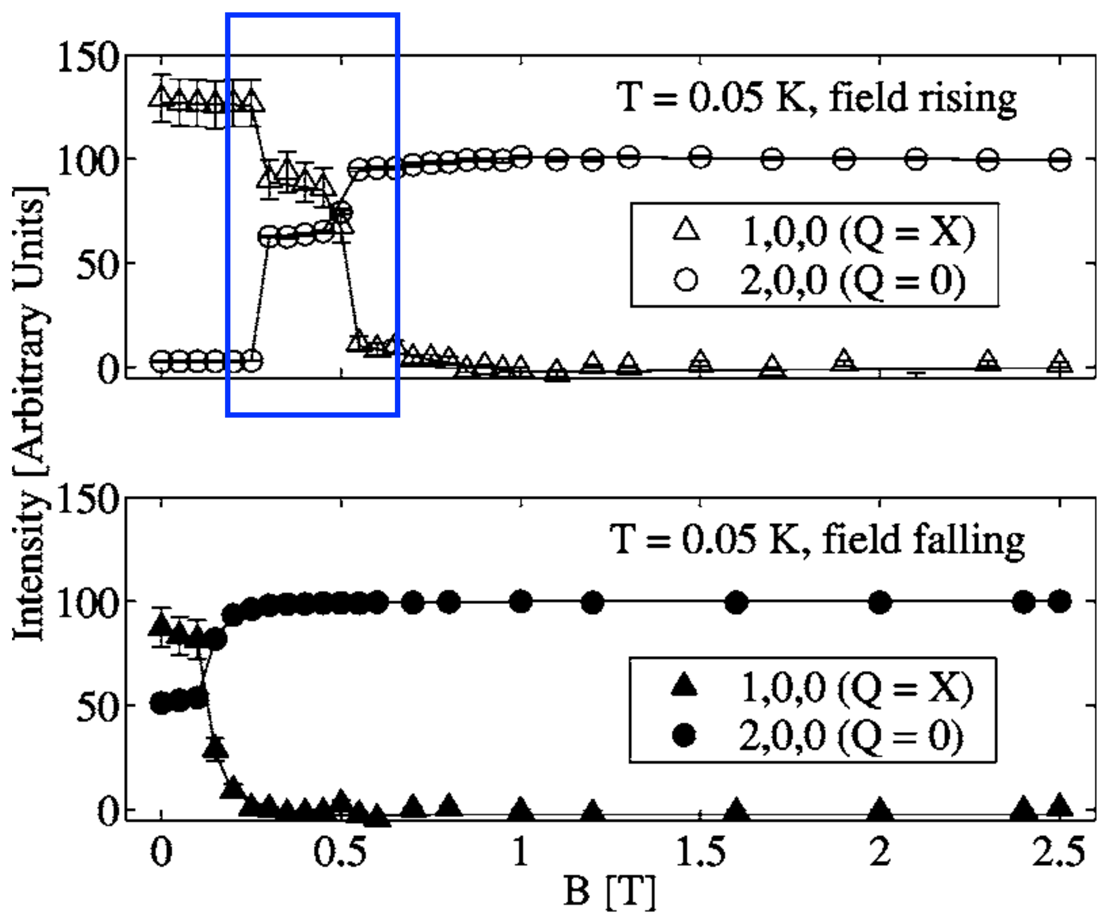
$$H = -t \sum_{\langle ij \rangle} \left(b_i^\dagger b_j + \text{h. c.} \right) + \boxed{V} \sum_{\langle ij \rangle} n_i n_j - \mu \sum_i n_i,$$

Correct effective model?

Experimental Signatures



H. Fukazawa *et al.*, Phys. Rev. B **65**, 054410 (2002)



T. Fennel *et al.*, Phys. Rev. B **72,** 224411 (2005)

High T experiment

Field too high, out of equilibrium?

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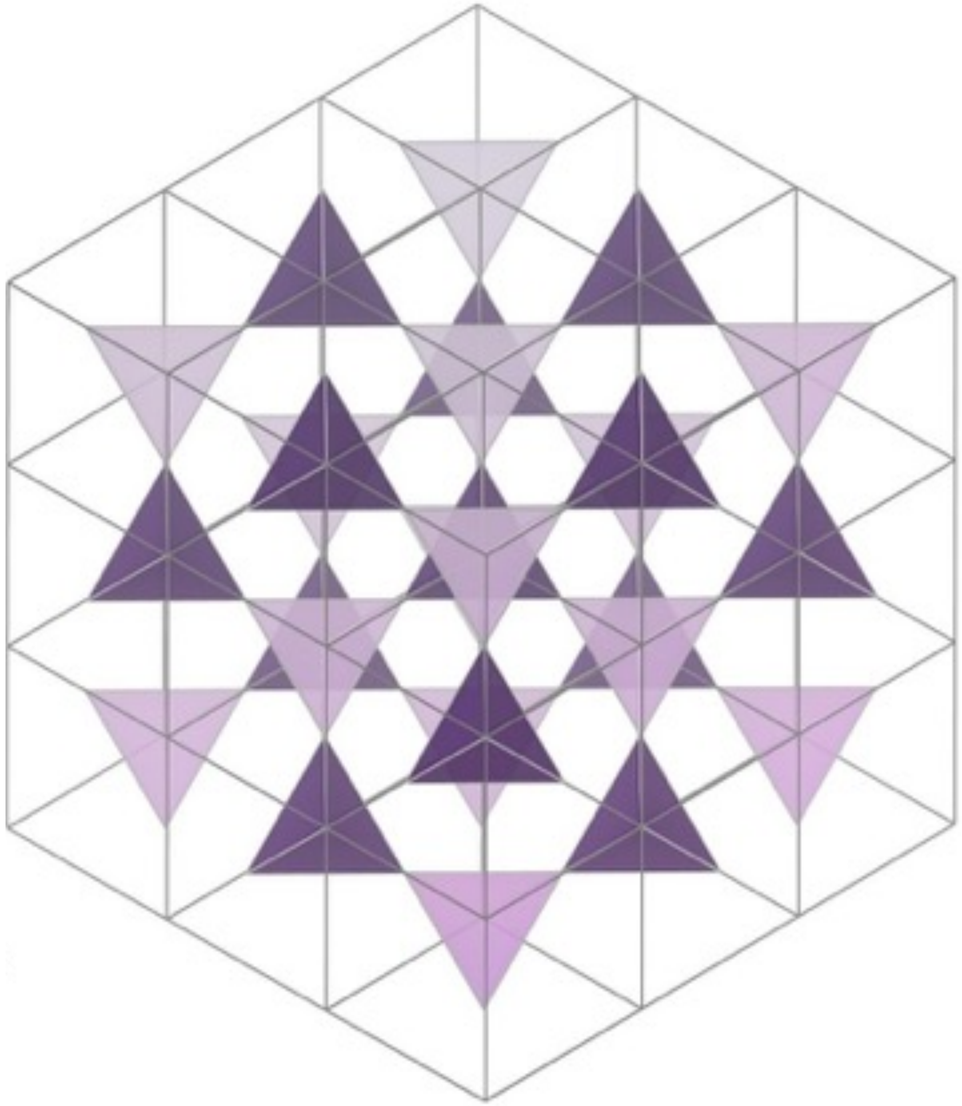
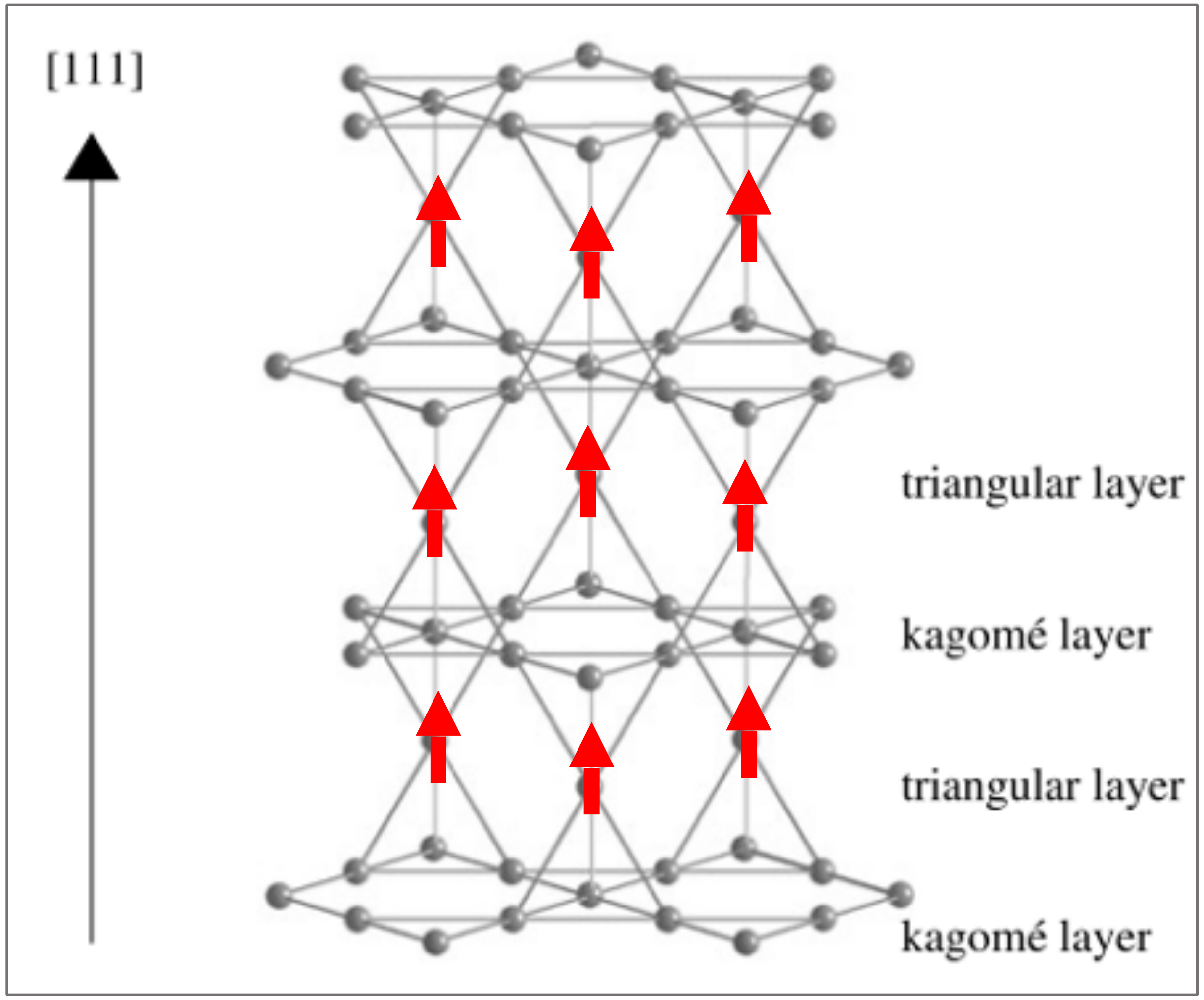
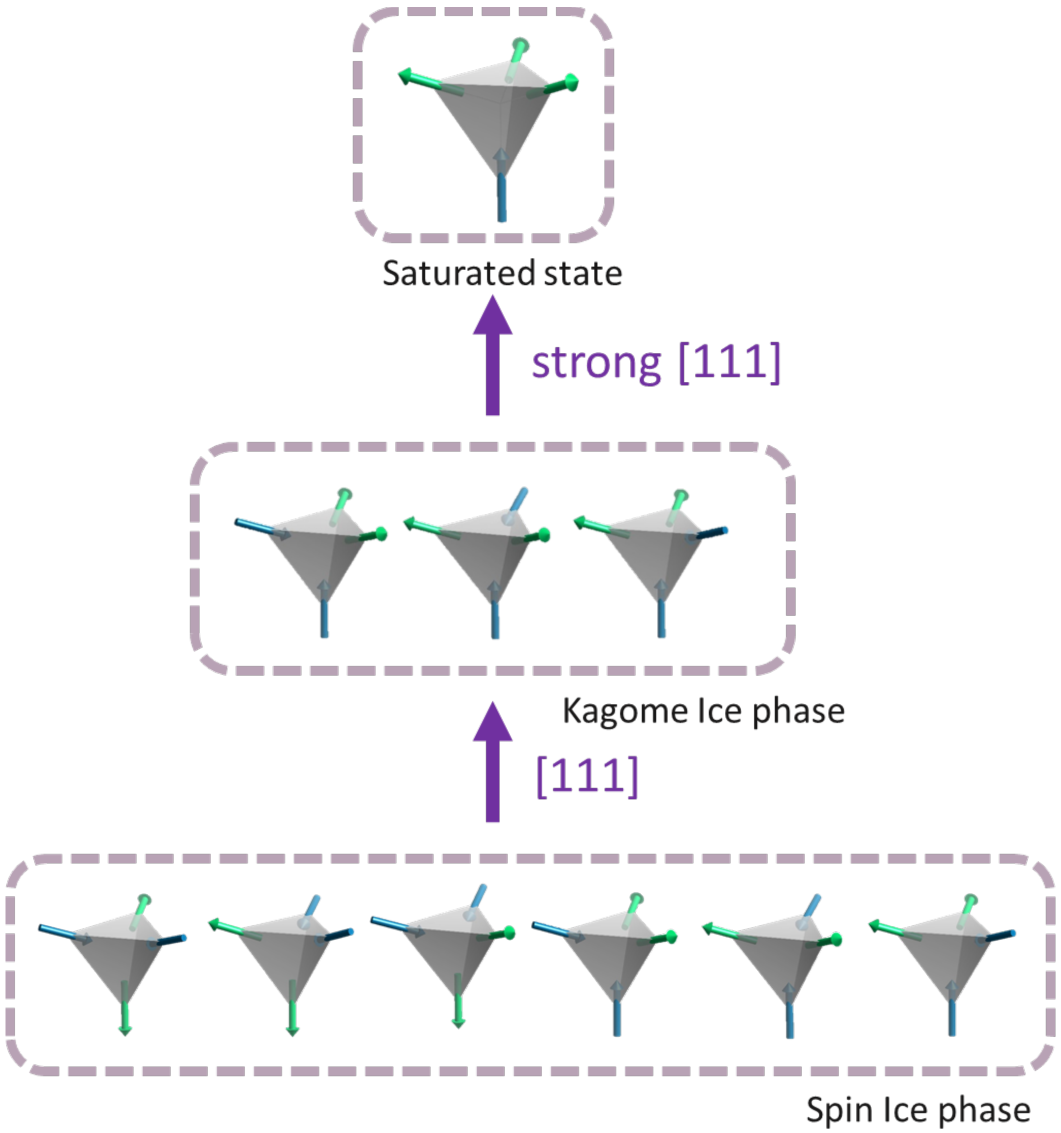
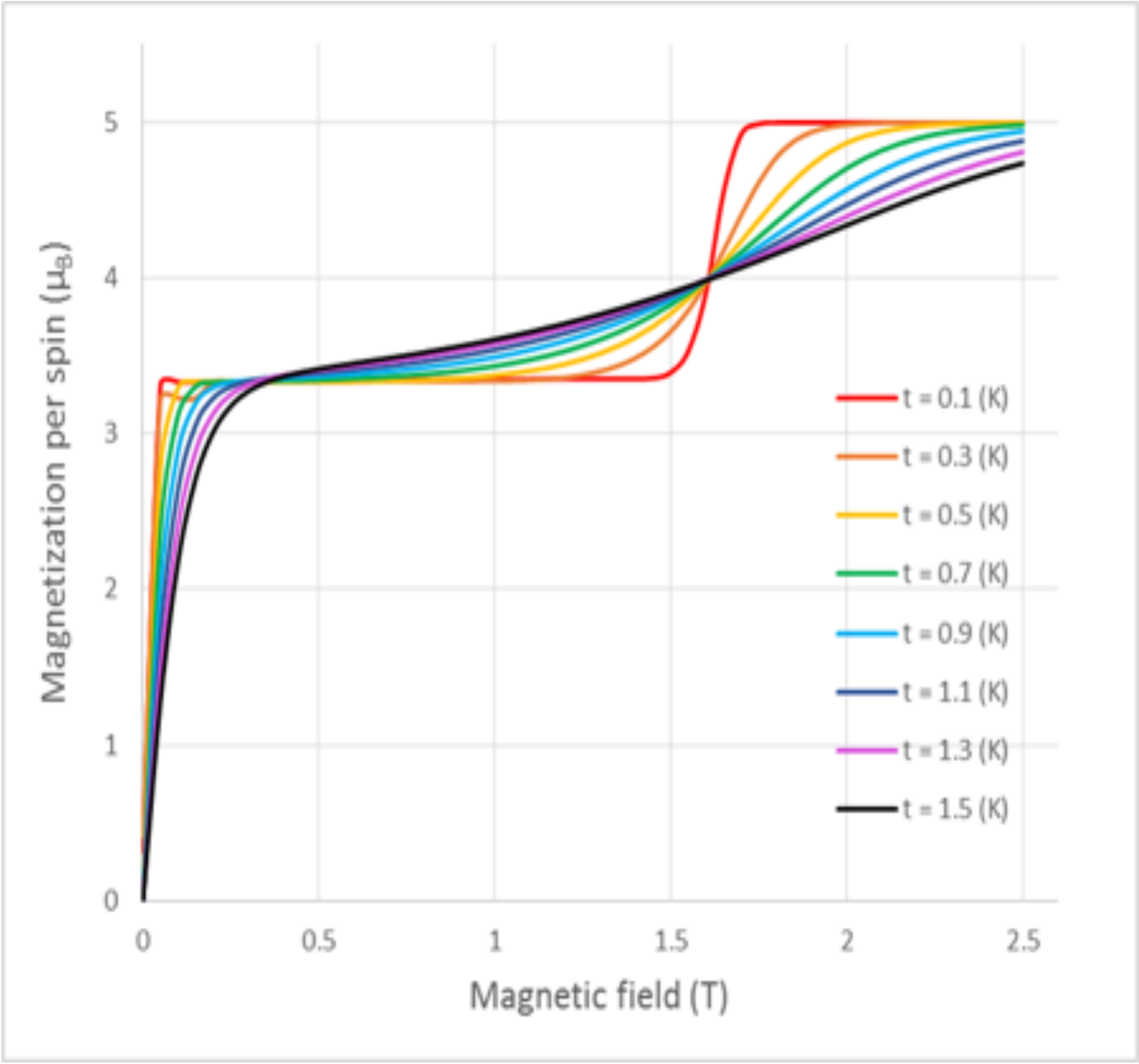
[111]: Kagome Planes



