

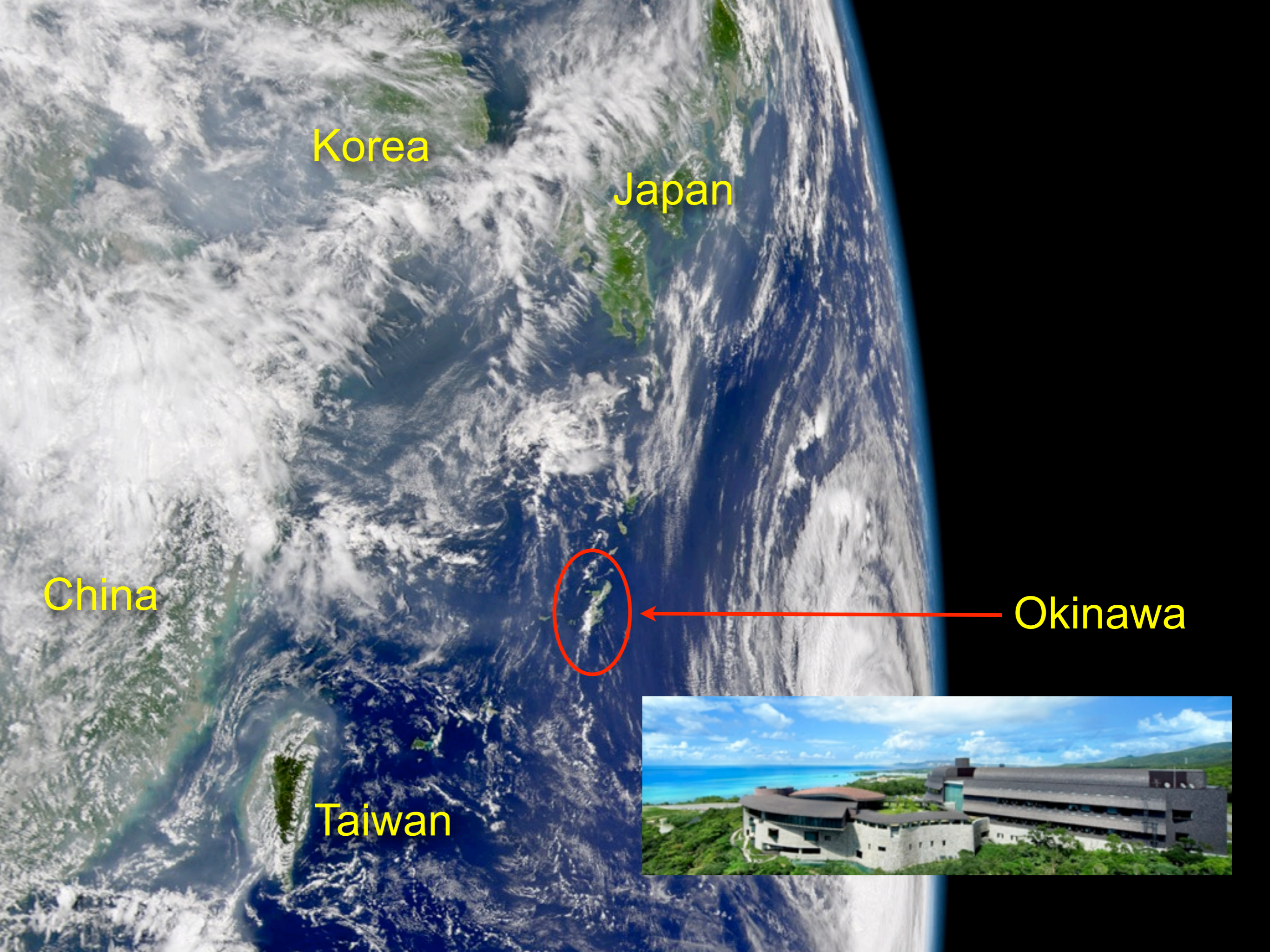
Quantum Ice

nic shannon



OIST

OKINAWA INSTITUTE OF SCIENCE AND TECHNOLOGY GRADUATE UNIVERSITY



Korea

Japan

China

Okinawa

Taiwan

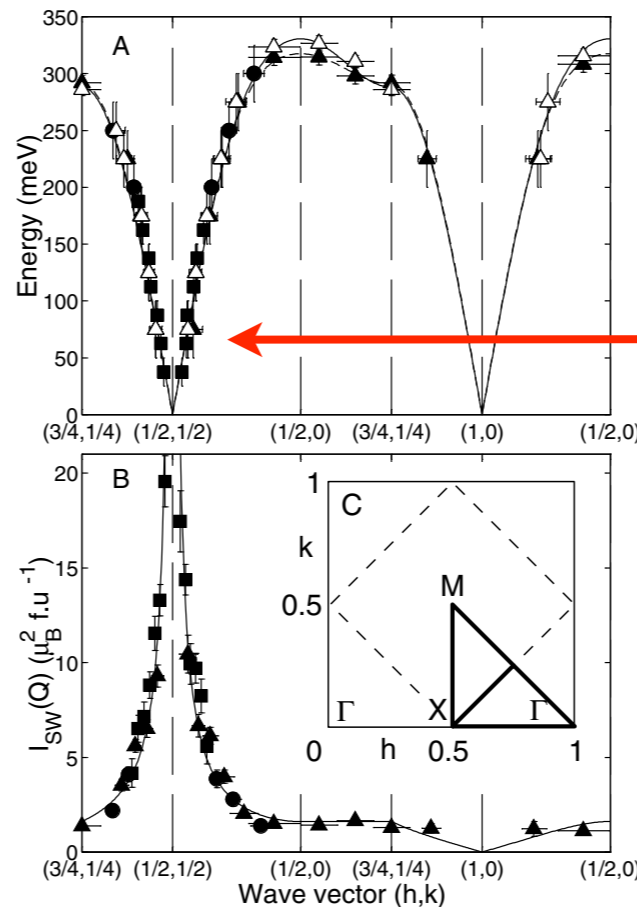
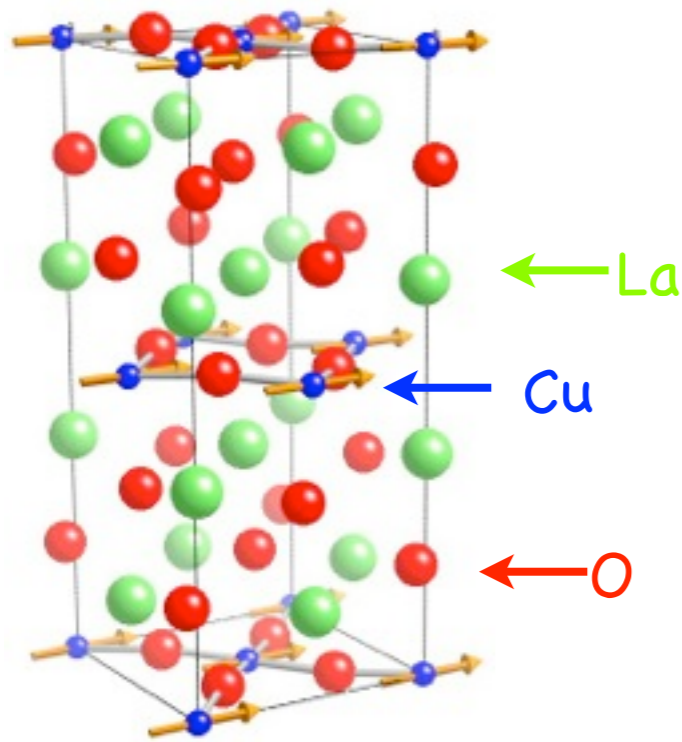


spin liquids are boring...



...because there's nothing to measure

La₂CuO₄ - everybody's favourite Néel AF :



spin wave excitation
dispersing out of
ordering vector

R. Coldea *et al.*,
Phys. Rev. Lett. **86**, 5377 (2001).

spin wave excitations which control all properties of La₂CuO₄ for $T < T_N = 250$ K
follow directly from broken spin-rotation symmetry...



...but not everything works like this

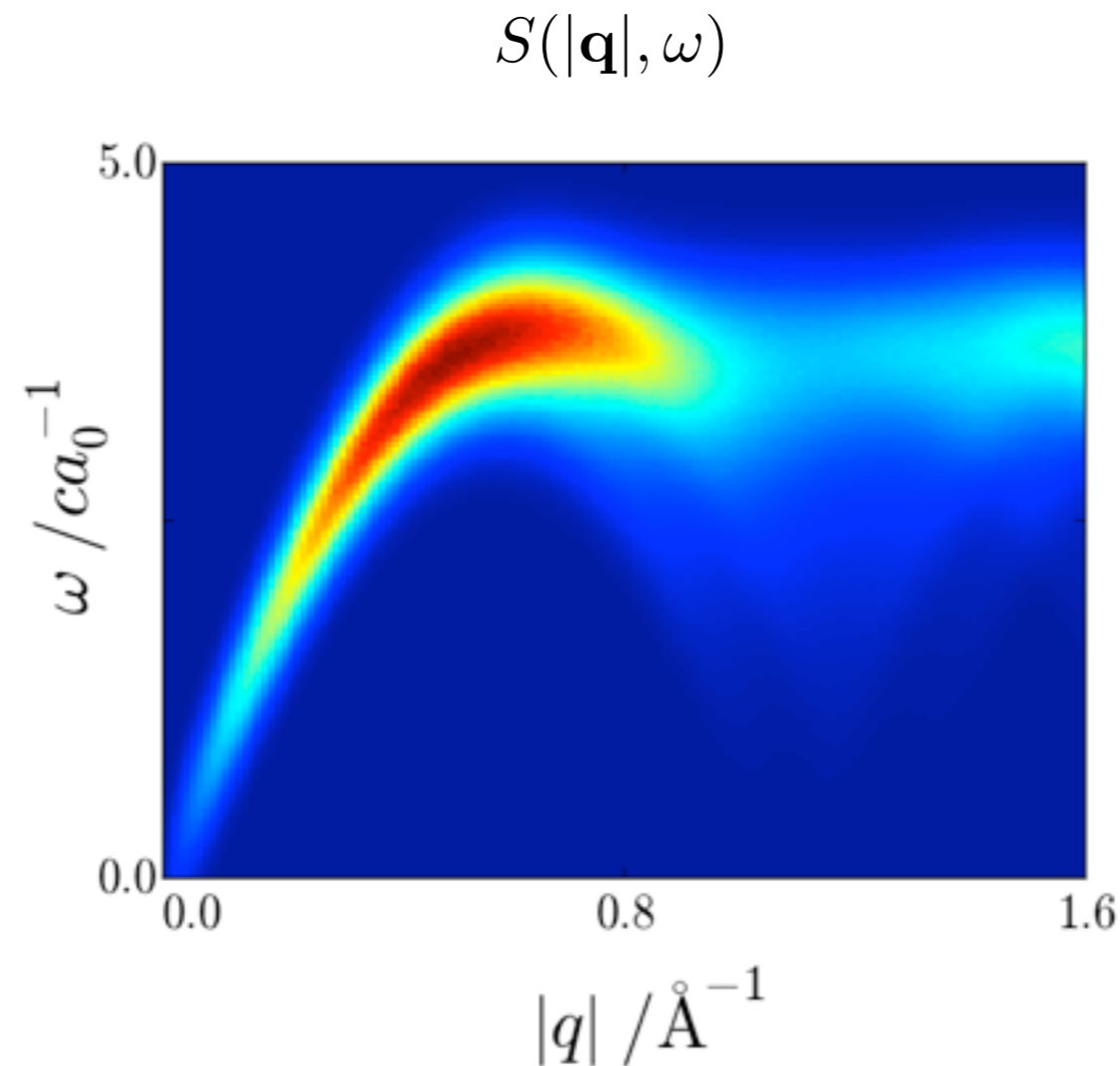


just because there's no
broken symmetry...



...doesn't mean
there's no physical effect!

a quantum spin liquid with artificial light ?



...prediction for inelastic neutron scattering taken from microscopic model of a quantum spin ice



wouldn't have happened without



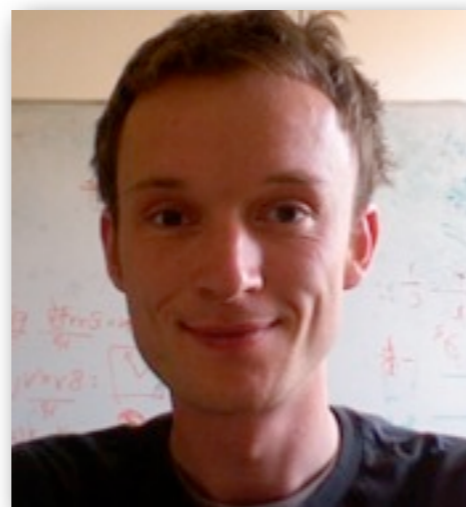
Karlo Penc

Budapest



Peter Fulde

APCTP



Frank Pollmann

MPI-PKS



Olga Sikora

NTU



Owen Benton

Bristol/OIST



MAX-PLANCK-GESELLSCHAFT

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Engineering and Physical Sciences
Research Council



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THE JOURNAL OF CHEMICAL PHYSICS

VOLUME 1

AUGUST, 1933

NUMBER 8

A Theory of Water and Ionic Solution, with Particular Reference to Hydrogen and Hydroxyl Ions

J. D. BERNAL AND R. H. FOWLER, *University of Cambridge, England*

(Received April 29, 1933)

SHORT SUMMARY

ON the basis of the model of the water molecule derived from spectral and x-ray data and a proposed internal structure for water, the following properties of water and ionic solutions have been deduced quantitatively in good agreement with experiment.

- (1) The crystal structure of ice.
- (2) The x-ray diffraction curve for water.
- (3) The total energy of water and ice.
- (4) The degree of hydration of positive and negative ions in water.

ice, and of the density of water, leads to the proposing of an irregular four-coordinated structure for water. This structure is tested and found to account for the positions of the maxima of x-ray diffraction by water, and for the change in these positions with temperature, which are quite different from those of a simple liquid such as mercury. Three different intermolecular arrangements are postulated for water at different temperatures.

(1) Ice-tridymite-like (four-coordinated) at low temperatures below 4°C.

how does water ice work ?

water molecules link up through hydrogen bonds...