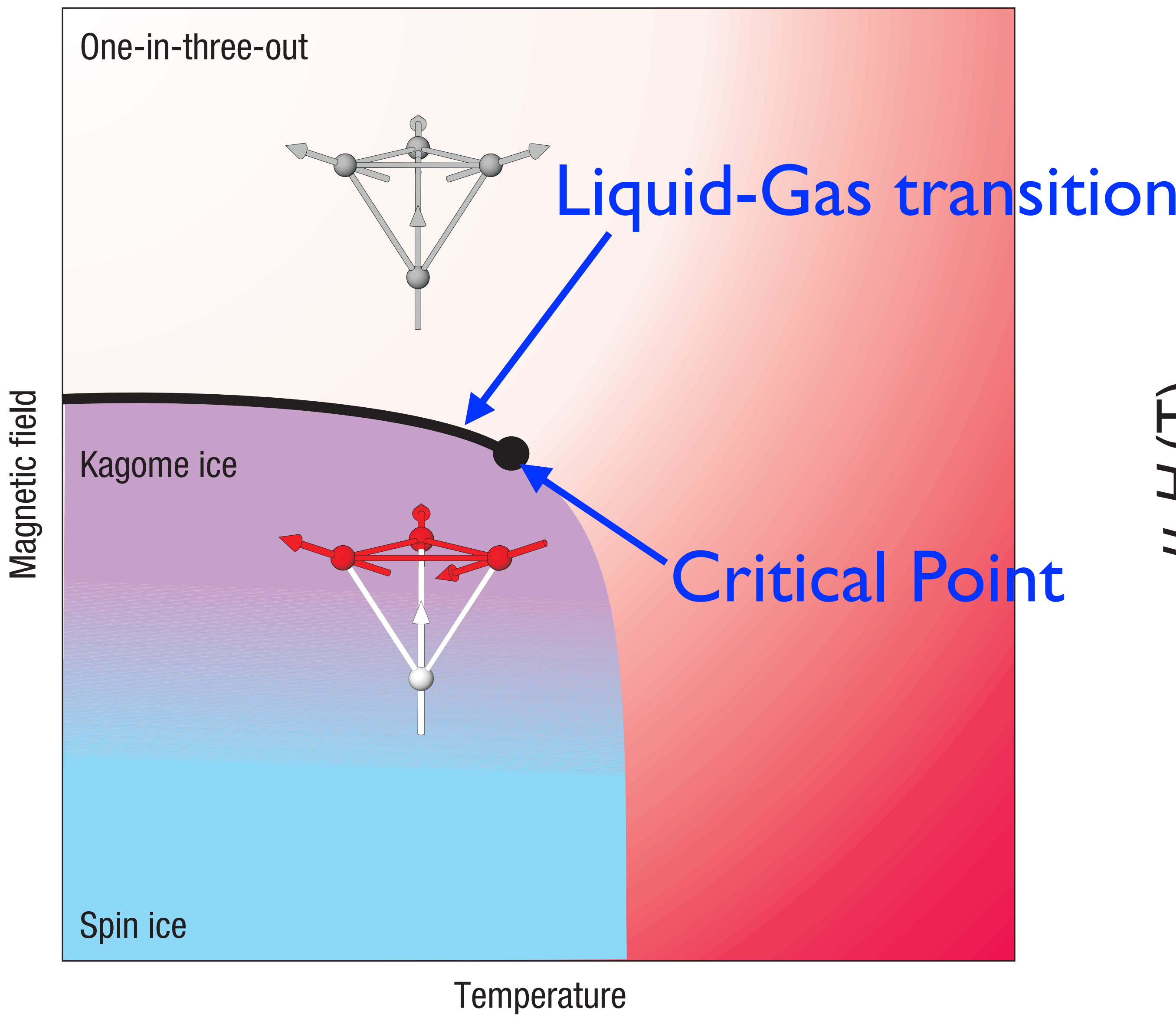


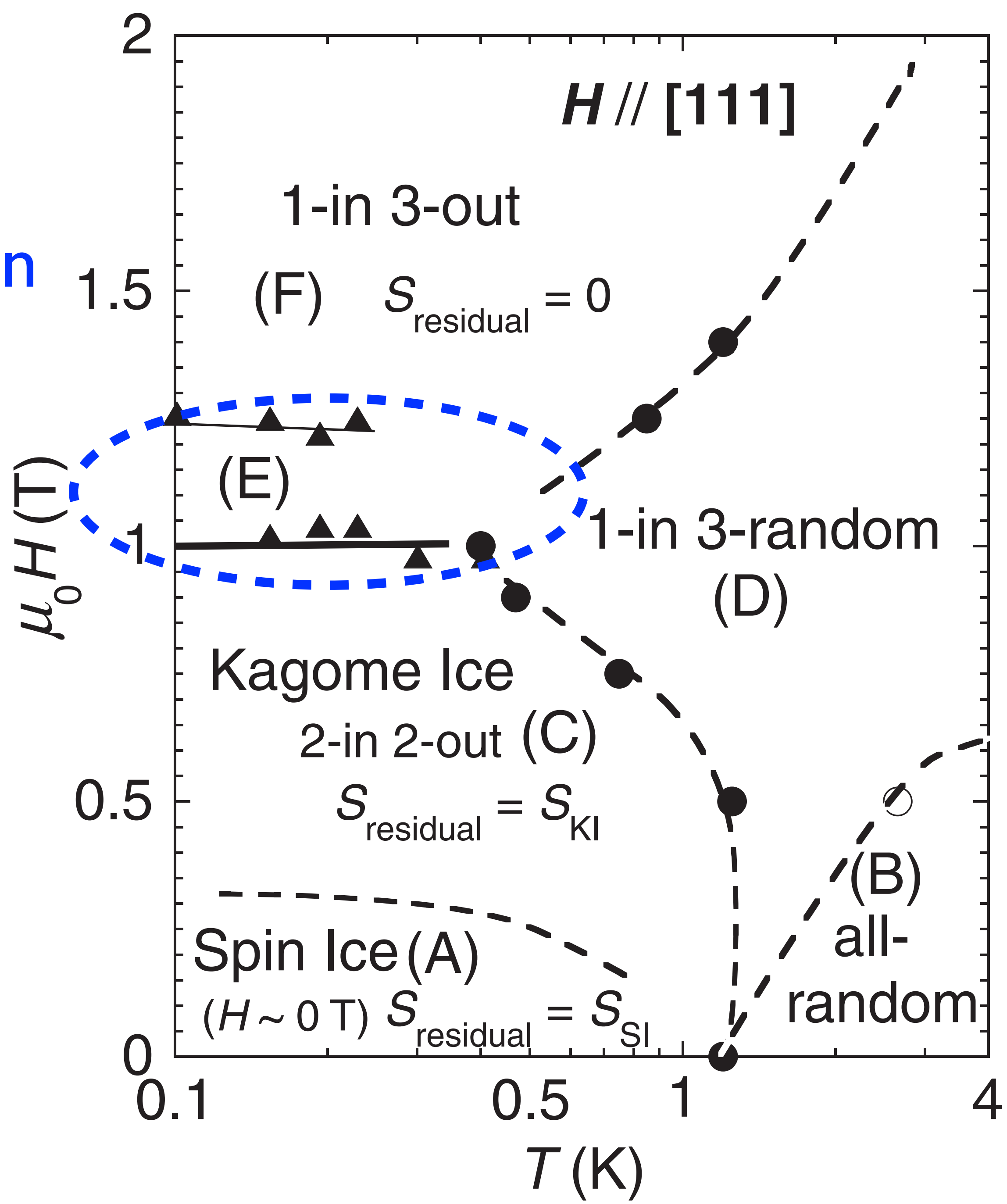
Two-Stage Magnetization Plateau





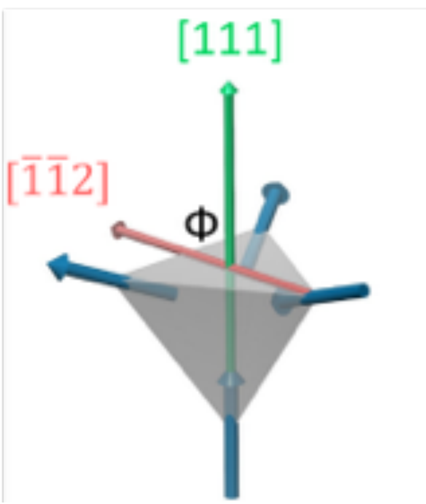
Phase diagram



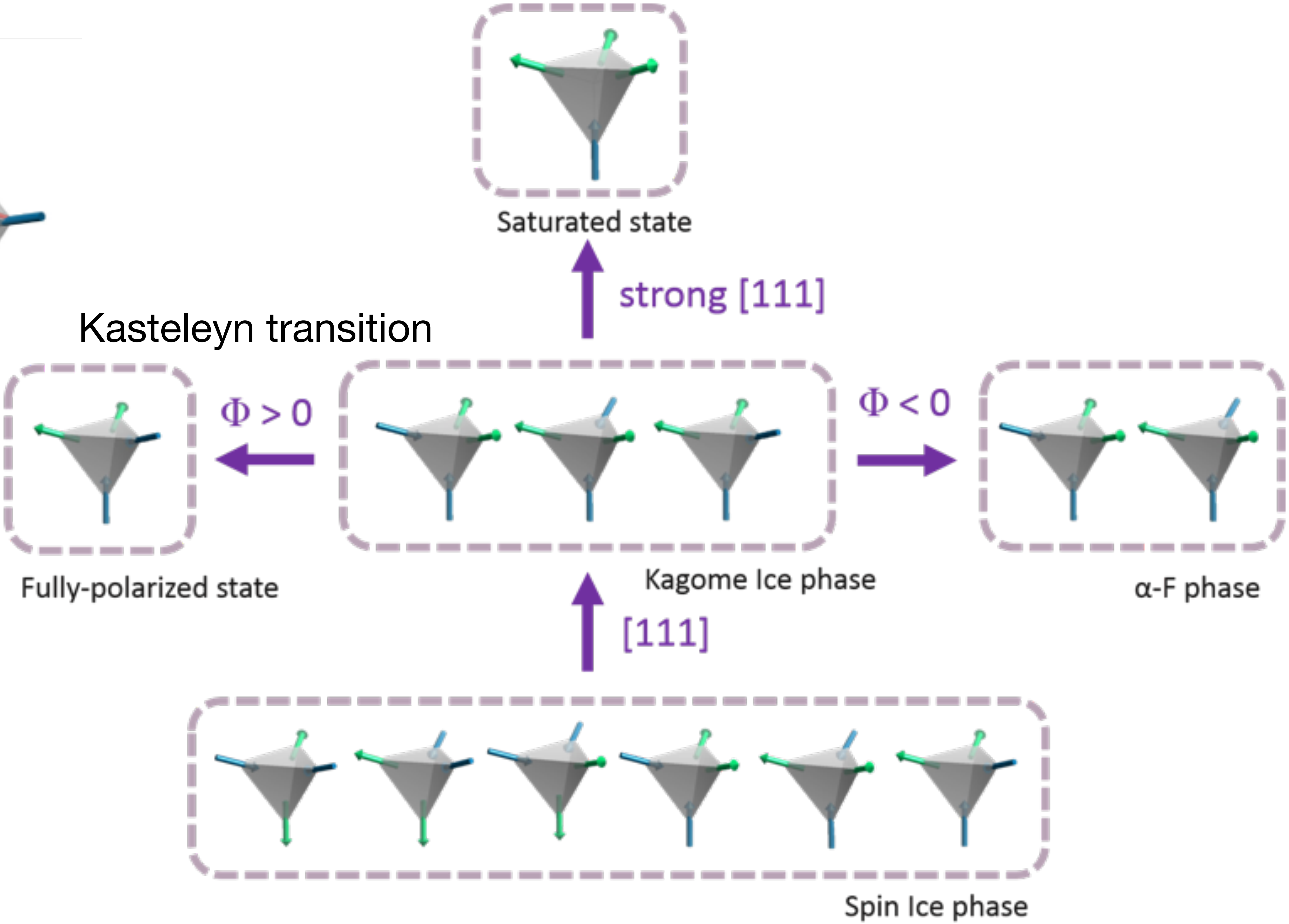


T. Fennell et al., Nature Physics 3, 566 - 572 (2007)
T. Sakakibara, T. Tayama, Z. Hiroi, K. Matsuhira, and S. Takagi, Phys. Rev. Lett. 90, 207205 (2003).
R. Higashinaka, H. Fukazawa, K. Deguchi, and Y. Maeno, J. Phys. Soc. Jpn. 73, 2845 (2004).