...to form a tetrahedrally co-ordinated lattice :

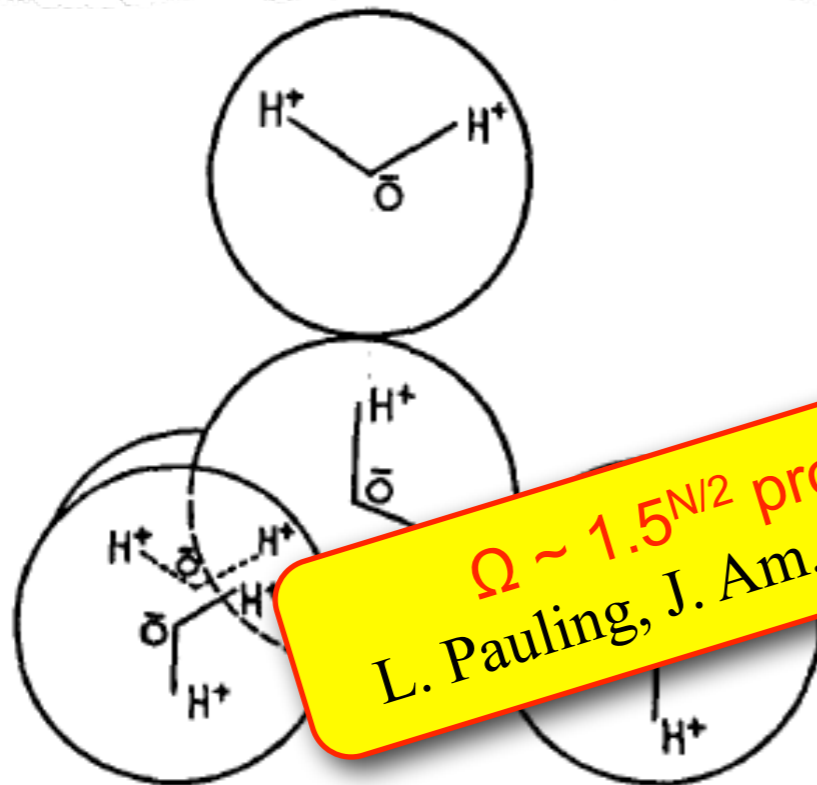
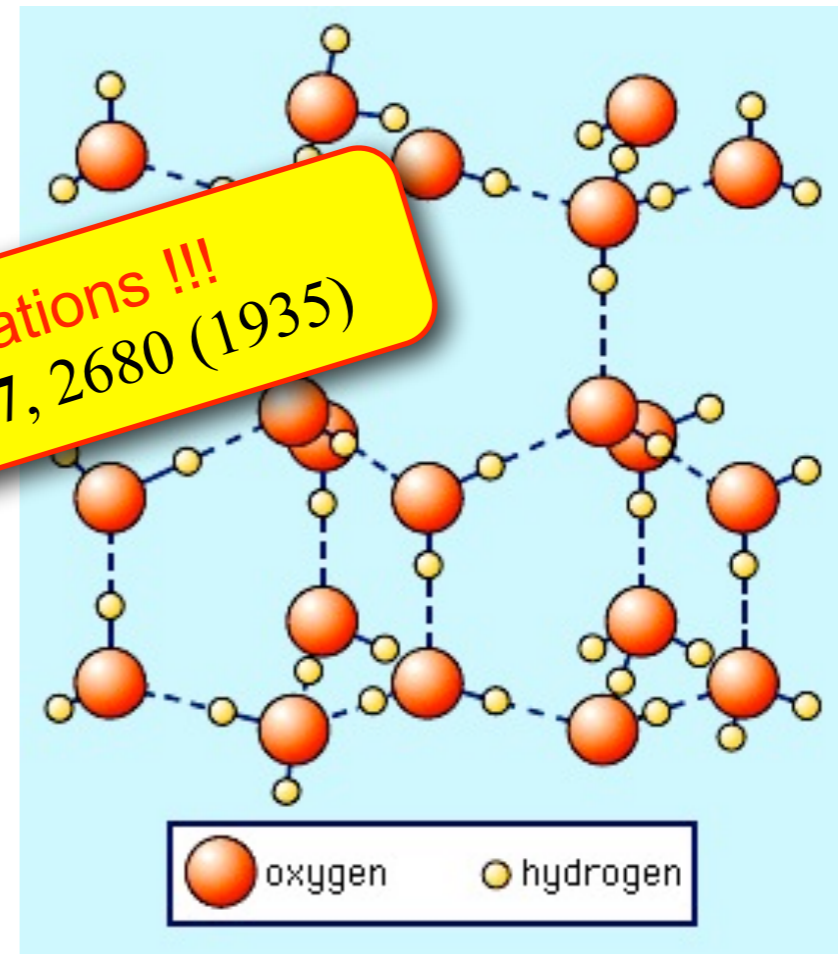


FIG. 4. Tetrahedral coordination of water molecules. The four molecules surrounding one water molecule are shown. Of these, two are in the plane of the paper, one above and one below it.

$\Omega \sim 1.5^{N/2}$ proton configurations !!!
L. Pauling, J. Am. Chem. Soc. 27, 2680 (1935)



J. D. Bernal and R. H. Fowler, J. Chem. Phys. 1, 515 (1933).

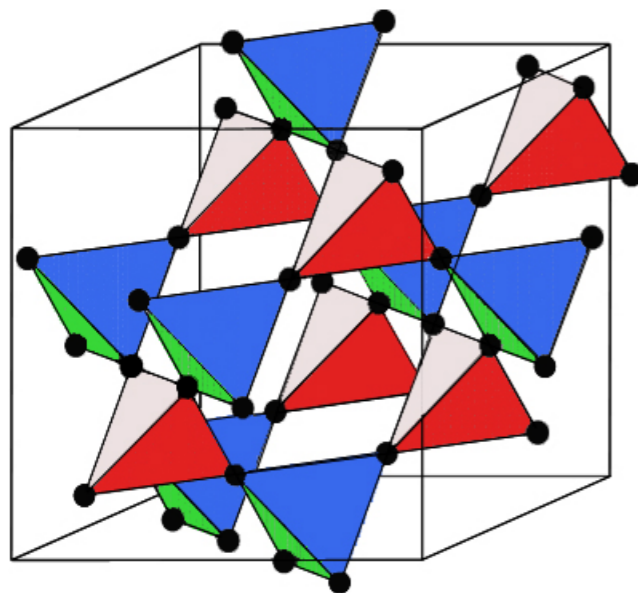


the fashionable form of ice...

$\text{Ho}_2\text{Ti}_2\text{O}_7$
 $\text{Dy}_2\text{Ti}_2\text{O}_7$

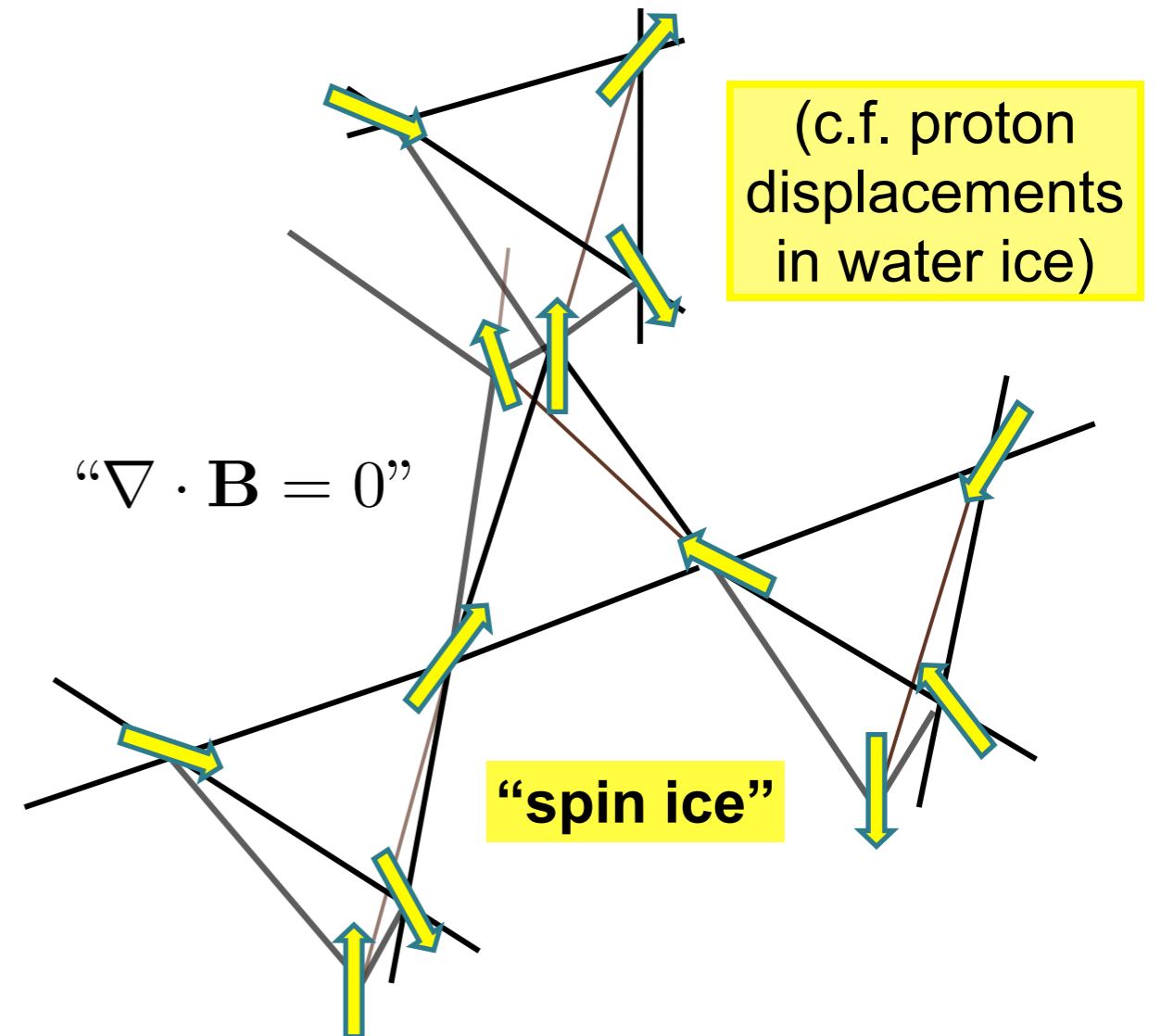


ferromagnetic interactions select states with two in and two out spins per tetrahedron



magnetic
 Ho^{8+} or
 Dy^{8+} ions
live on a
pyrochlore
lattice

strong easy axis anisotropy forces
spins to point in or out of tetrahedron



M.J. Harris et al., Phys. Rev. Lett. **79**, 2554 (1997)
A.P. Ramirez et al., Nature **399**, 333 (1999)



spin ice and its monopoles...



...discussed in all the most reputable sources of scientific information !



Rods of Neutron Scattering Intensity in $\text{Yb}_2\text{Ti}_2\text{O}_7$: Compelling Evidence for Significant Anisotropic Exchange in a Magnetic Pyrochlore Oxide

Jordan D. Thompson,¹ Paul A. McClarty,¹ Henrik M. Rønnow,² Louis P. Regnault,³ Andreas Sorge,^{4,5} and Michel J. P. Gingras^{1,5,6}

Quantum Excitations in Quantum Spin Ice

Kate A. Ross,¹ Lucie Savary,² Bruce D. Gaulin,^{1,3,4} and Leon Balents^{5,*}

ARTICLE

Received 27 Jan 2012 | Accepted 5 Jul 2012 | Published 7 Aug 2012

DOI: 10.1038/ncomms1989

Higgs transition from a magnetic Coulomb liquid to a ferromagnet in $\text{Yb}_2\text{Ti}_2\text{O}_7$

Lieh-Jeng Chang^{1,2}, Shigeki Onoda³, Yixi Su⁴, Ying-Jer Kao⁵, Ku-Ding Tsuei⁶, Yukio Yasui^{7,8}, Kazuhisa Kakurai² & Martin Richard Lees⁹

Vindication of $\text{Yb}_2\text{Ti}_2\text{O}_7$ as a Model Exchange Quantum Spin Ice

R. Applegate,¹ N. R. Hayre,¹ R. R. P. Singh,¹ T. Lin,² A. G. R. Day,^{2,3} and M. J. P. Gingras^{1,2,4}

Power-Law Spin Correlations in the Pyrochlore Antiferromagnet $\text{Tb}_2\text{Ti}_2\text{O}_7$

T. Fennell,^{1,*} M. Kenzelmann,² B. Roessli,¹ M. K. Haas,³ and R. J. Cava³



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spin ice goes quantum...

Large Quantum Spin Ice

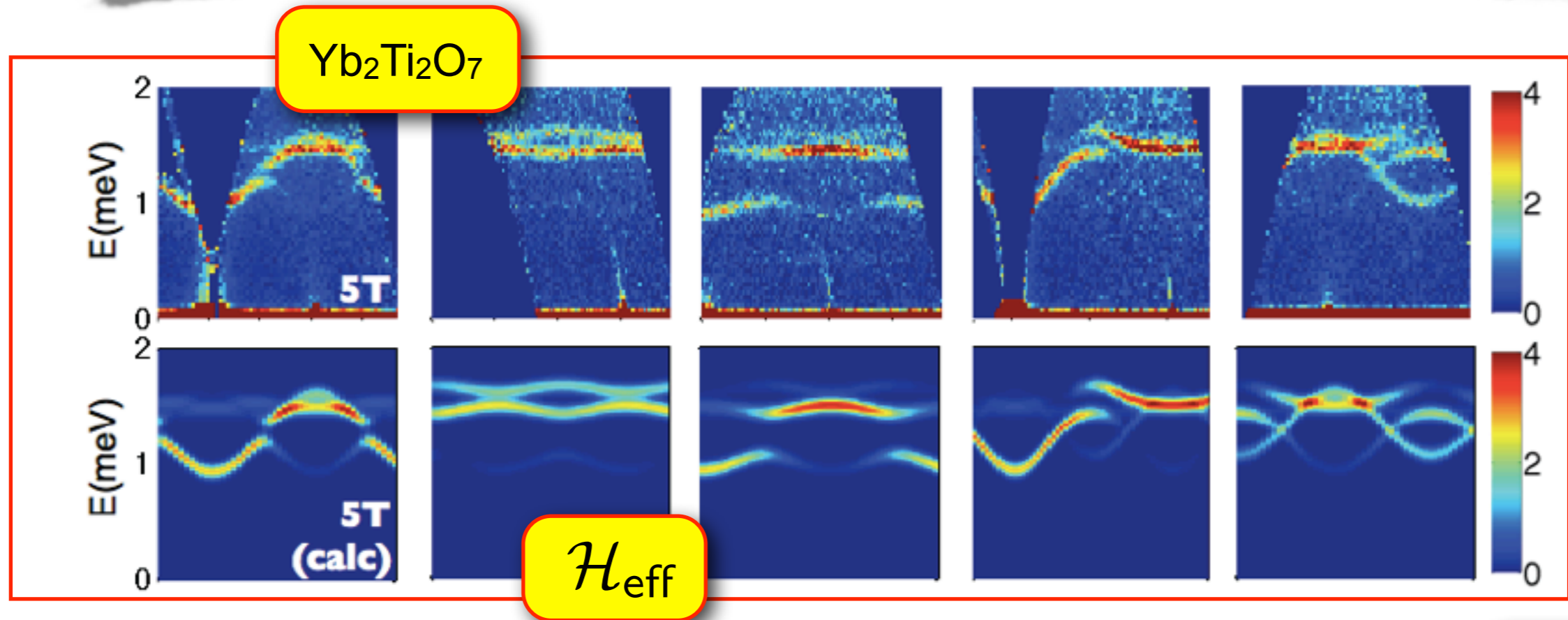
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Quantum Excitations in Quantum Spin Ice

 Kate A. Ross,¹ Lucile Savary,² Bruce D. Gaulin,^{1,3,4} and Leon Balents^{5,*}


$$\mathcal{H}_{\text{eff}} = \sum_{\langle ij \rangle} \left\{ J_{zz} S_i^z S_j^z - J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+) \right. \\ \left. + J_{\pm\pm} [\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^-] \right. \\ \left. + J_{z\pm} [S_i^z (\zeta_{ij} S_j^+ + \zeta_{ij}^* S_j^-) + i \leftrightarrow j] \right\}$$

 fit to spin wave spectrum
in saturated state

$$J_{zz} = 0.17 \pm 0.04 \text{ meV}$$

$$J_{\pm} = 0.05 \pm 0.01 \text{ meV}$$

$$J_{z\pm} = 0.14 \pm 0.01 \text{ meV}$$

$$J_{\pm\pm} = 0.05 \pm 0.01 \text{ meV}$$

 Ising term
⇒ ice

 transverse
term
⇒ dynamics

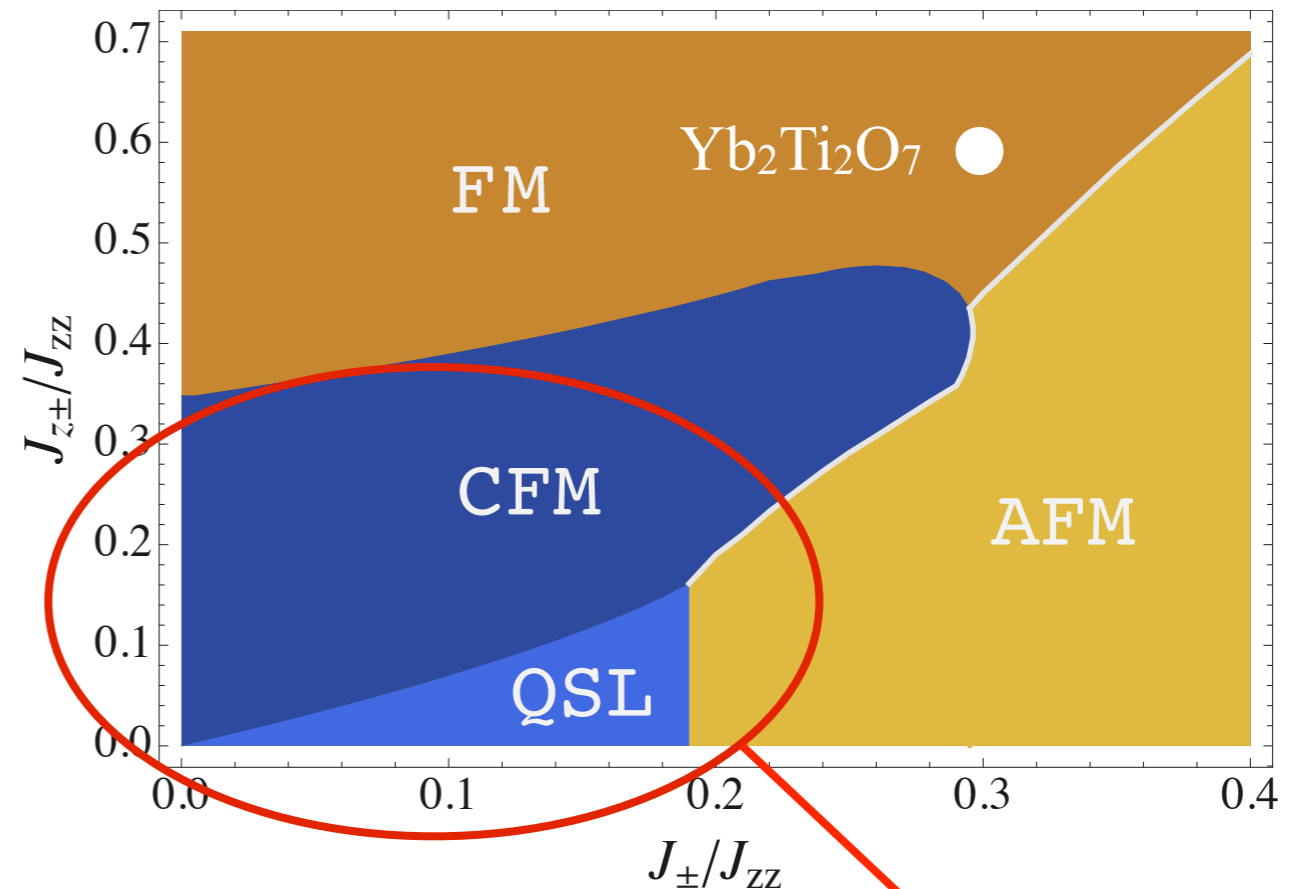

Coulombic Quantum Liquids in Spin-1/2 Pyrochlores

Lucile Savary^{1,2} and Leon Balents³

most general nearest-neighbour
exchange Hamiltonian :

$$H = \sum_{\langle ij \rangle} \left[J_{zz} S_i^z S_j^z - J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+) \right. \\ \left. + J_{\pm\pm} [\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^-] \right. \\ \left. + J_{z\pm} [S_i^z (\zeta_{ij} S_j^+ + \zeta_{ij}^* S_j^-) + i \leftrightarrow j] \right],$$

phase diagram within
Gauge mean field theory :



quantum spin
liquids derived from
spin ice

N.B. see also :
S. Onoda *et al.*, Phys. Rev. B **83**, 094411 (2011)
S-B. Lee *et al.*, arXiv:1204.2262



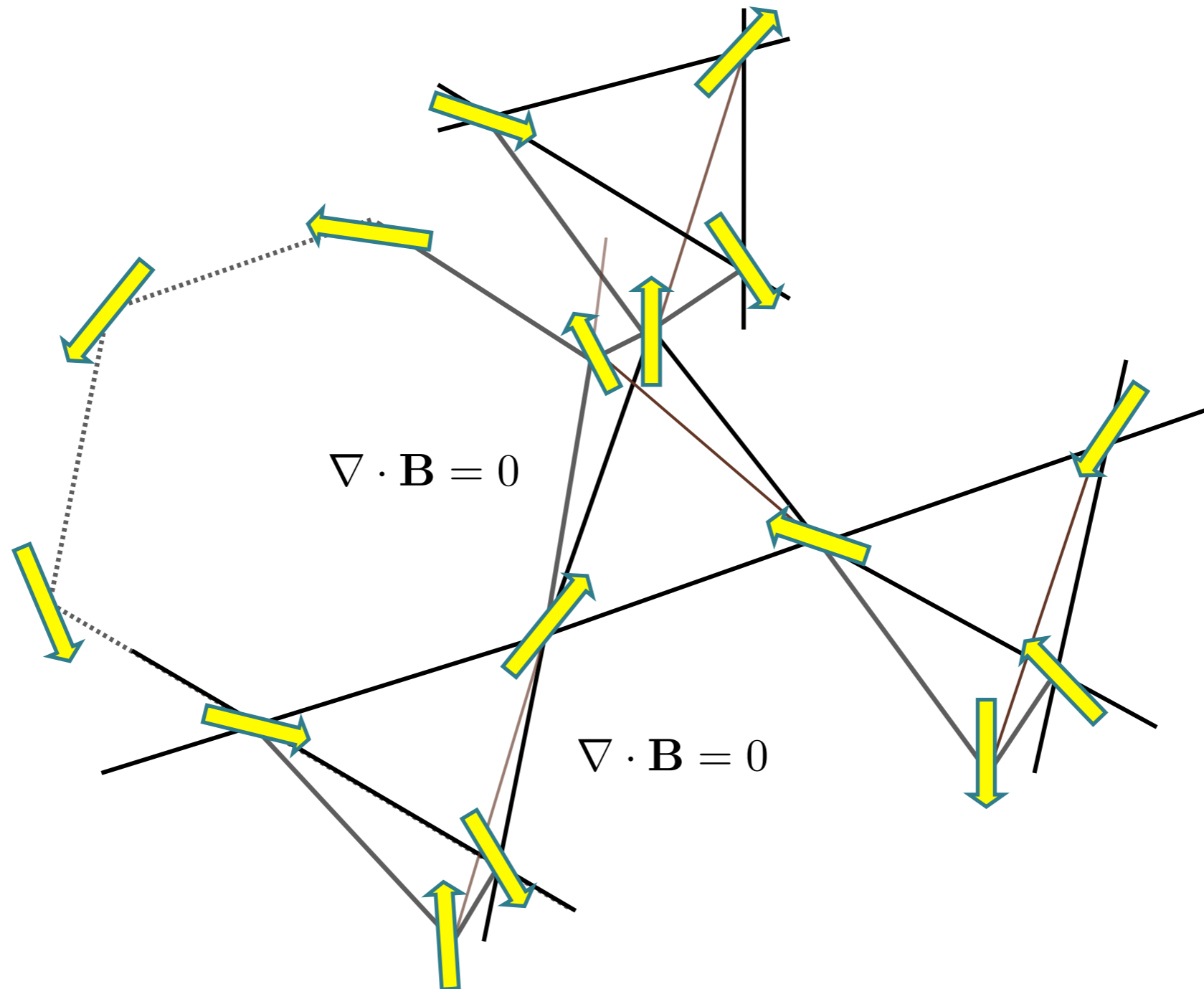


how might things change in
a quantum ice ?