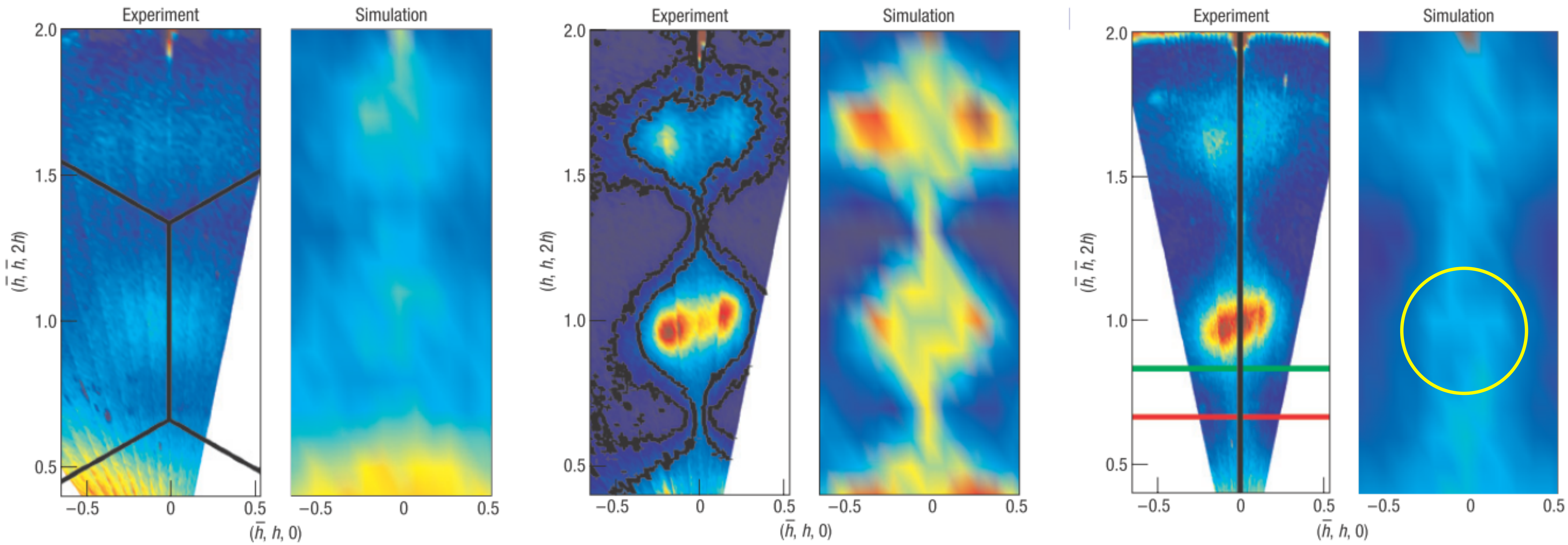
Tilted Kagome Ice



Kasteleyn transition



Anomalous critical scattering



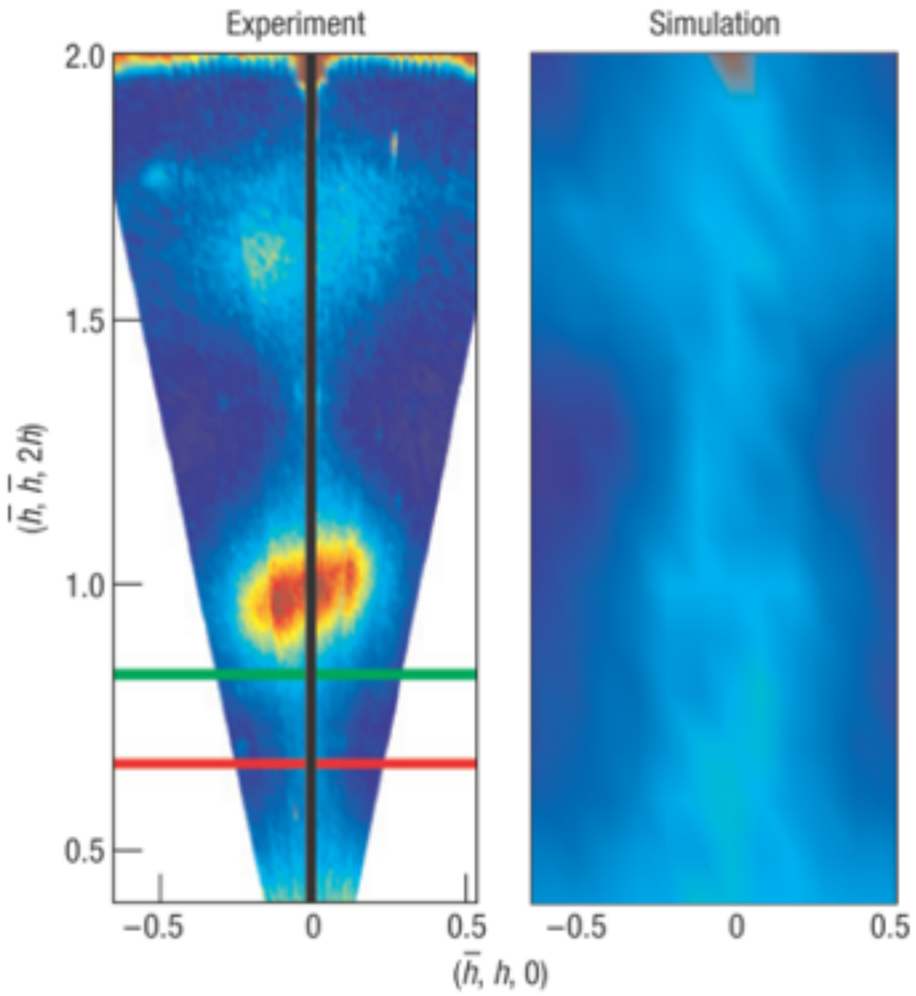
(0 T, 0.5 K) (1.0 T, 0.5 K) (1.6 T, 0.5 K)

- For negative tilted field off [111], critical scattering can not be captured by nearest-neighbor spin ice model

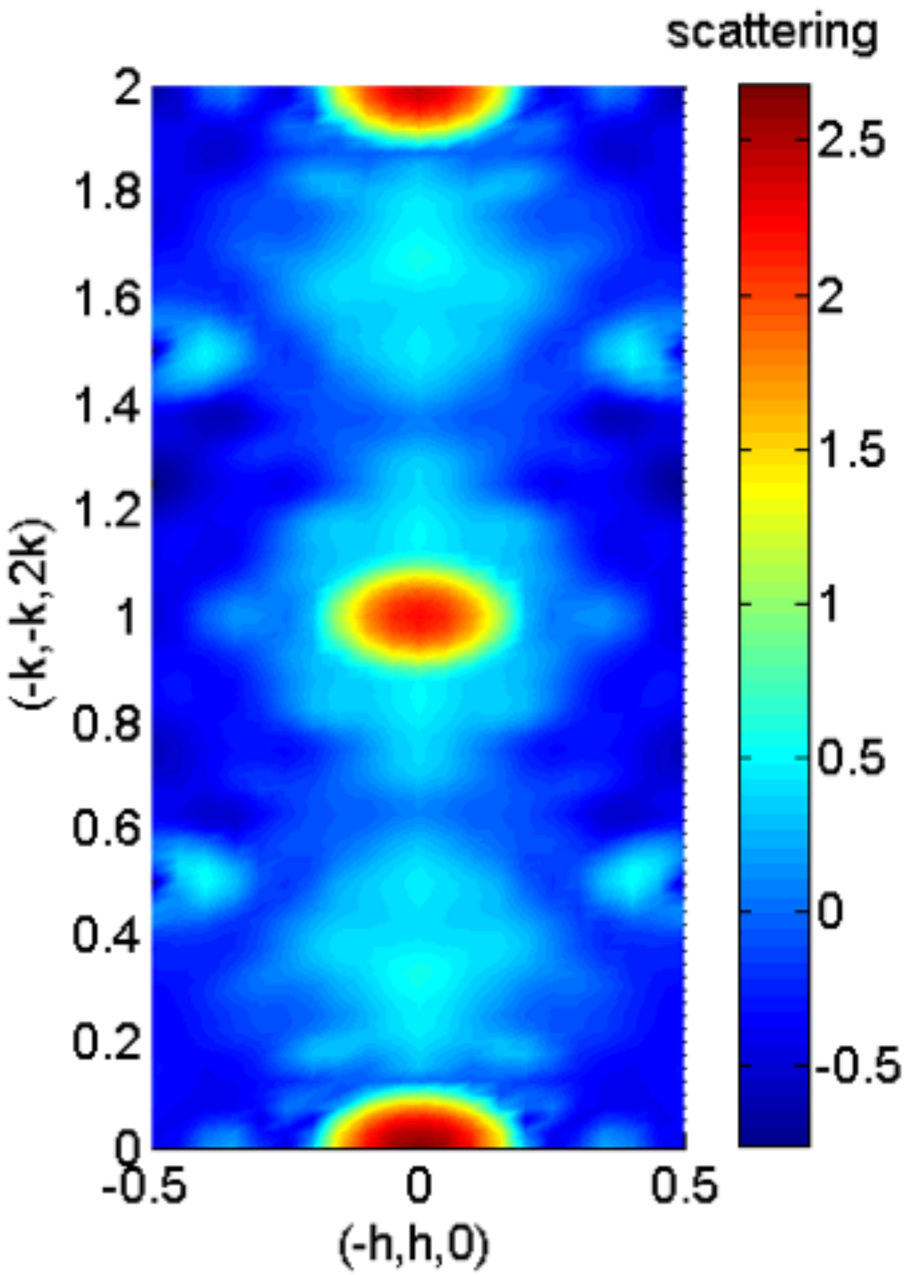
$$H = -J \sum_{<(i,a),(j,b)>} \mathbf{S}_i^a \cdot \mathbf{S}_j^b + D r_{nn}^3 \sum_{i<j,a,b} \left(\frac{\mathbf{S}_i^a \cdot \mathbf{S}_j^b}{|\mathbf{R}_{ij}^{ab}|^3} - \frac{3(\mathbf{S}_i^a \cdot \mathbf{R}_{ij}^{ab})(\mathbf{S}_j^b \cdot \mathbf{R}_{ij}^{ab})}{|\mathbf{R}_{ij}^{ab}|^5} \right) + \mathbf{H} \cdot \sum_i \mathbf{S}_i$$

T. Fennell *et al.*,
Nature Physics **3**, 566 - 572 (2007)

Dipolar Kagome Ice



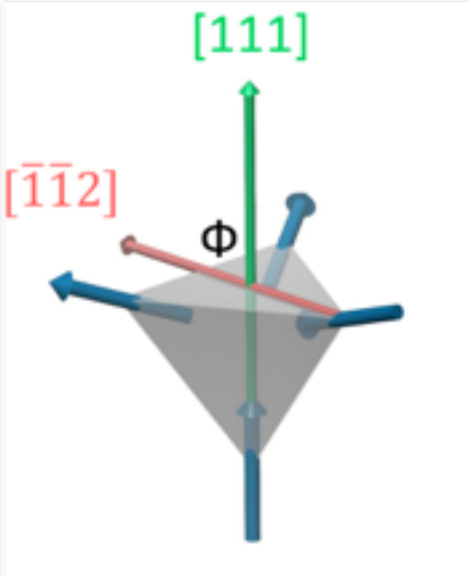
T. Fennell *et al.*,
Nature Physics **3**, 566 - 572 (2007)