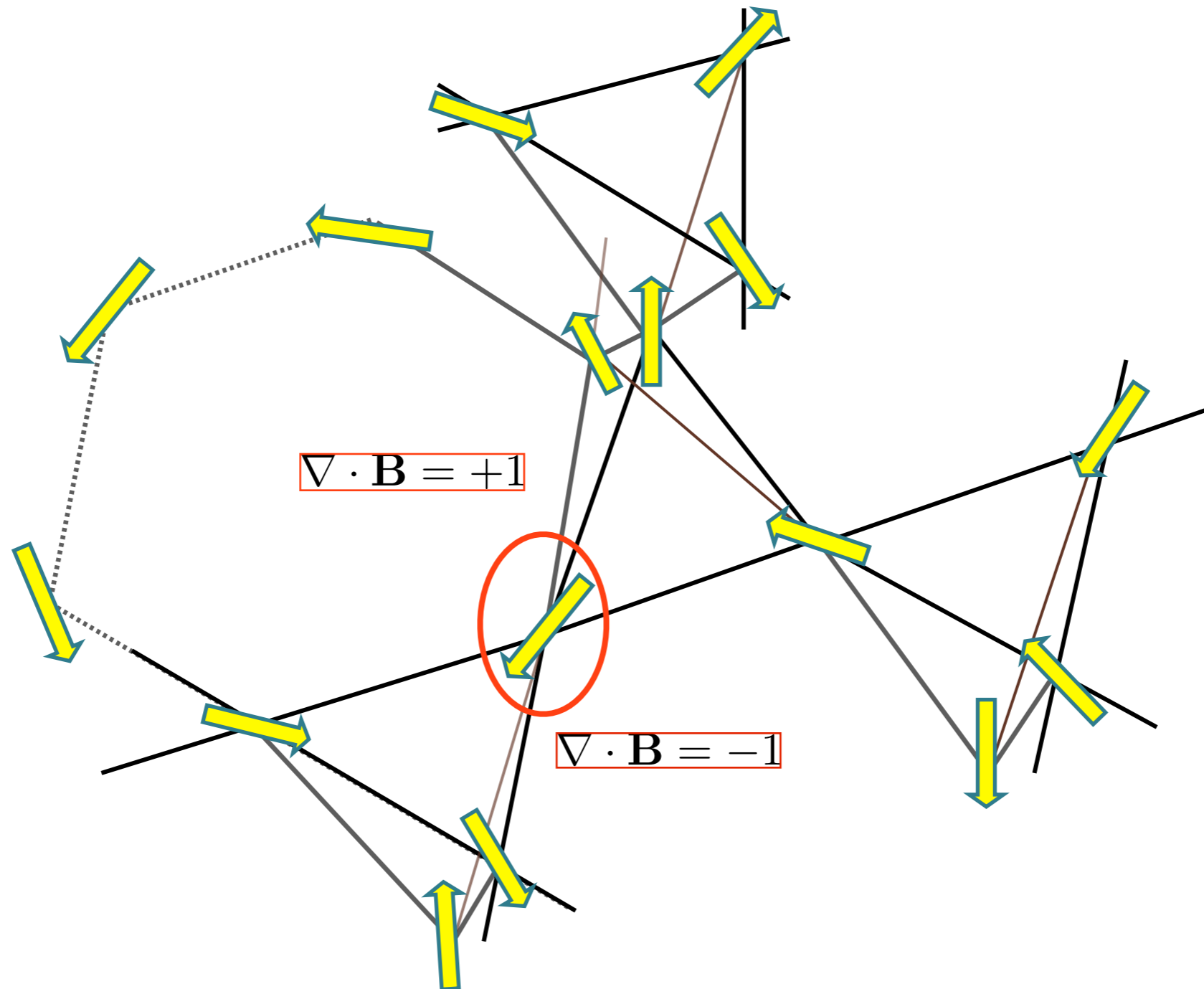
$$\hbar \neq 0$$

\Rightarrow tunnelling between ice states

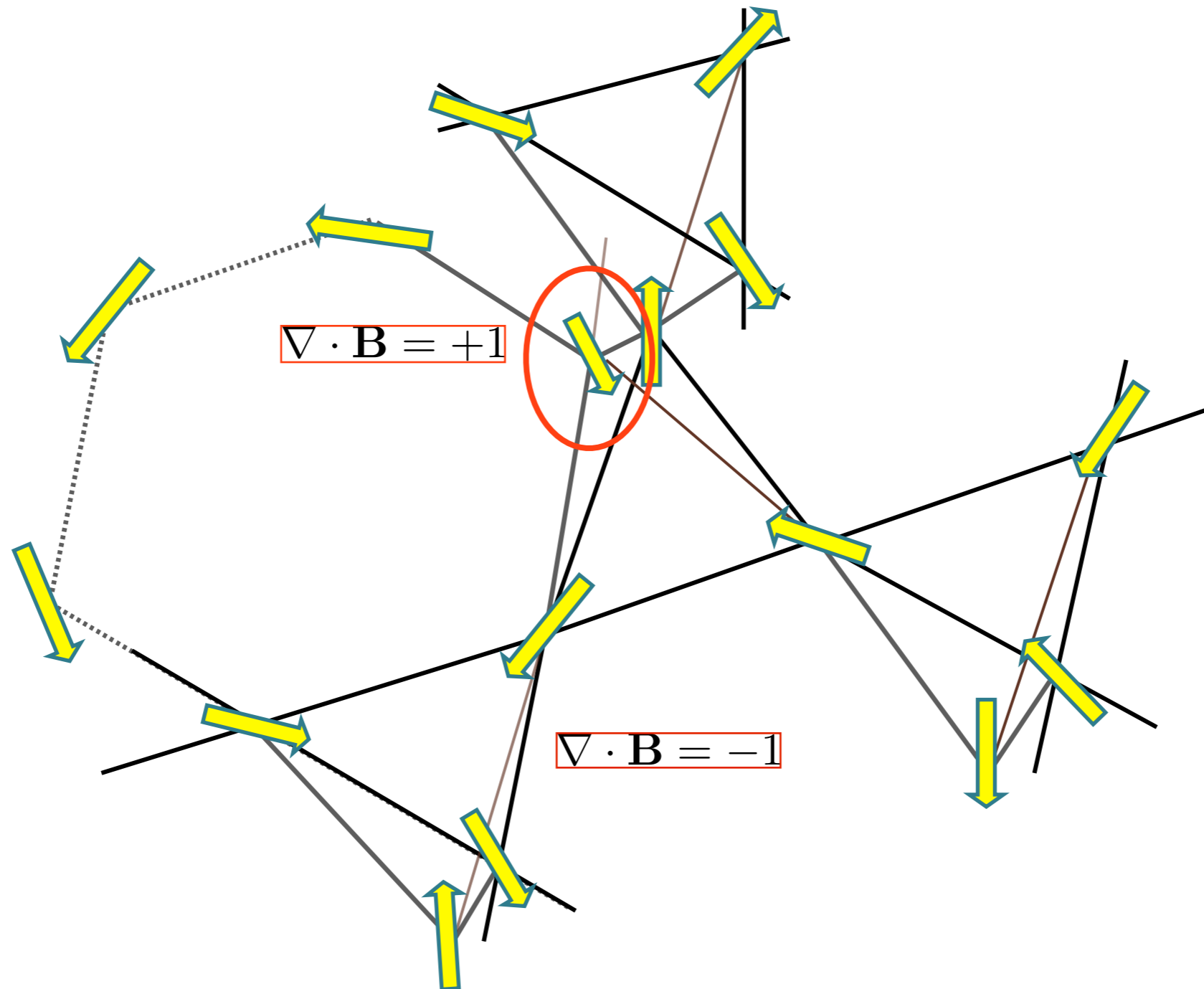
what kind of dynamics are there in ice ?



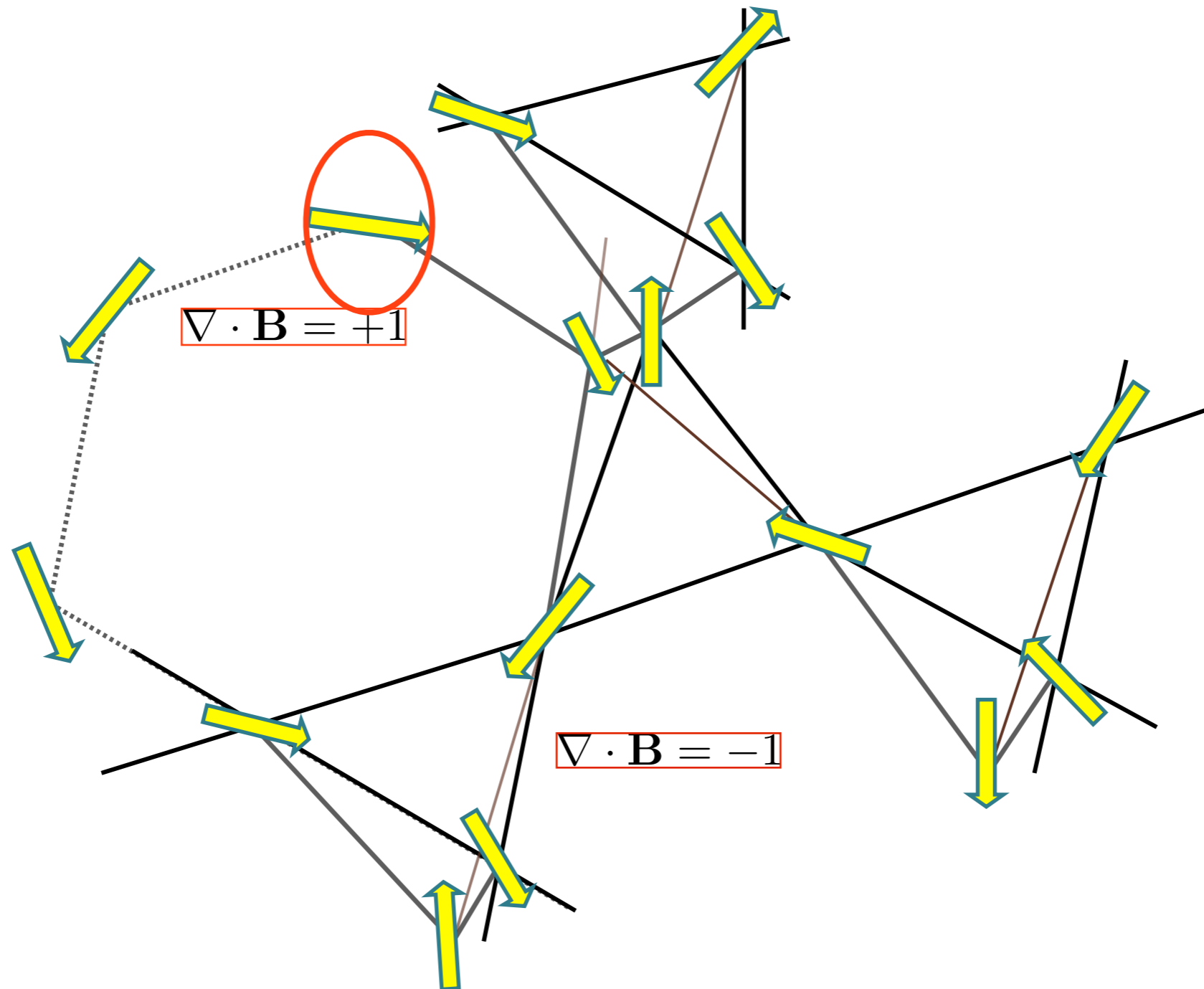
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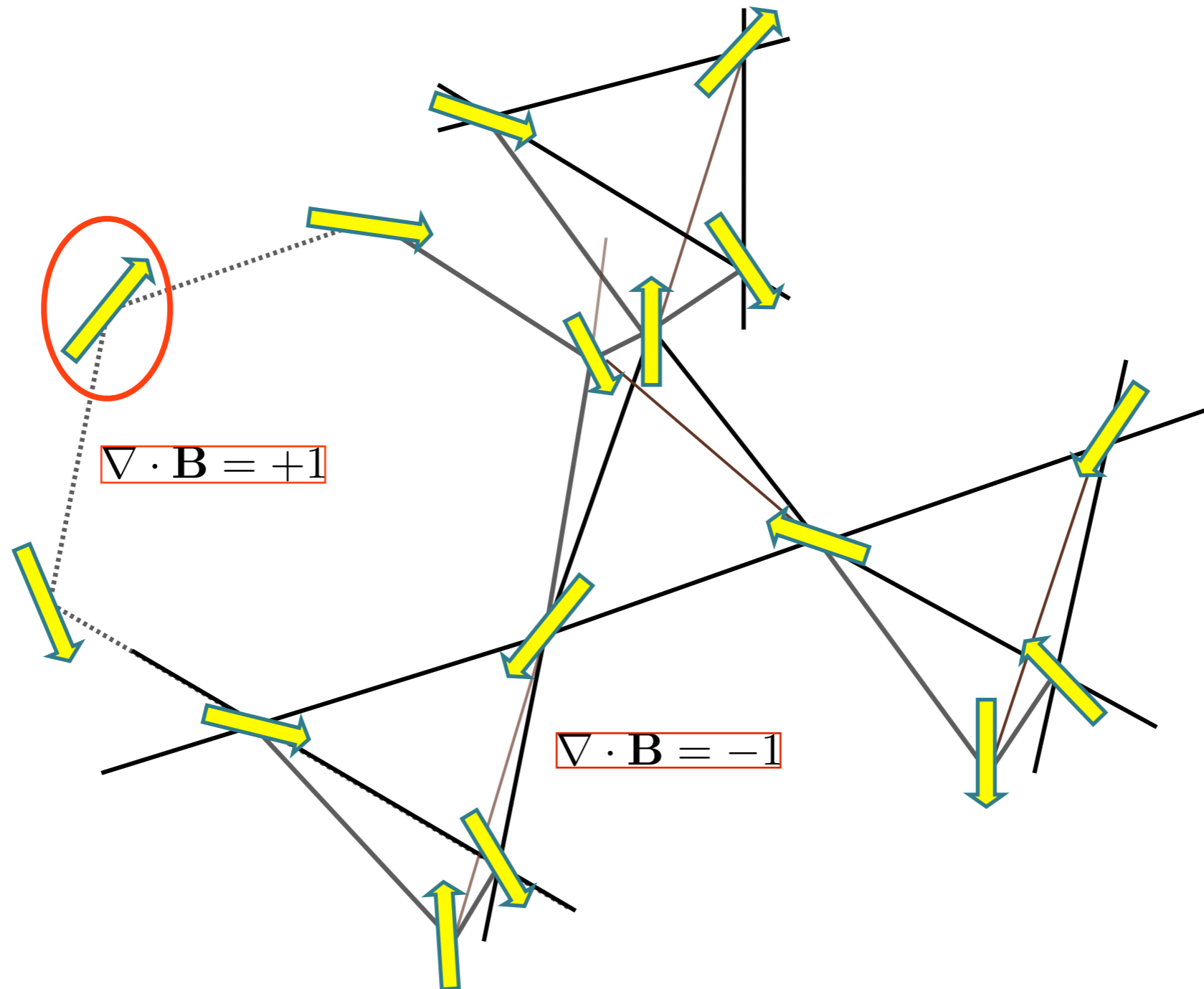
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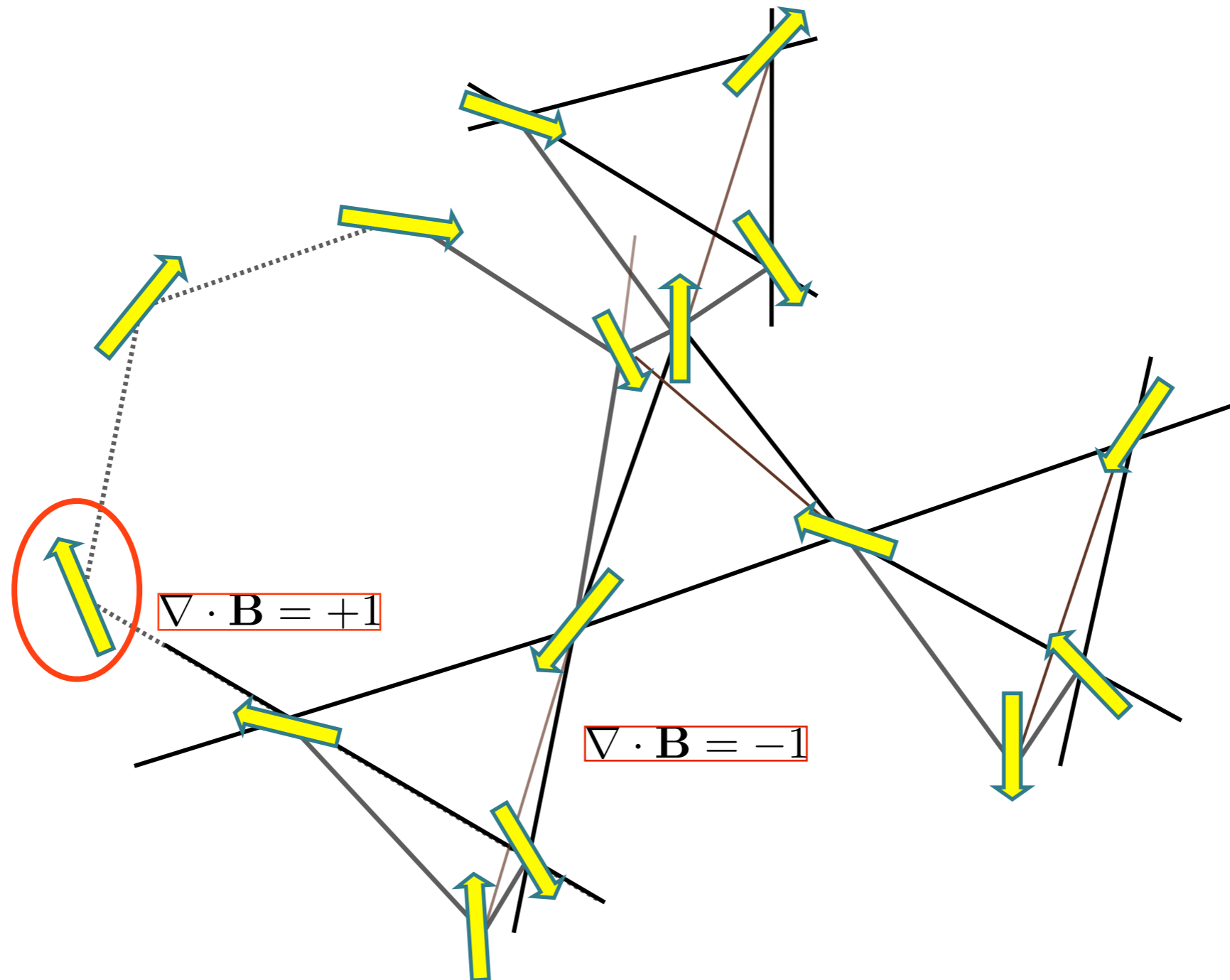
what kind of dynamics are there in ice ?



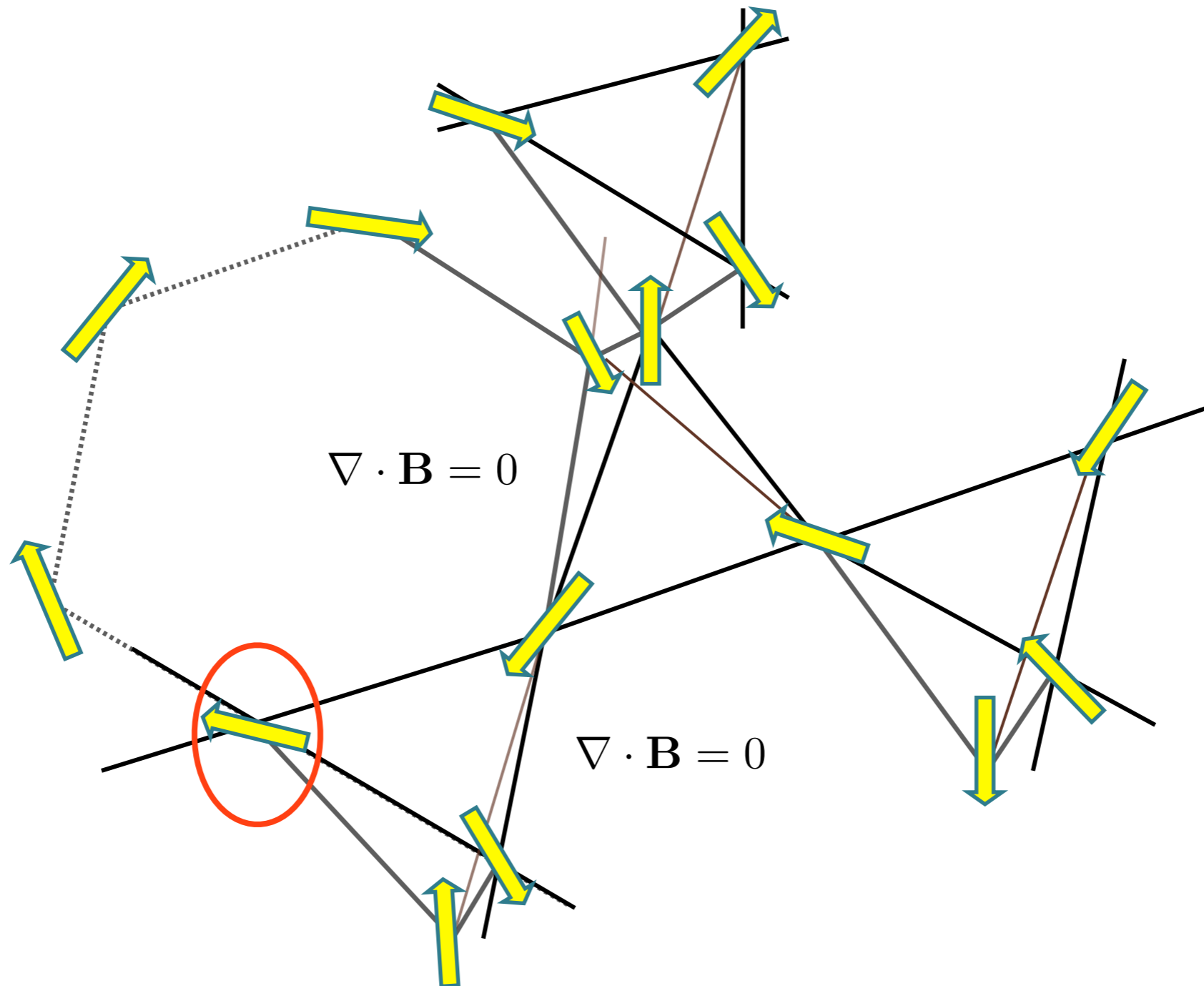
what kind of dynamics are there in ice ?



what kind of dynamics are there in ice ?

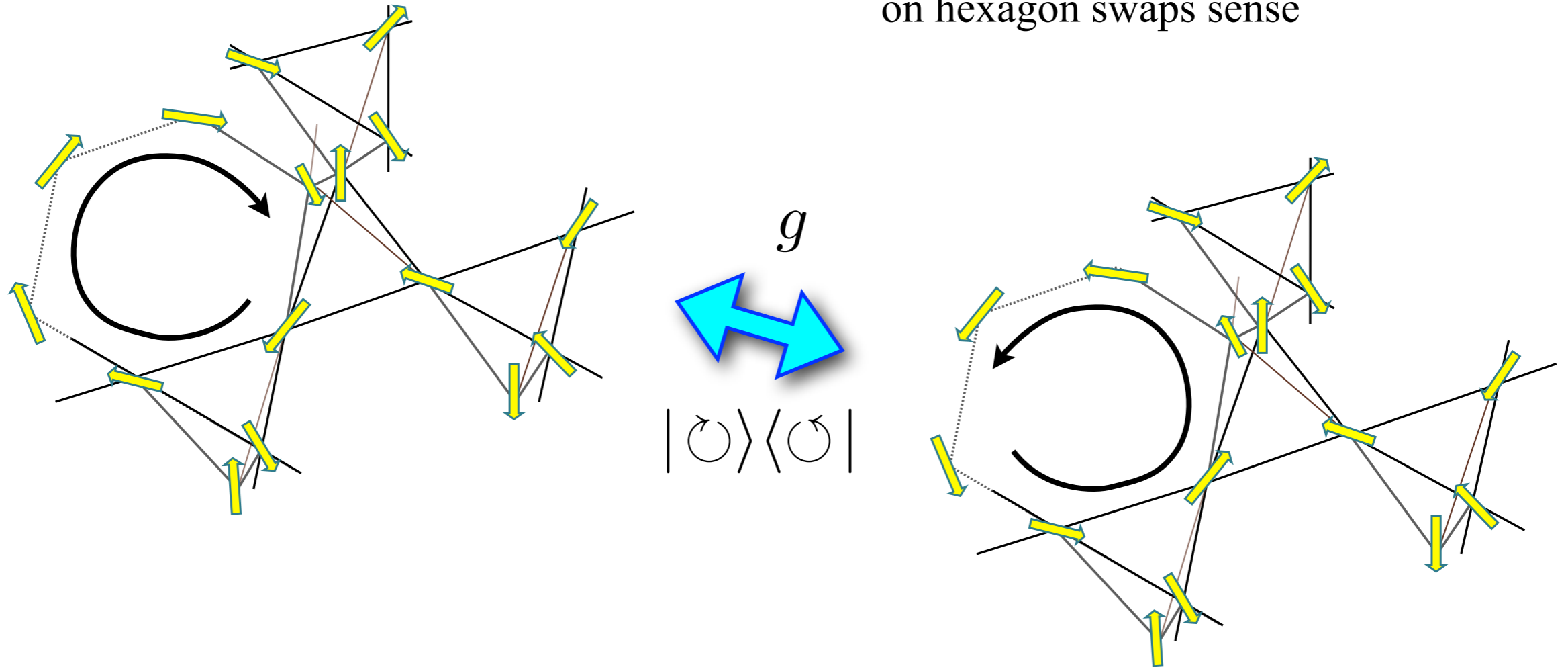


what kind of dynamics are there in ice ?



what kind of dynamics are there in ice ?

circulation of “magnetic” field
on hexagon swaps sense



what does this kind of quantum
tunneling do to (spin) ice ?



Cyclic exchange, isolated states, and spinon deconfinement in an XXZ Heisenberg model on the checkerboard lattice

Nic Shannon,^{1,2} Grégoire Misguich,³ and Karlo Penc⁴

$S=1/2$ easy-axis magnet on a checkerboard lattice...

$$\mathcal{H}_{xxz} = J_z \sum_{\langle ij \rangle} S_i^z S_j^z + J_{xy} \sum_{\langle ij \rangle} S_i^x S_j^x + S_i^y S_j^y$$

strong anisotropy

$$J_z \gg J_{xy}$$

selects ice manifold

Hamiltonian acting on ice states...

$$\mathcal{H} = -g \sum_{\square} |\circlearrowleft\rangle\langle\circlearrowleft| + |\circlearrowright\rangle\langle\circlearrowright| + \mu \sum_{\square} |\circlearrowleft\rangle\langle\circlearrowright| + |\circlearrowright\rangle\langle\circlearrowleft|$$

degenerate perturbation theory

$$g = \frac{2J_{xy}^2}{J_z}$$

...makes model exactly soluble for $\mu=g$

...2D quantum ice, *aka* the quantum six vertex model

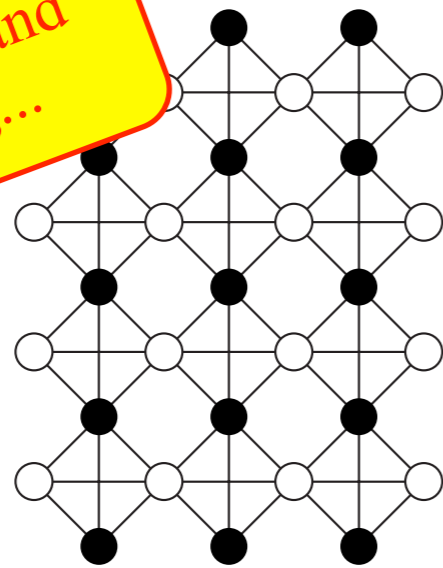


what happens happens at $T=0$?

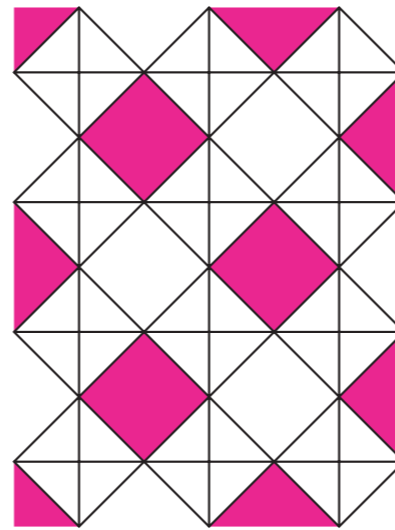
ground state phase diagram from exact diagonalisation :

NS et al. Phys. Rev. B **69**, 220403(R) (2004)

in 2D, all phases are ordered and confining...

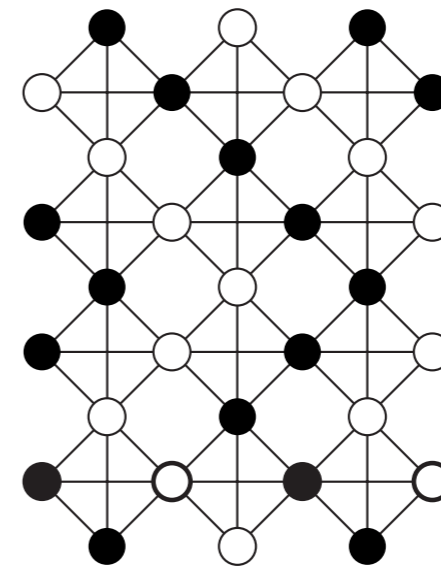


Néel



plaquette

RK



disordered



cf. O. F. Syljusen and S. Chakravarty, Phys. Rev. Lett. **96**, 147004 (2006).

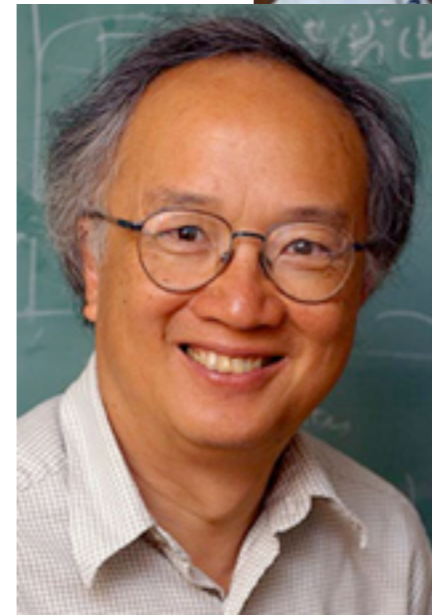
S. Chakravarty, Phys. Rev. B **66**, 224505 (2002).



no
spin liquid
then



v.



et al.