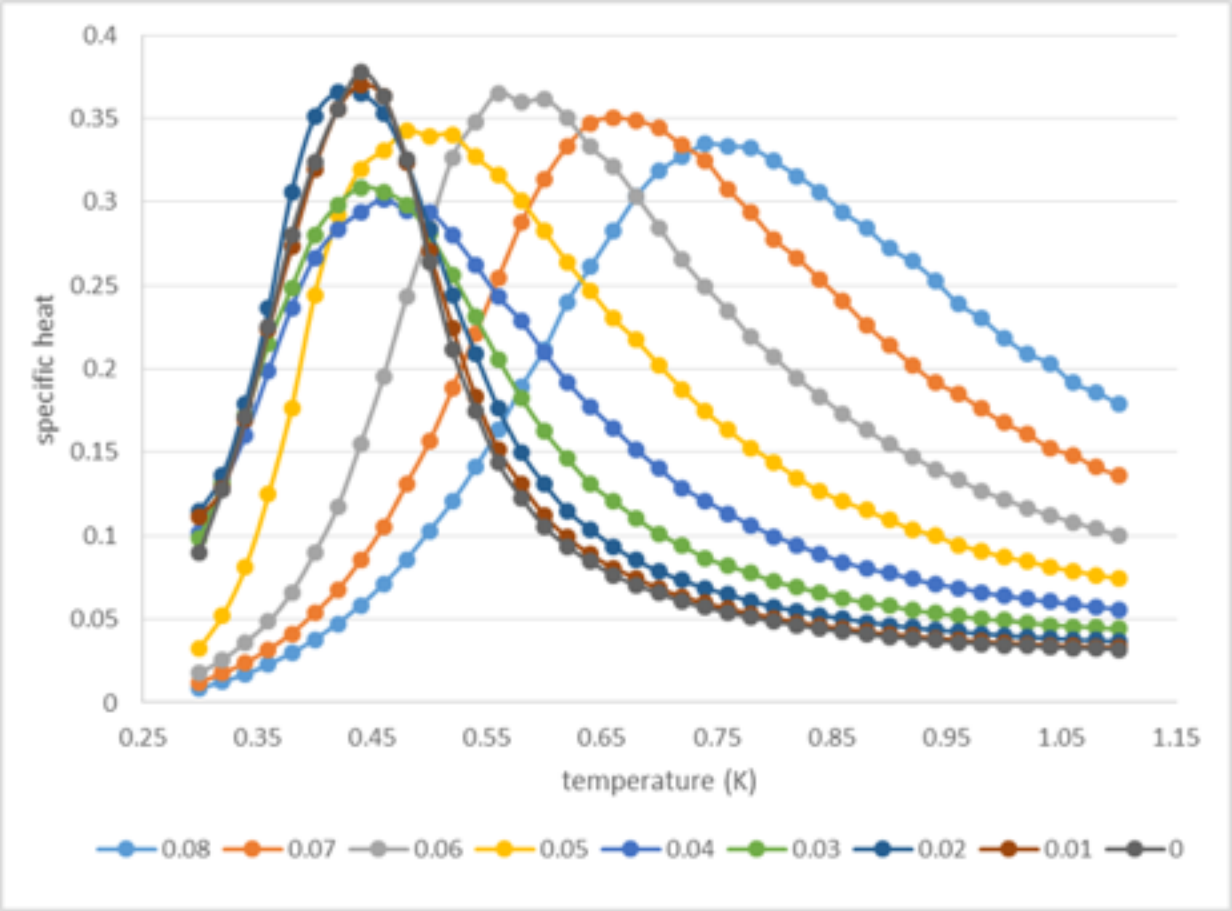


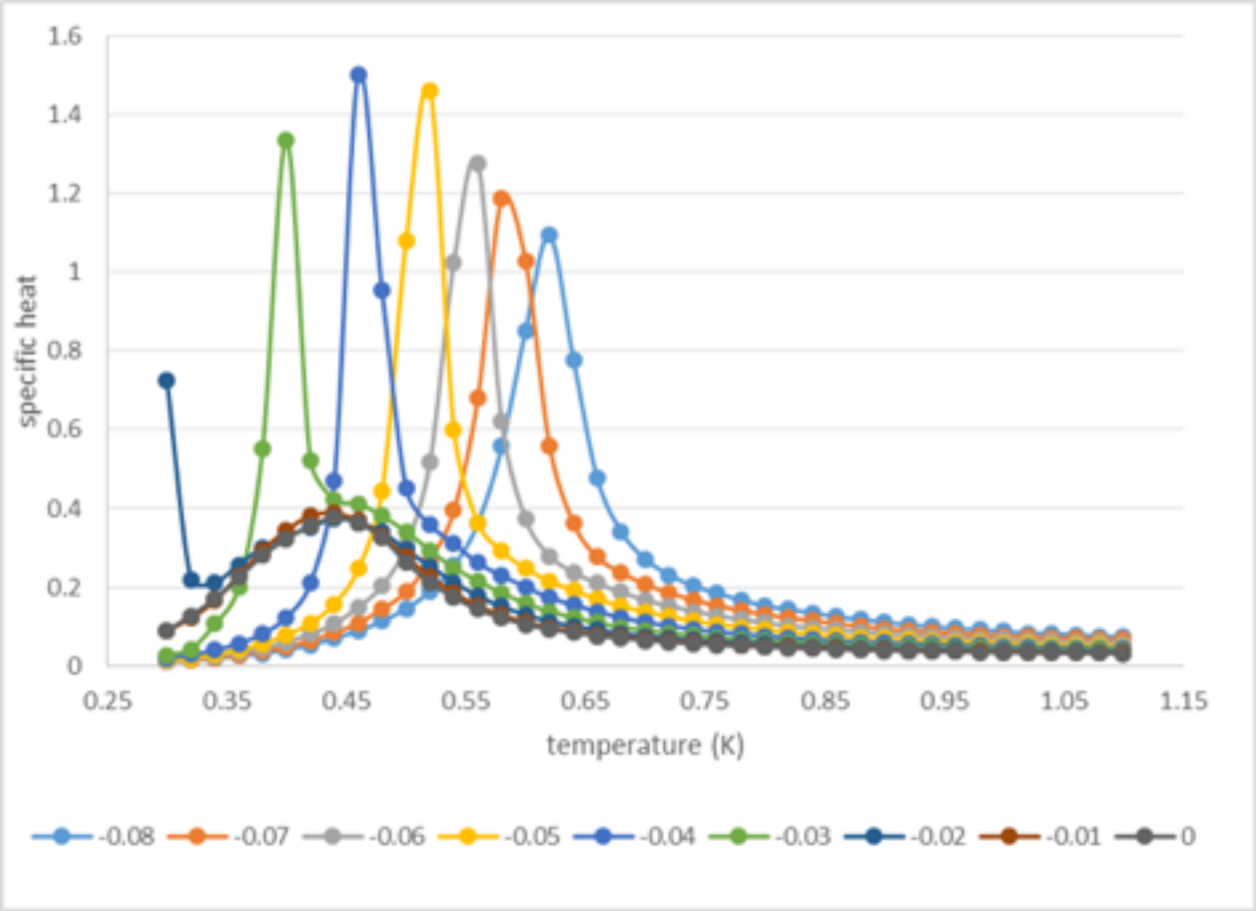
$$H = -J \sum_{<(i,a),(j,b)>} \mathbf{S_i^a} \cdot \mathbf{S_j^b} + D r_{nn}^3 \sum_{i<j,a,b} \left(\frac{\mathbf{S_i^a} \cdot \mathbf{S_j^b}}{|\mathbf{R_{ij}^{ab}}|^3} - \frac{3(\mathbf{S_i^a} \cdot \mathbf{R_{ij}^{ab}})(\mathbf{S_j^b} \cdot \mathbf{R_{ij}^{ab}})}{|\mathbf{R_{ij}^{ab}}|^5} \right) + \mathbf{H} \cdot \sum_i \mathbf{S_i}$$

Specific Heat

- Broad peaks in the positive tilted field
- Sharp peaks in the negative tilted field