Pyrochlore photons: The $U(1)$ spin liquid in a $S = \frac{1}{2}$ three-dimensional frustrated magnet

Michael Hermele,¹ Matthew P. A. Fisher,² and Leon Balents¹

$S=1/2$ easy-axis magnet on a pyrochlore lattice...

$$\mathcal{H}_{xxz} = J_z \sum_{\langle ij \rangle} S_i^z S_j^z + J_{xy} \sum_{\langle ij \rangle} S_i^x S_j^x + S_i^y S_j^y$$

strong anisotropy

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selects ice manifold

Hamiltonian acting on ice states...

$$\mathcal{H} = -g \sum_{\text{hex}} |\circlearrowleft\rangle\langle\circlearrowleft| + |\circlearrowright\rangle\langle\circlearrowright| + \mu \sum_{\text{hex}} |\circlearrowleft\rangle\langle\circlearrowleft| + |\circlearrowright\rangle\langle\circlearrowright|$$

degenerate perturbation theory

$$g = \frac{12J_{xy}^3}{J_z^2}$$

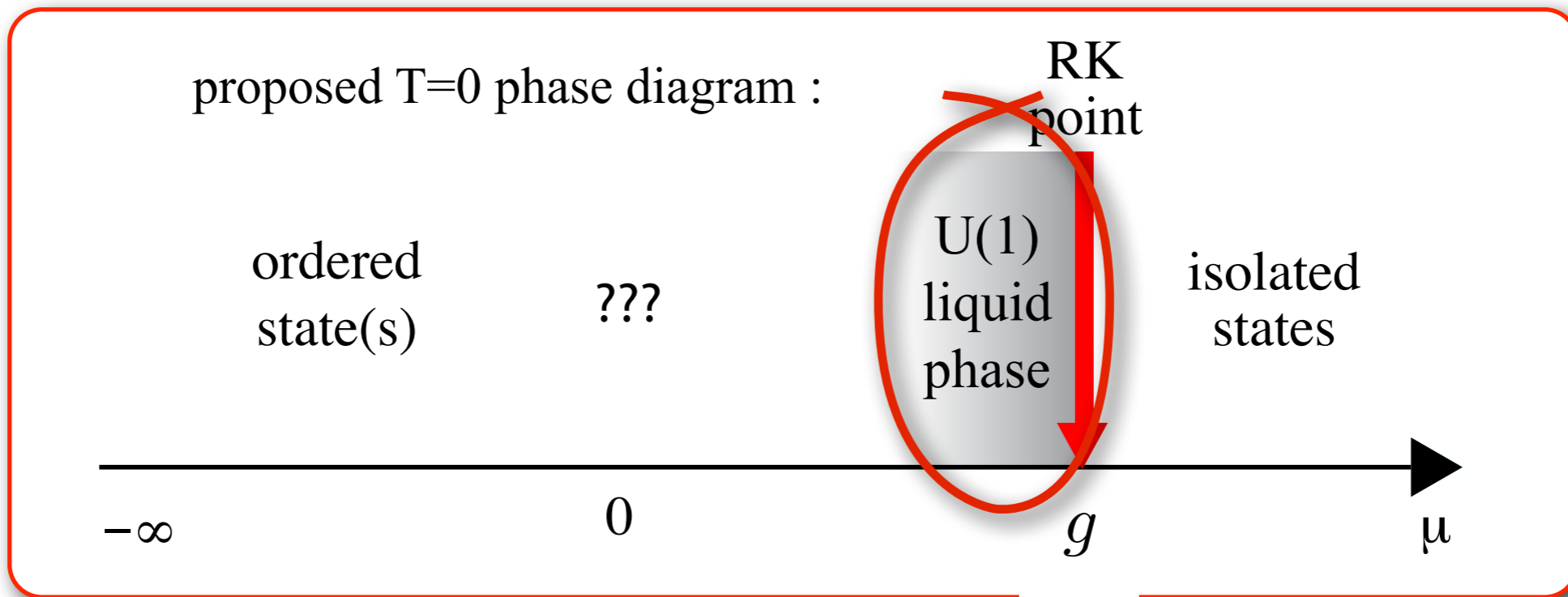
...extra term makes model exactly soluble for $\mu=g$



Pyrochlore photons: The $U(1)$ spin liquid in a $S = \frac{1}{2}$ three-dimensional frustrated magnet

Michael Hermele,¹ Matthew P. A. Fisher,² and Leon Balents¹

...microscopic model is equivalent to compact, frustrated, lattice $U(1)$ gauge theory



...equivalent proposal for 3D Quantum Dimer Model :
 R. Moessner and S Sondhi, Phys. Rev. B **68**, 184512 (2003)

$$\mathcal{H}_\mu = -g \sum_{\langle ij \rangle} |\langle i | \sigma_j^x | i \rangle\rangle \langle i | \sigma_j^x | i \rangle + \mu \sum_{\langle ij \rangle} |\langle i | \sigma_j^z | i \rangle\rangle \langle i | \sigma_j^z | i \rangle + \dots$$



does this idea work ?



Unusual Liquid State of Hard-Core Bosons on the Pyrochlore Lattice

Argha Banerjee,¹ Sergei V. Isakov,² Kedar Damle,¹ and Yong Baek Kim²

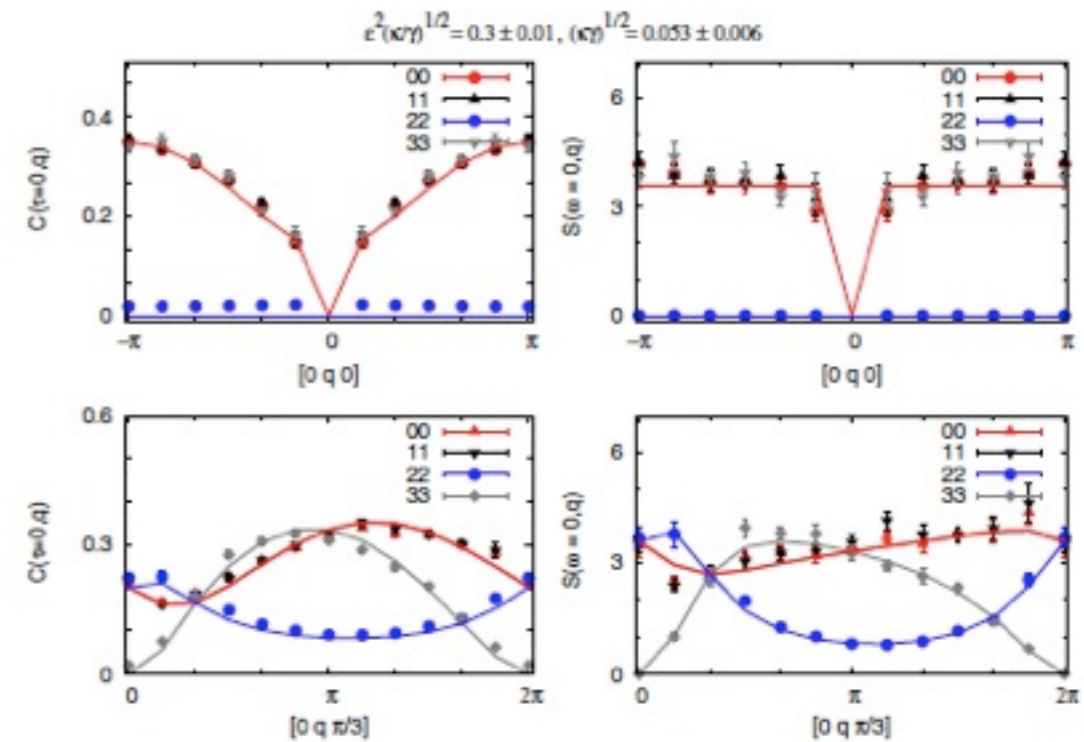
consider hard-core Bosons with
strong nearest neighbour
interactions $V \gg t$
on a pyrochlore lattice

$\mathcal{H}_{\text{charge-ice}}$

$$= -t \sum_{\langle ij \rangle} (b_i^\dagger b_j + b_j^\dagger b_i) \\ + V \sum_{\langle ij \rangle} \left(n_i - \frac{1}{2} \right) \left(n_j - \frac{1}{2} \right)$$

quantum charge ice with tunneling

$$g = 12t^3 / V^2$$



finite temperature correlation functions,
calculated using QMC, and compared to the
predictions of a U(1) gauge theory



...suppose it orders at a lower temperature

effective Hamiltonian in ice manifold



$$\mathcal{H} = -g \sum_{\square} |\uparrow\rangle\langle\uparrow| + |\downarrow\rangle\langle\downarrow| + \mu \sum_{\square} |\uparrow\rangle\langle\downarrow| + |\downarrow\rangle\langle\uparrow|$$

all off-diagonal
matrix elements
have definite
(negative) sign

suitable for T=0 GFMC simulation as no sign problems
(but simulation doesn't need to converge in finite time !)



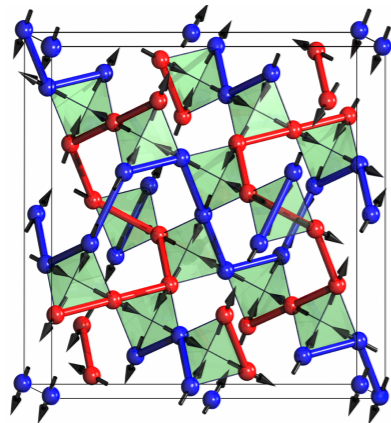
...some CPU-centuries later



Quantum Ice: A Quantum Monte Carlo Study

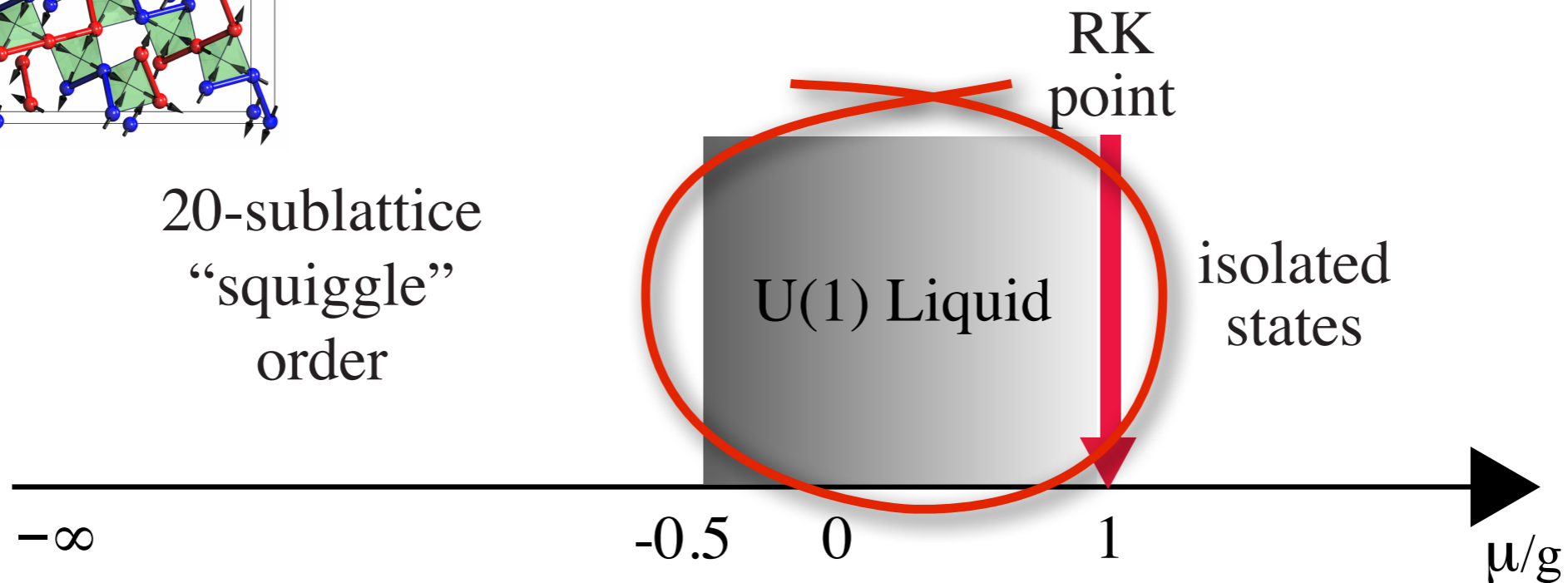
Nic Shannon,¹ Olga Sikora,¹ Frank Pollmann,² Karlo Penc,³ and Peter Fulde^{2,4}

$$\mathcal{H}_\mu = -g \sum_{\langle ij \rangle} |\uparrow_i\rangle\langle\uparrow_j| + |\downarrow_i\rangle\langle\downarrow_j| + \mu \sum_{\langle ij \rangle} |\uparrow_i\rangle\langle\uparrow_j| + |\downarrow_i\rangle\langle\downarrow_j|$$



20-sublattice
"squiggle"
order

extended T=0 quantum
spin liquid phase !



ground state phase diagram from Quantum Monte Carlo simulation



so what's a quantum U(1) liquid ?

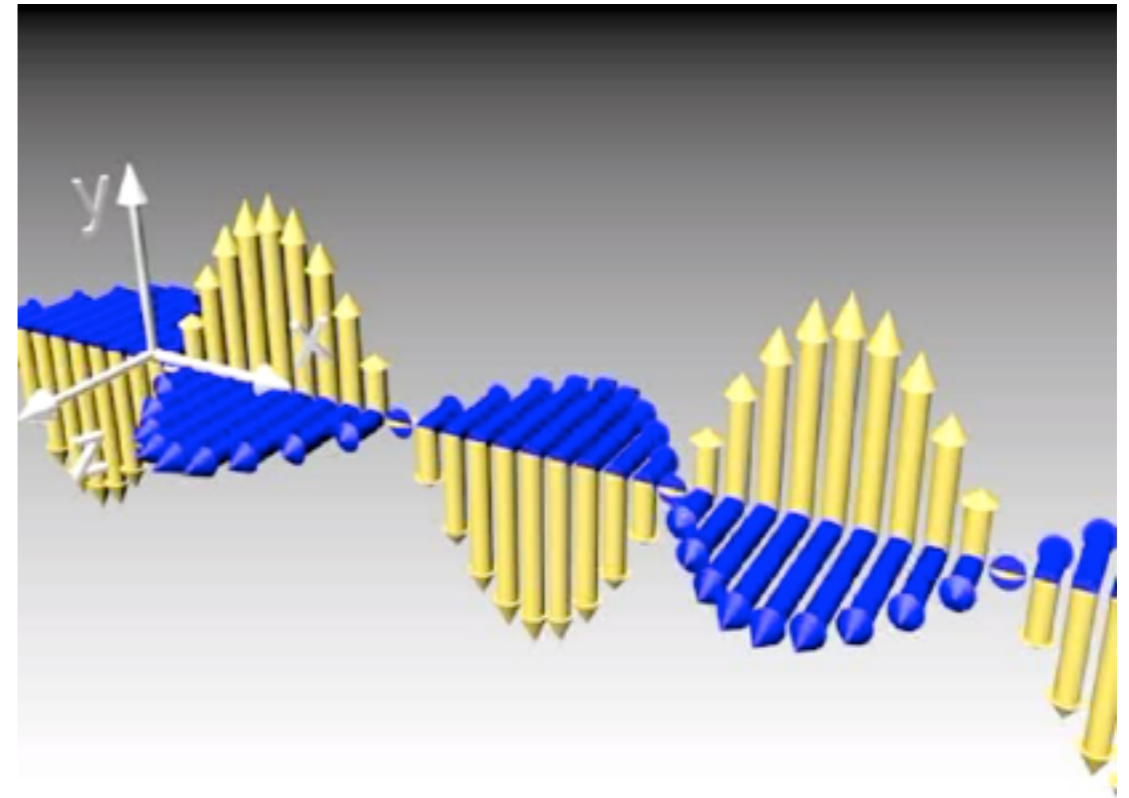
- * **quantum (spin) liquid** with excitations described by the Maxwell action of classical electromagnetism
- * supports both electric and magnetic charges (**magnetic monopoles**)
- * electric and magnetic fields couple to give **photons**

$$S = \int d^3x dt [\mathbf{E}^2 - c^2 \mathbf{B}^2]$$



so what's a quantum U(1) liquid ?

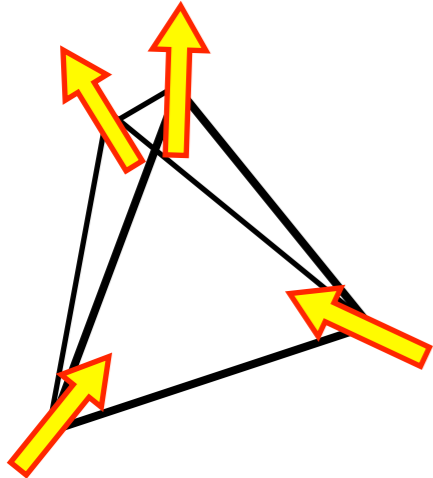
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$$S = \int d^3x dt [\mathbf{E}^2 - c^2 \mathbf{B}^2]$$



how does this happen ?



$$\nabla \cdot \mathbf{B} = 0 \quad \dots \text{by explicit construction}$$

$$\text{solve as : } \mathbf{B} = \nabla \times \mathbf{A} \quad \text{and chose Coulomb gauge : } \nabla \cdot \mathbf{A} = 0$$

quantum ice has local dynamics :

$$\mathcal{H} = -g \sum_{\text{hex}} |\circlearrowleft\rangle\langle\circlearrowleft| + |\circlearrowright\rangle\langle\circlearrowright|$$

tunneling between ice states \Rightarrow gauge field varies in time
simplest guess for effective field theory in a liquid phase is **Maxwell** action :

$$S = \int d^3x dt [\mathbf{E}^2 - c^2 \mathbf{B}^2]$$

$$\partial_t \mathbf{A} - \nabla A_0$$



same story, longer equations...

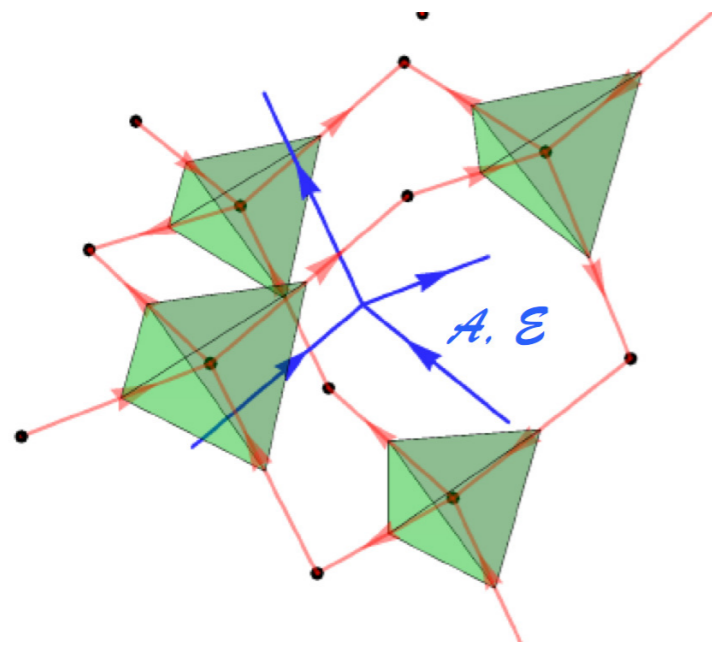
compact U(1) lattice gauge theory...

$$\mathcal{H}'_{U(1)} = \frac{U}{2} \sum_{\mathbf{r} \in A, n} \left[(\nabla_{\square} \times \mathcal{A})_{(\mathbf{r}, n)} \right]^2 + \frac{1}{2\mathcal{K}} \sum_{\mathbf{s} \in A', m} \left[\frac{\partial \mathcal{A}_{(\mathbf{s}, m)}}{\partial t} \right]^2 + \frac{\mathcal{W}}{2} \sum_{\mathbf{s} \in A', m} \left[(\nabla_{\square} \times \nabla_{\square} \times \mathcal{A})_{(\mathbf{s}, m)} \right]^2$$

“ice”
term

tunnelling
term

“μ” term
(relevant at RK point)



...theory is quadratic in gauge field

can diagonalise problem by introducing suitable photon basis :

$$\begin{aligned} \mathcal{A}_{(\mathbf{s}, m)} = & \sqrt{\frac{2}{N}} \sum_{\mathbf{k}} \sum_{\lambda=1}^4 \sqrt{\frac{\mathcal{K}}{\omega_{\lambda}(\mathbf{k})}} \\ & \times \left(\exp[-i\mathbf{k} \cdot (\mathbf{s} + \mathbf{e}_m/2)] \eta_{m\lambda}(\mathbf{k}) a_{\lambda}(\mathbf{k}) \right. \\ & \left. + \exp[i\mathbf{k} \cdot (\mathbf{s} + \mathbf{e}_m/2)] \eta_{\lambda m}^*(\mathbf{k}) a_{\lambda}^{\dagger}(\mathbf{k}) \right) \end{aligned}$$



DYNAMICAL STABILITY OF LOCAL GAUGE SYMMETRY

Creation of Light From Chaos

D. FOERSTER

Service de Physique Théorique, CEN Saclay, F-91190 Gif-sur-Yvette, France

H.B. NIELSEN

*The Niels Bohr Institute, University of Copenhagen, and NORDITA,
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and

M. NINOMIYA

The Niels Bohr Institute, University of Copenhagen, DK-2100 Copenhagen ϕ , Denmark

Received 14 May 1980



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PHYSICAL REVIEW LETTERS

7 JANUARY 2002

Origin of Gauge Bosons from Strong Quantum Correlations

Xiao-Gang Wen*

VOLUME 89, NUMBER 27

PHYSICAL REVIEW LETTERS

30 DECEMBER 2002

Exotic Order in Simple Models of Bosonic Systems

O. I. Motrunich and T. Senthil

a spin liquid with many
advocates...



X.-G. Wen, Phys. Rev. B **68**, 115413 (2003).

R. Moessner and S. Sondhi, Phys. Rev. B **68**, 184512 (2003)

M. Hermele, L. Balents and M. Fisher, Phys. Rev. B **69**, 064404 (2004)

O. I. Motrunich and A. Vishwanath, Phys. Rev. B **70**, 075104 (2004)

O. I. Motrunich and T. Senthil, Phys. Rev. B **71**, 125102 (2005)

M. Levin and X.-G. Wen, Rev. Mod. Phys. **77**, 871 (2005)

(and many more...)

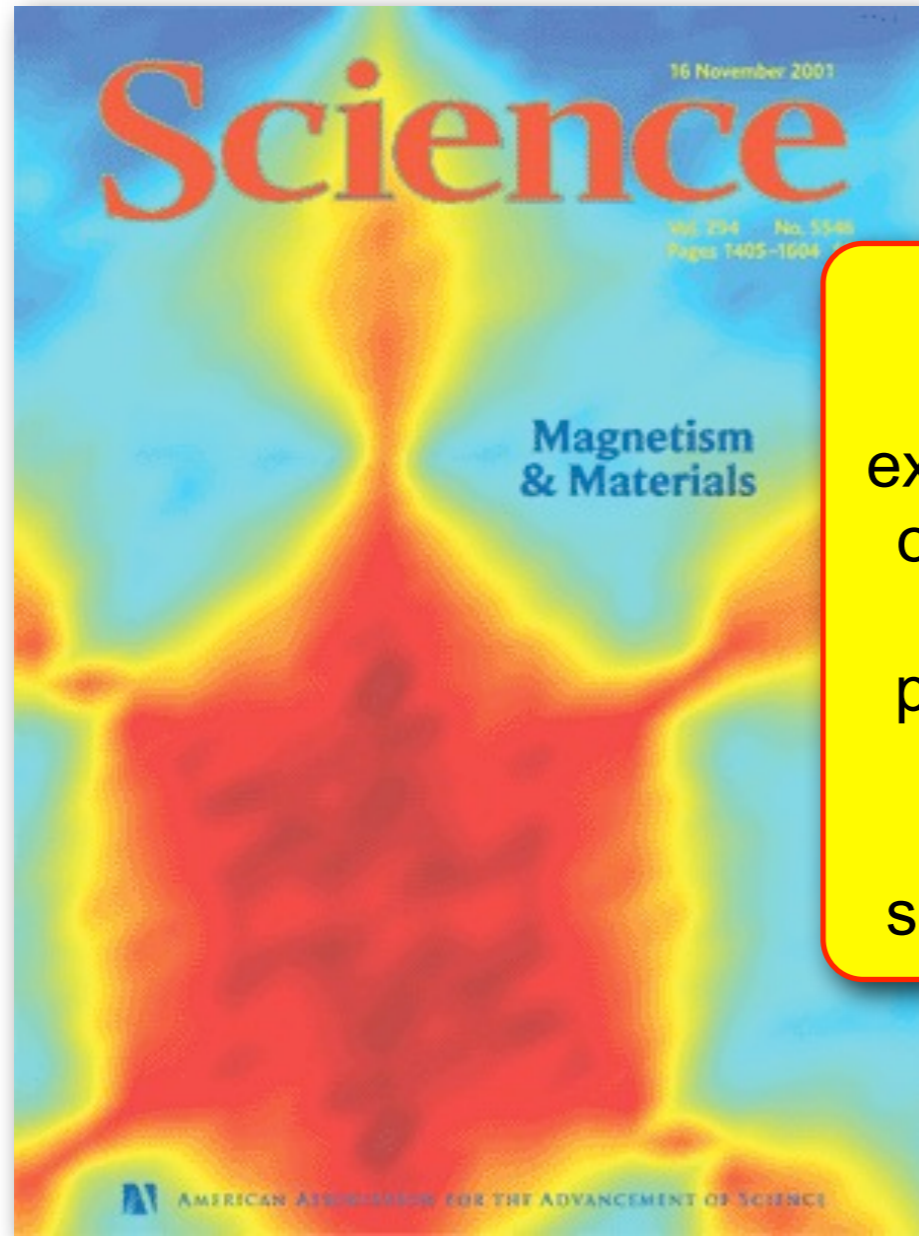
the story so far...

classical ice = magnetostatics

quantum ice = electromagnetism

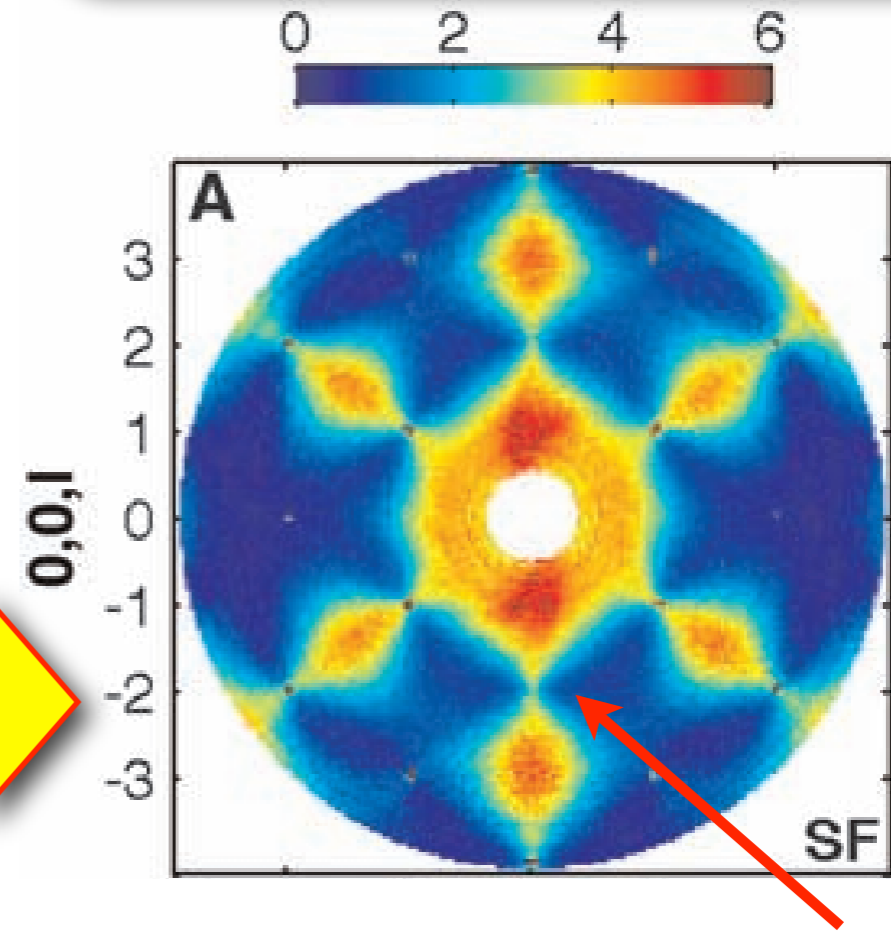
...so what ?