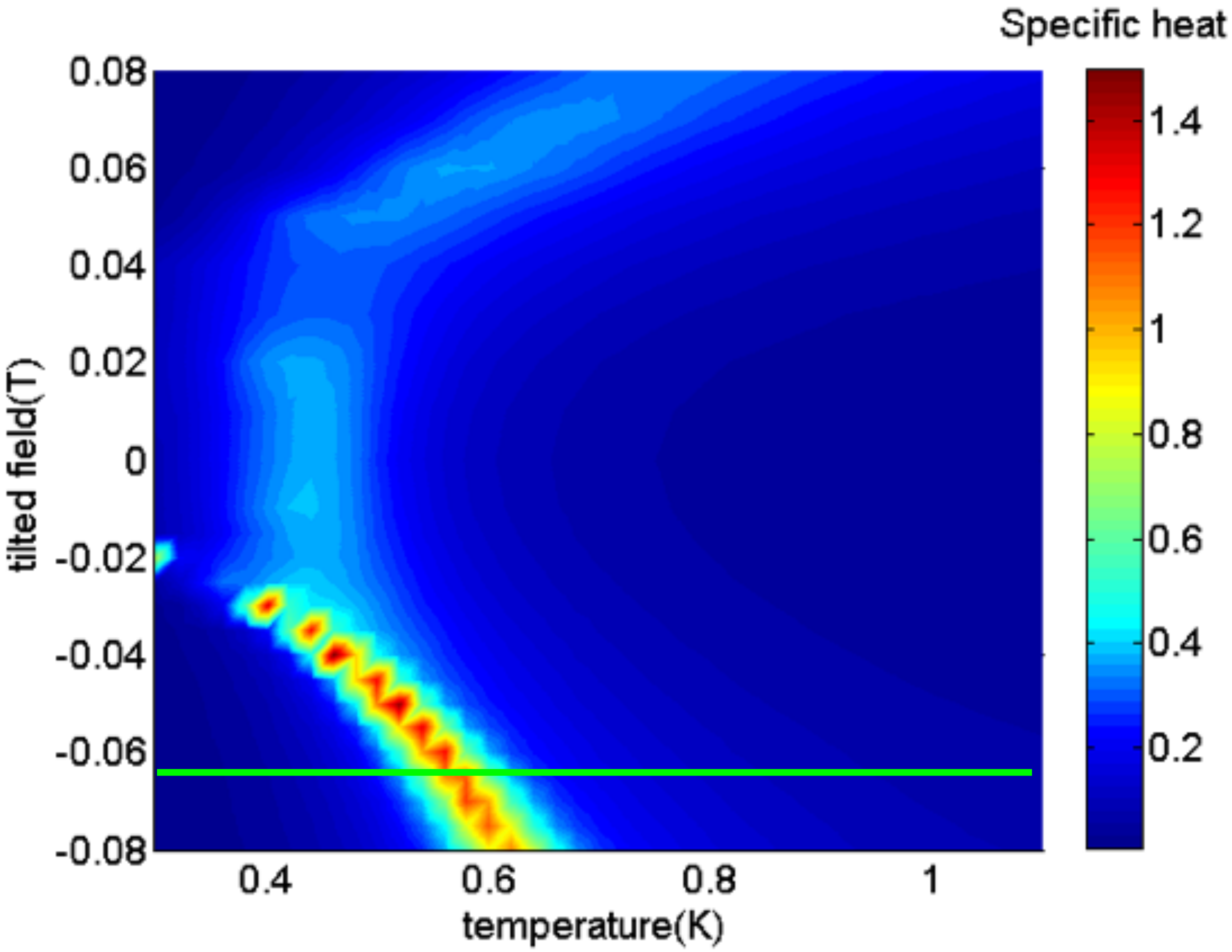


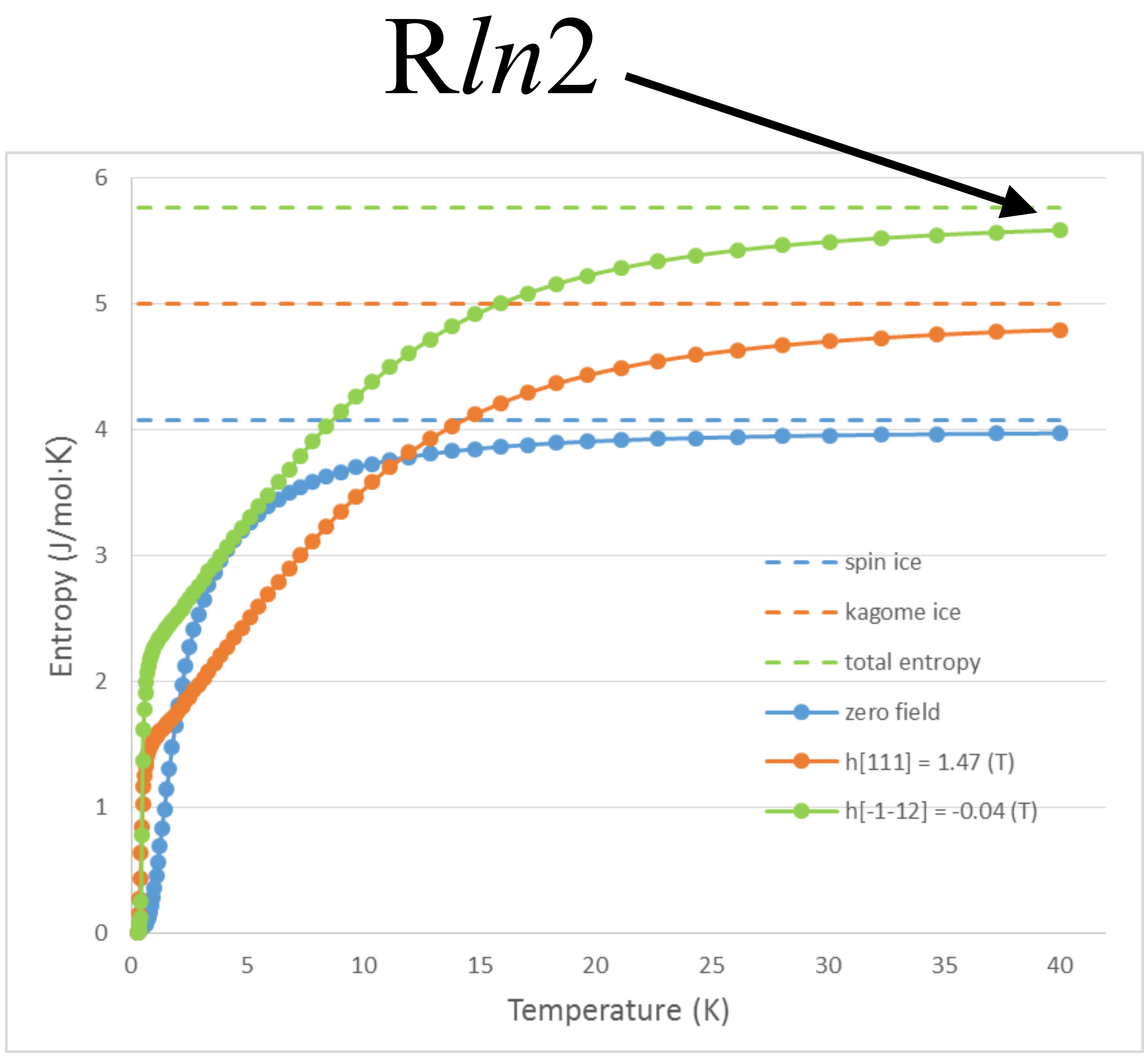




$\Phi>0$ $\Phi<0$

Residual Entropy

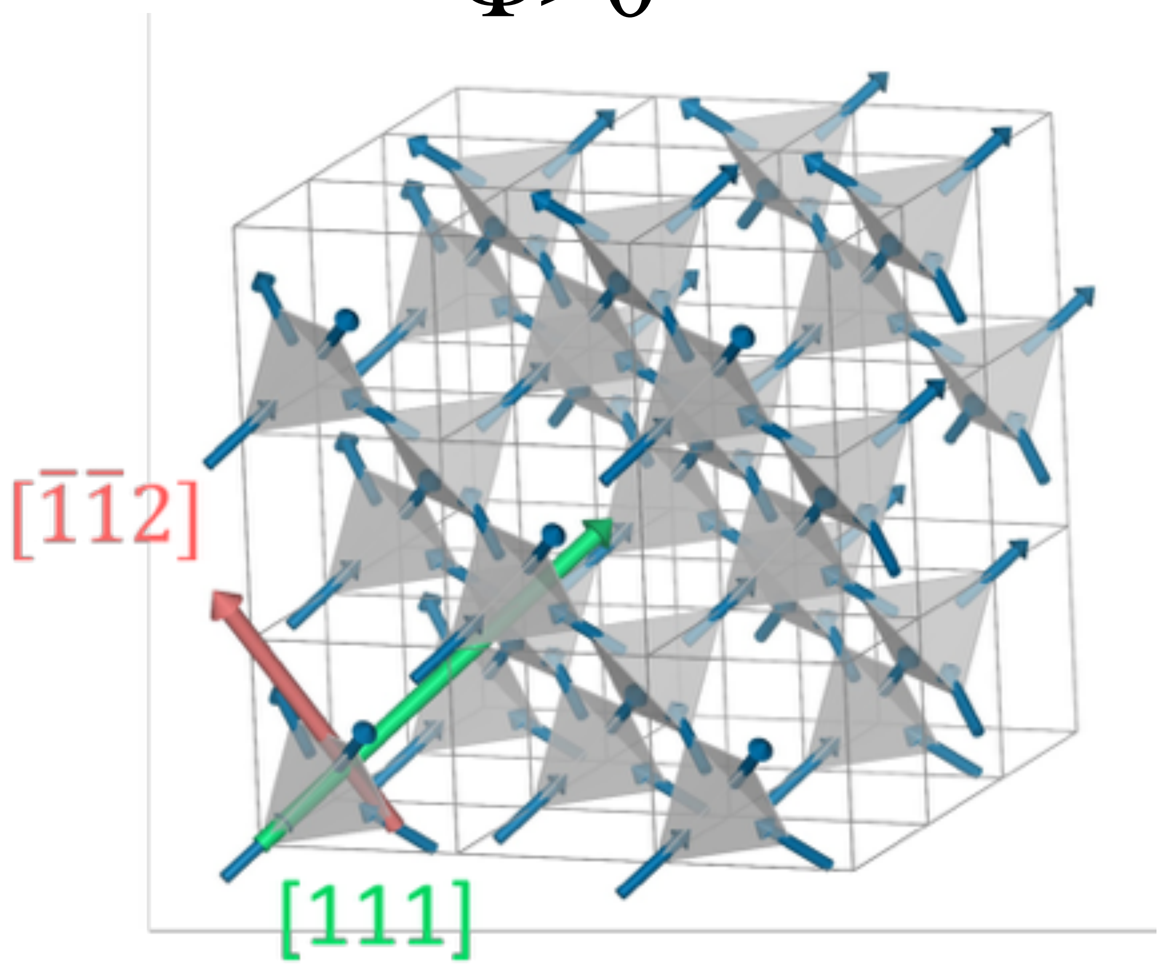




- All the entropy is recovered. Unique ground state.

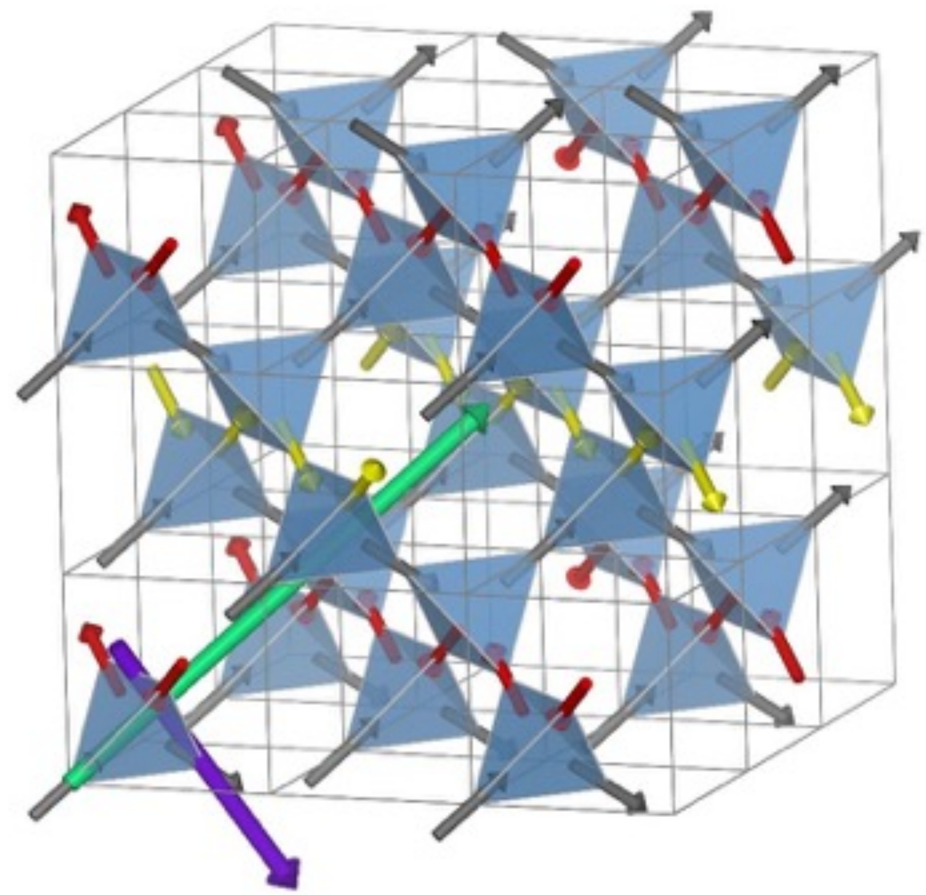
Tilted Dipolar Kagome Ice

$\Phi > 0$



FP state

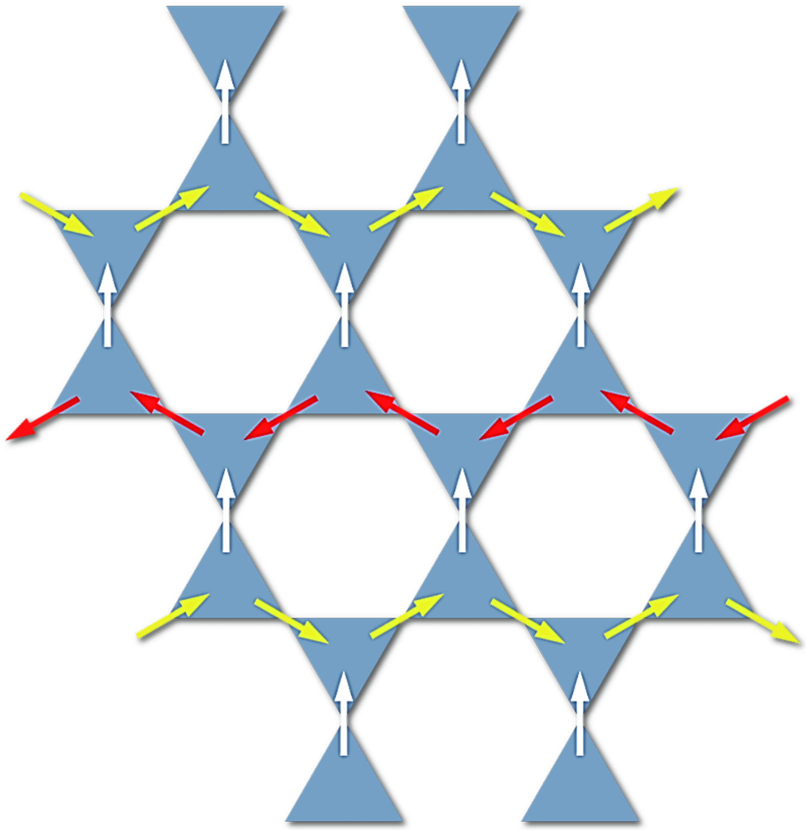
$\Phi < 0$



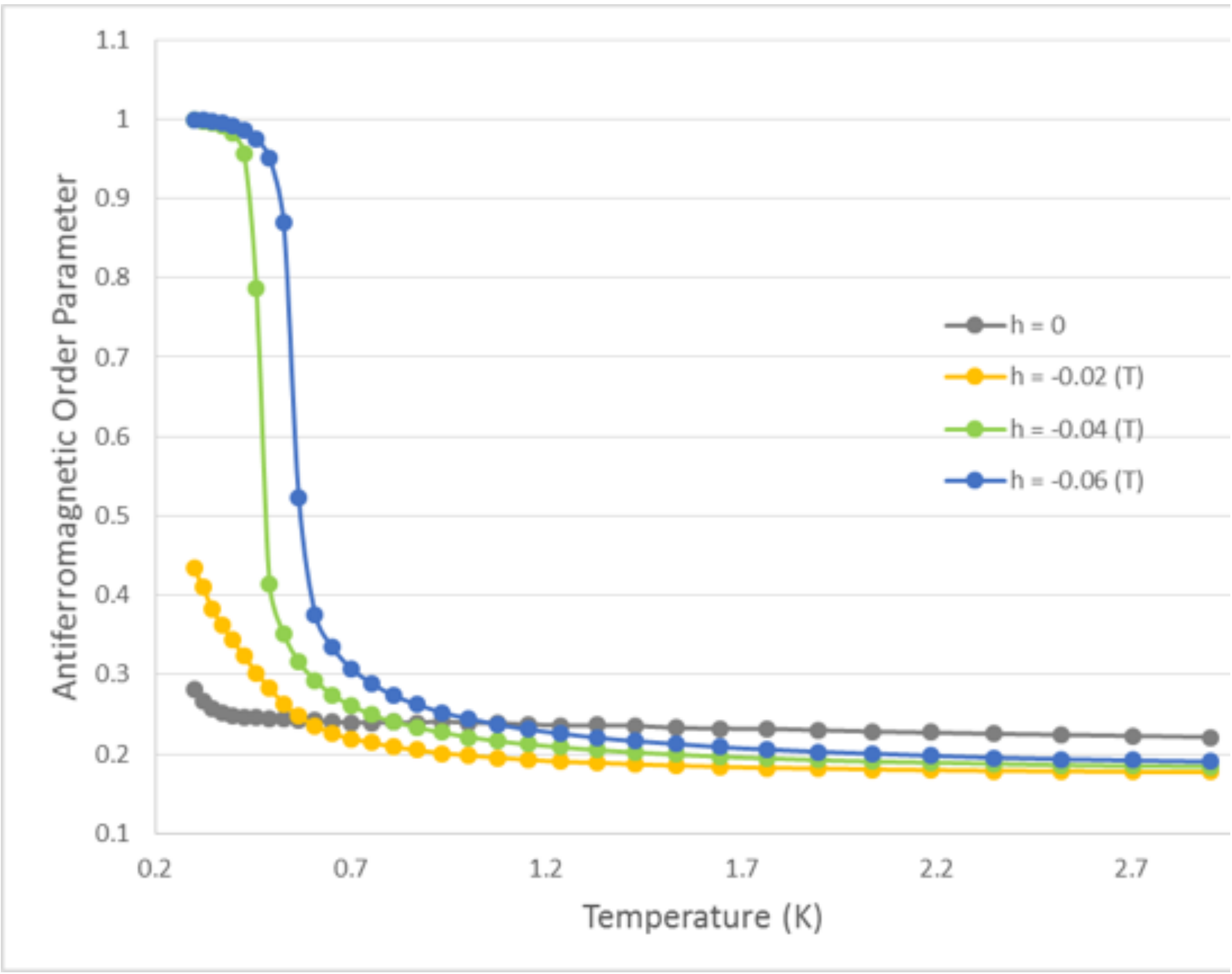
Q = X state

Neel Order on Kagome Plane

First order transition

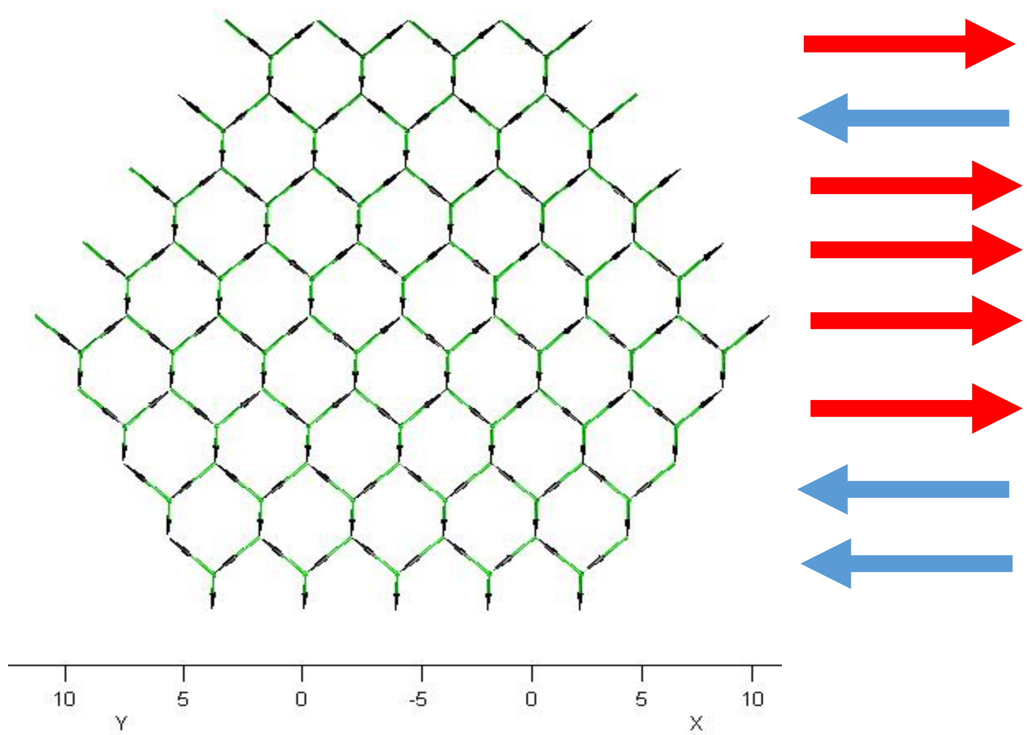


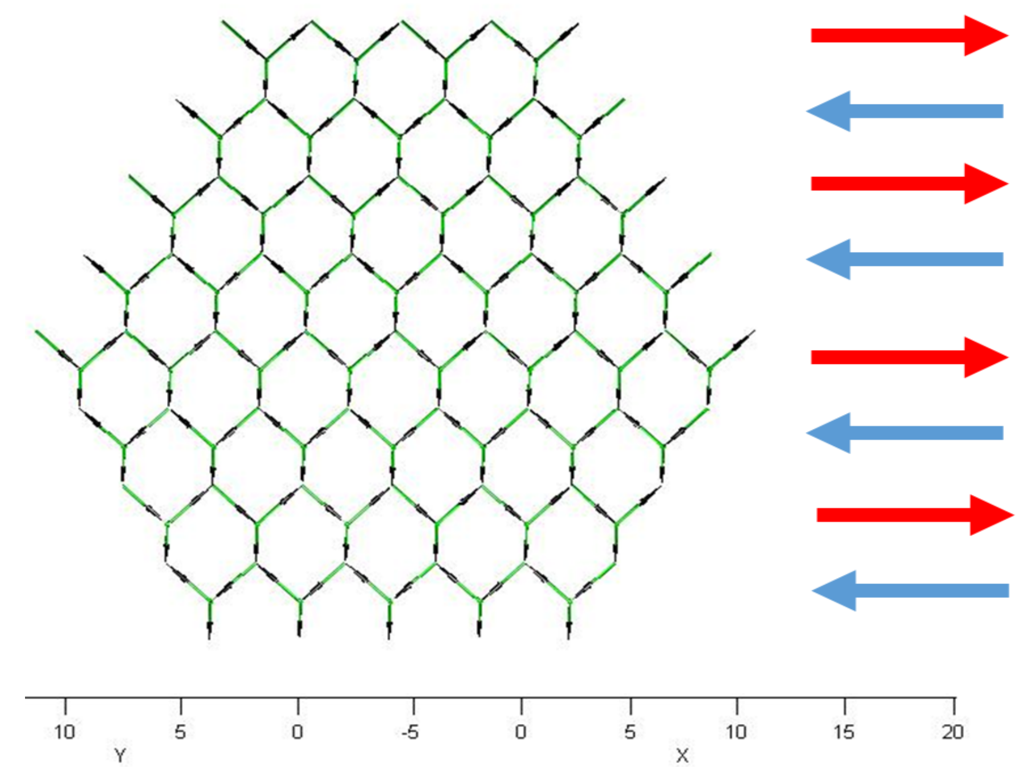
Q = X state



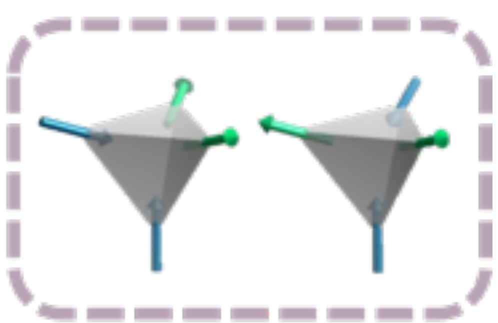
AFM ordering of β chain

Degeneracy Lifting





Dipolar interaction

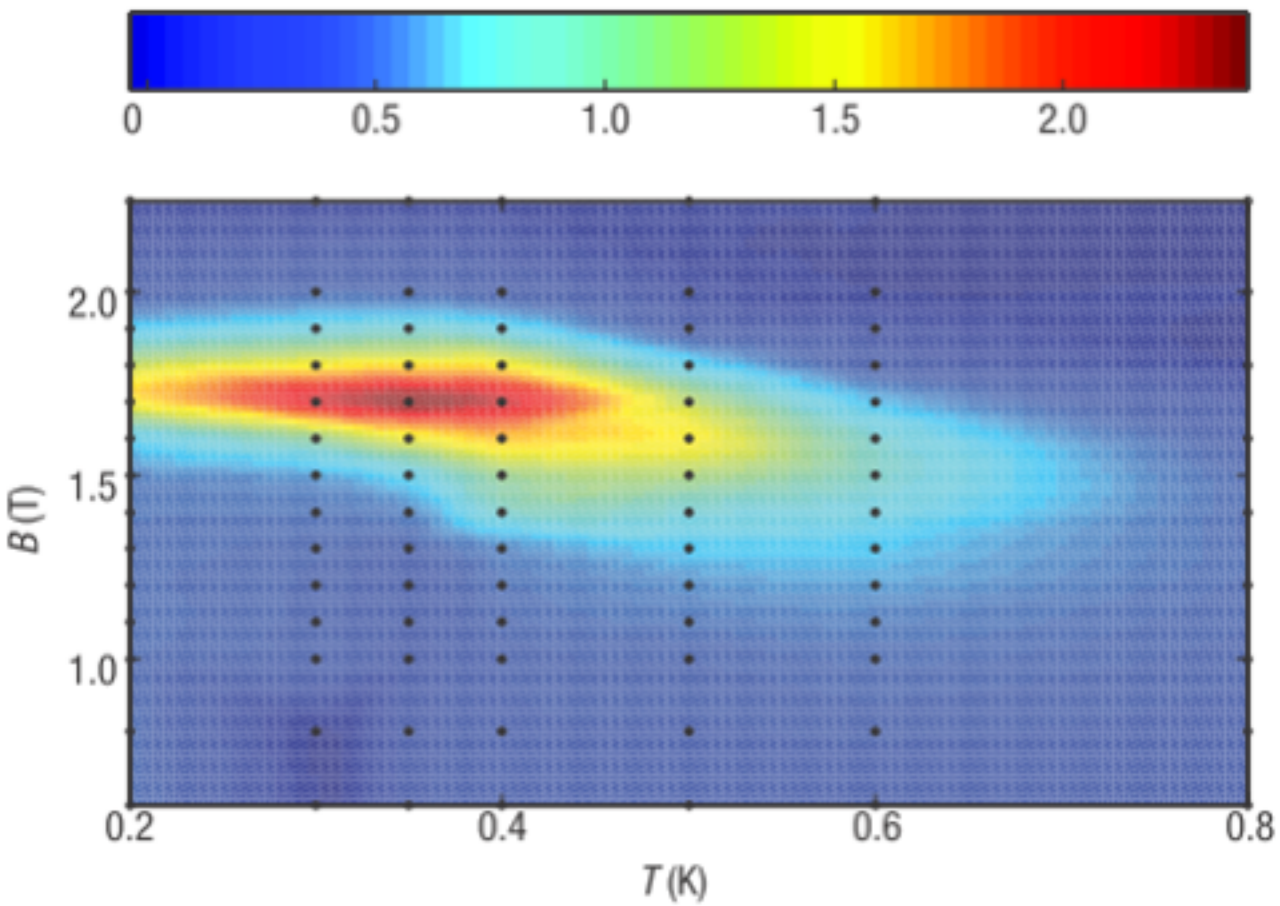


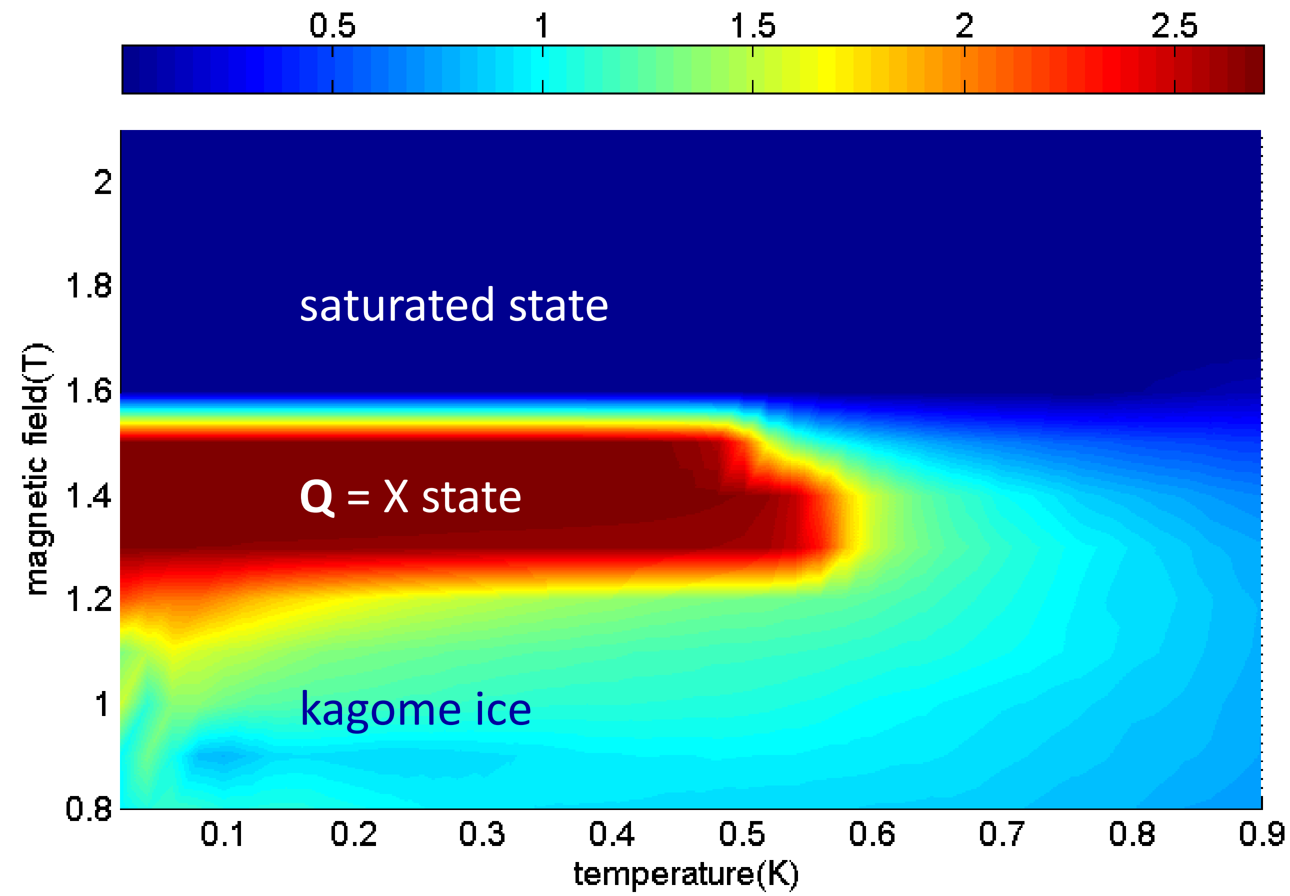
α-F phase

Q = X state

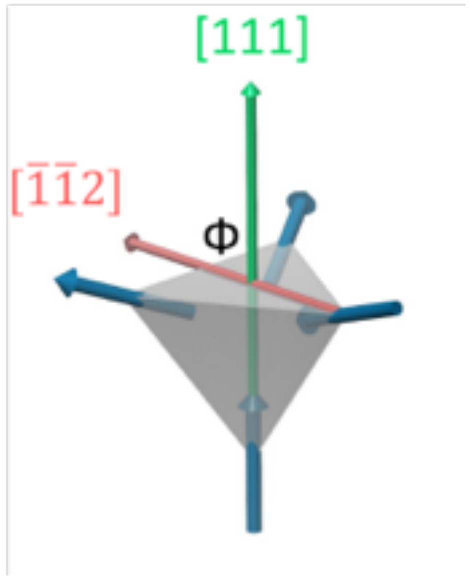
AFM ordering of β chain

Evolution of (-1,-1,2) Intensity





Experiment

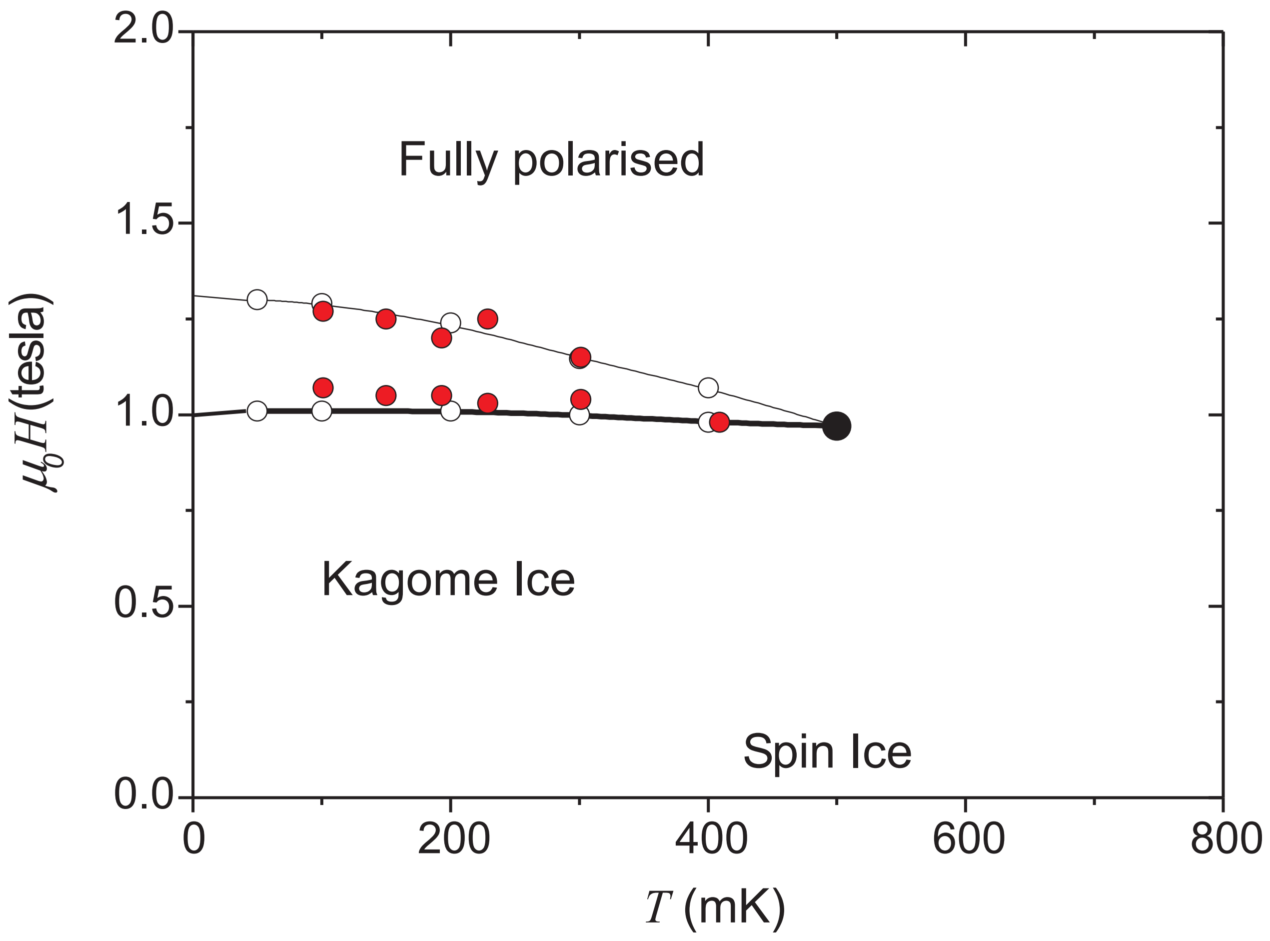


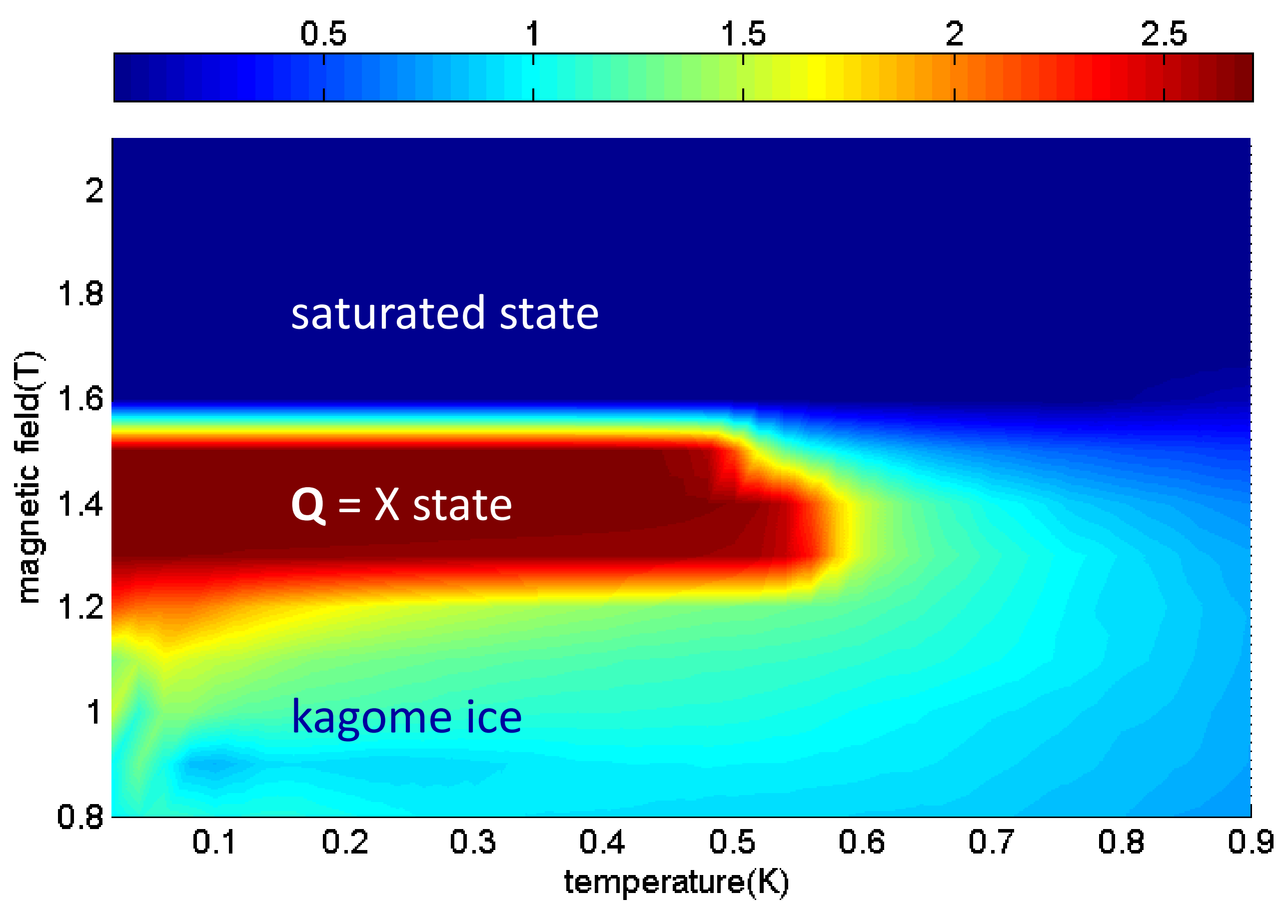
Theory

T. Fennell *et al.*,
Nature Physics **3**, 566 - 572 (2007)

Kao, Holdsworth, and YJK
Phys. Rev. B 93, 180410(R) (2016)

Experiments in $Dy_2Ti_2O_7$





AC susceptibility, Specific heat measurements

S.A. Grigera, et al., Papers in Physics 7, 070009 (2015).

Conclusions and outlooks

- We find a new intermediate half-polarized phase for a dipolar spin ice in a [100] field.
- Half-magnetization plateau is observed in the simulation, yet no clear experimental signatures available.
- AFM ordering transition from kagome ice to Q=X state in the negative tilted field.
- Simulation sees a first-order transition, but the neutron shows critical scattering. Other effects?
- Defect Dynamics? Similar phases in dipolar QSI?

Lin and YJK, Phys. Rev. B 88, 220402(R) (2013)

Kao, Holdsworth, and YJK Phys. Rev. B 93, 180410(R) (2016)