how can we “see” the ice rules ?



for spin
ices,
experiment
of choice
is spin-
polarised
elastic
neutron
scattering

experimental results for
 $\text{Ho}_2\text{Ti}_2\text{O}_7$ in spin-flip channel



pinch point

T. Fennell et al, Science **326**, 415 (2009).



what does a quantum ice look like ?

quantum fluctuations in the quantum U(1) liquid suppress pinch points

$$S^{\alpha\beta}(\mathbf{q}) \propto \langle \mathcal{B}_\alpha(-\mathbf{q})\mathcal{B}_\beta(\mathbf{q}) \rangle = \frac{8\pi^4 q}{c} \left[\delta_{\alpha\beta} - \frac{q_\alpha q_\beta}{q^2} \right] \coth \left(\frac{cq}{2T} \right)$$

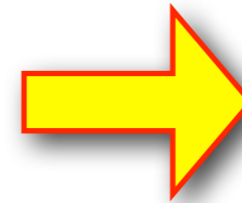
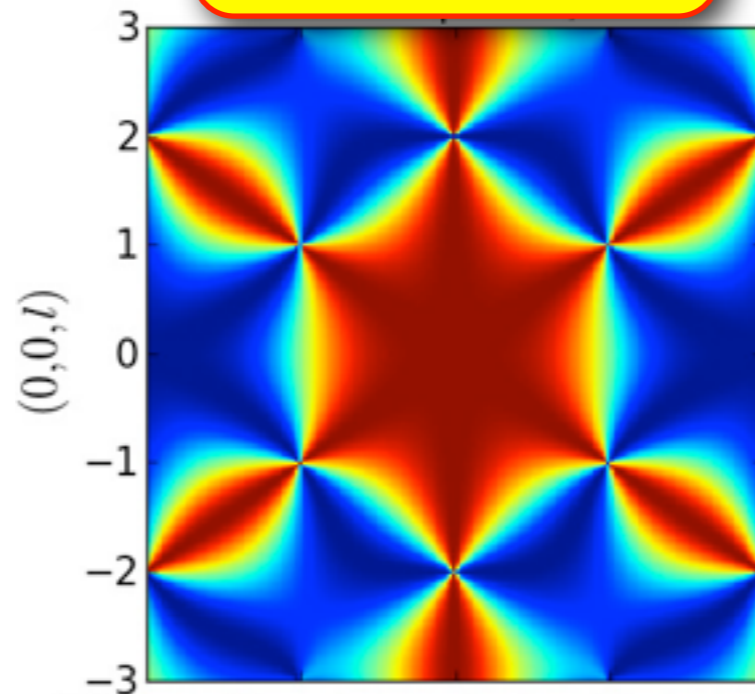
what would this look like in a quantum spin ice ?

predictions for quasi-elastic structure factor at T=0



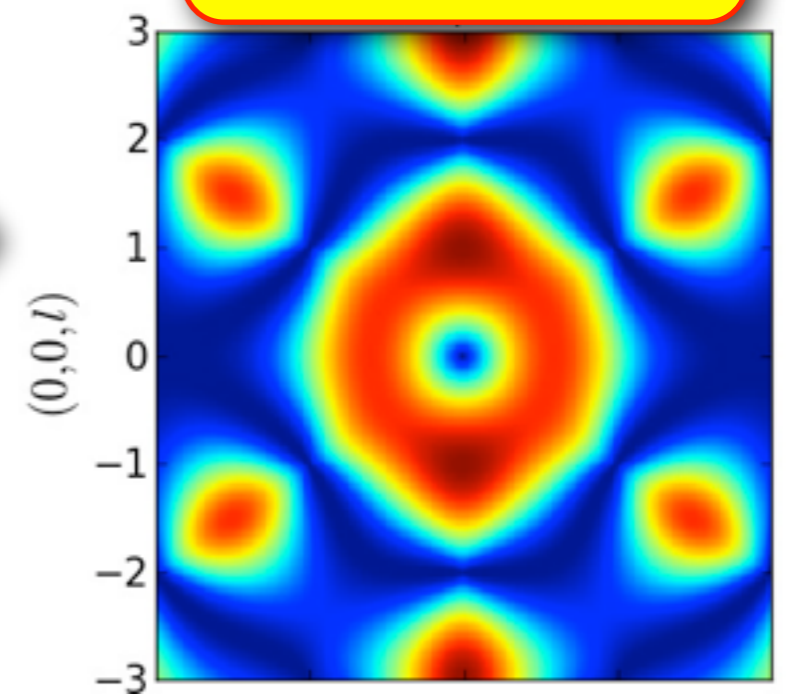
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classical ice



finite speed of light

quantum ice

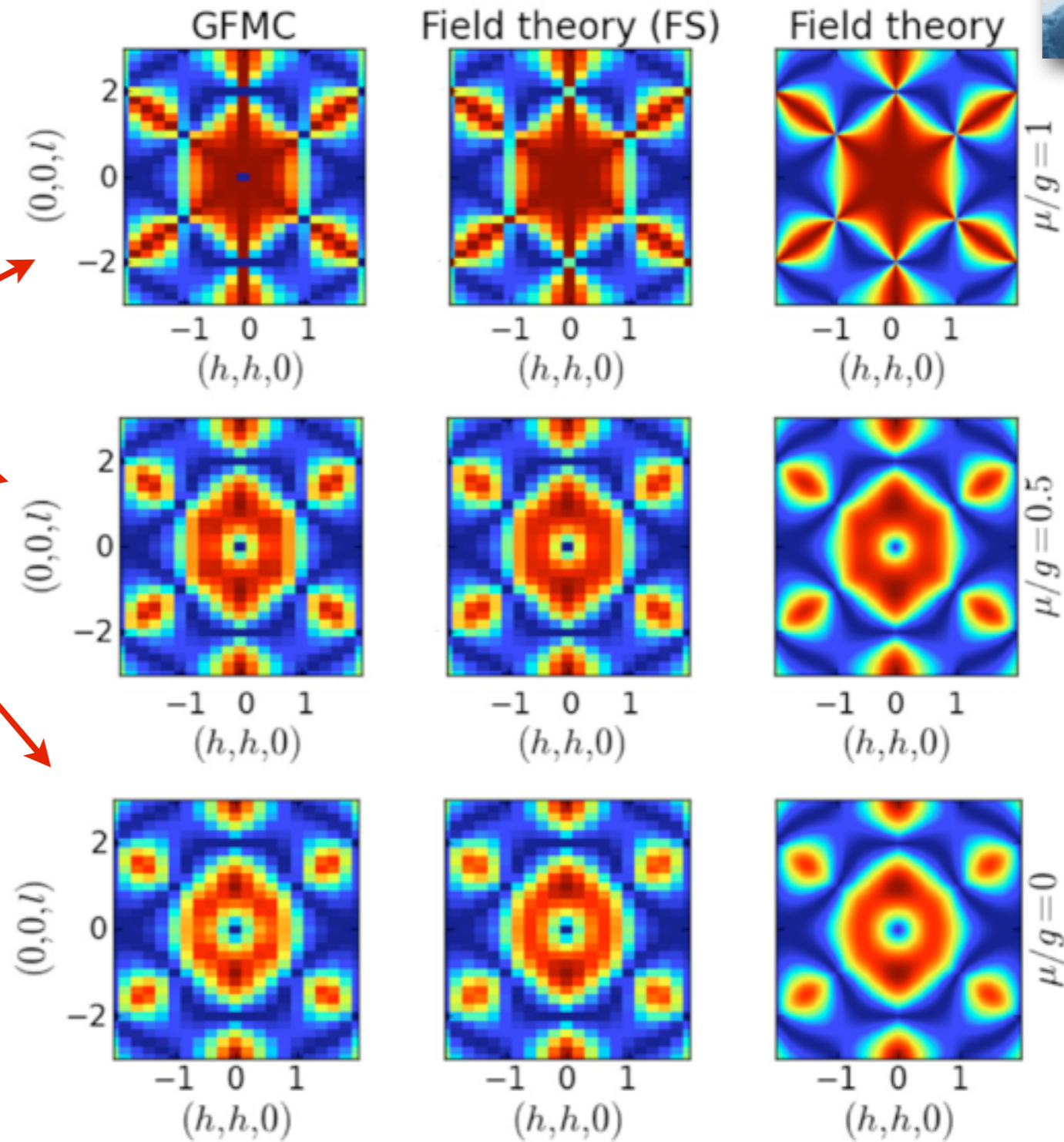
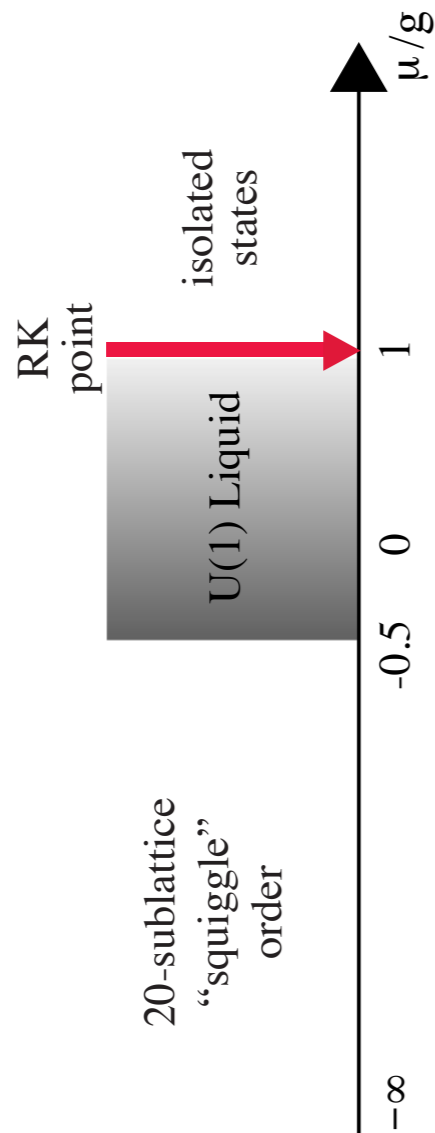


...no pinch points !



$$\mathcal{H}_\mu = -g \sum_{\langle ij \rangle} |\uparrow_i\rangle\langle\uparrow_j| + |\downarrow_i\rangle\langle\downarrow_j| + \mu \sum_{\langle ij \rangle} |\uparrow_i\rangle\langle\uparrow_j| + |\downarrow_i\rangle\langle\downarrow_j|$$

is this what we see in simulation ?

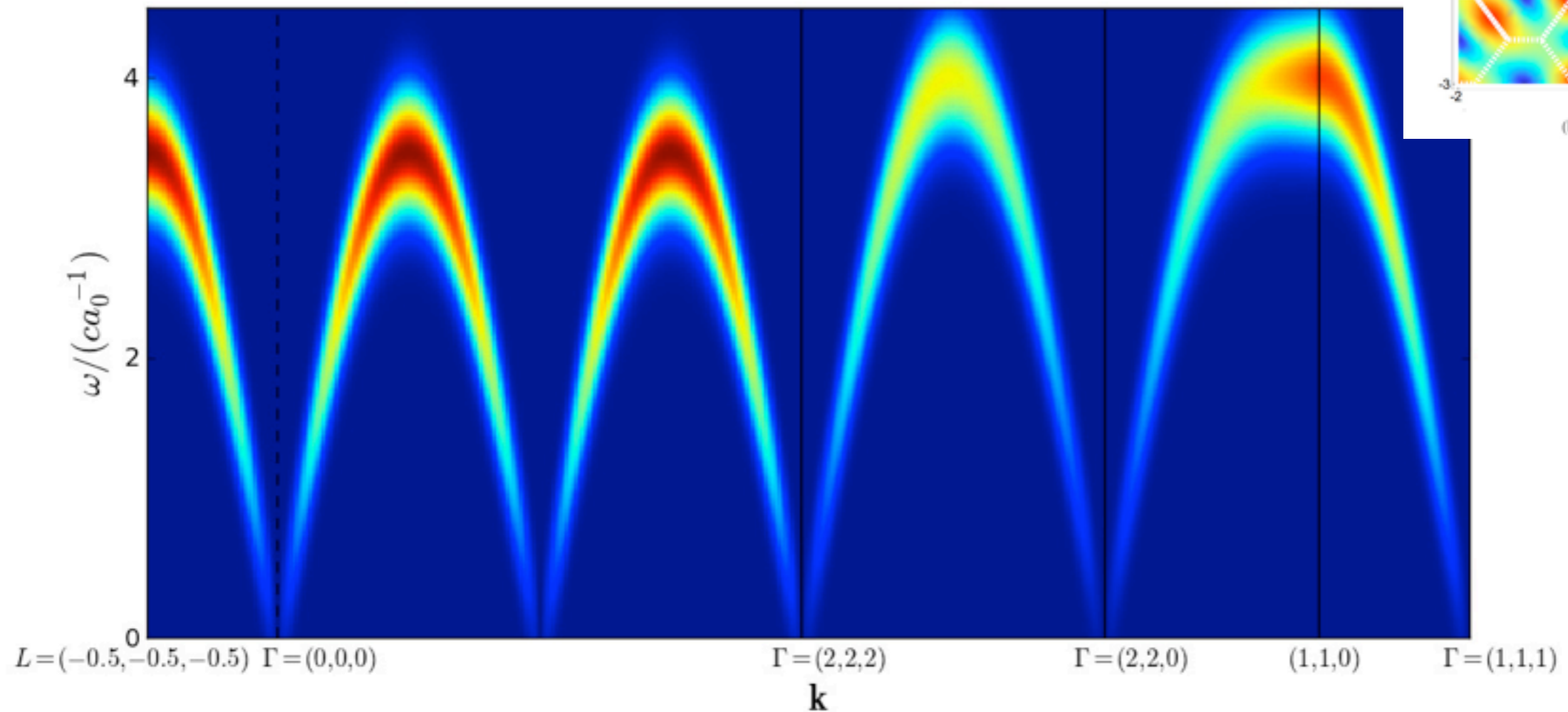


O. Benton *et al.*, Phys. Rev. B. **83**, 075174 (2012)



seeing the light ?

quantitative prediction for inelastic neutron scattering
from lattice gauge theory



O. Benton *et al.*, Phys. Rev. B. **83**, 075174 (2012) 

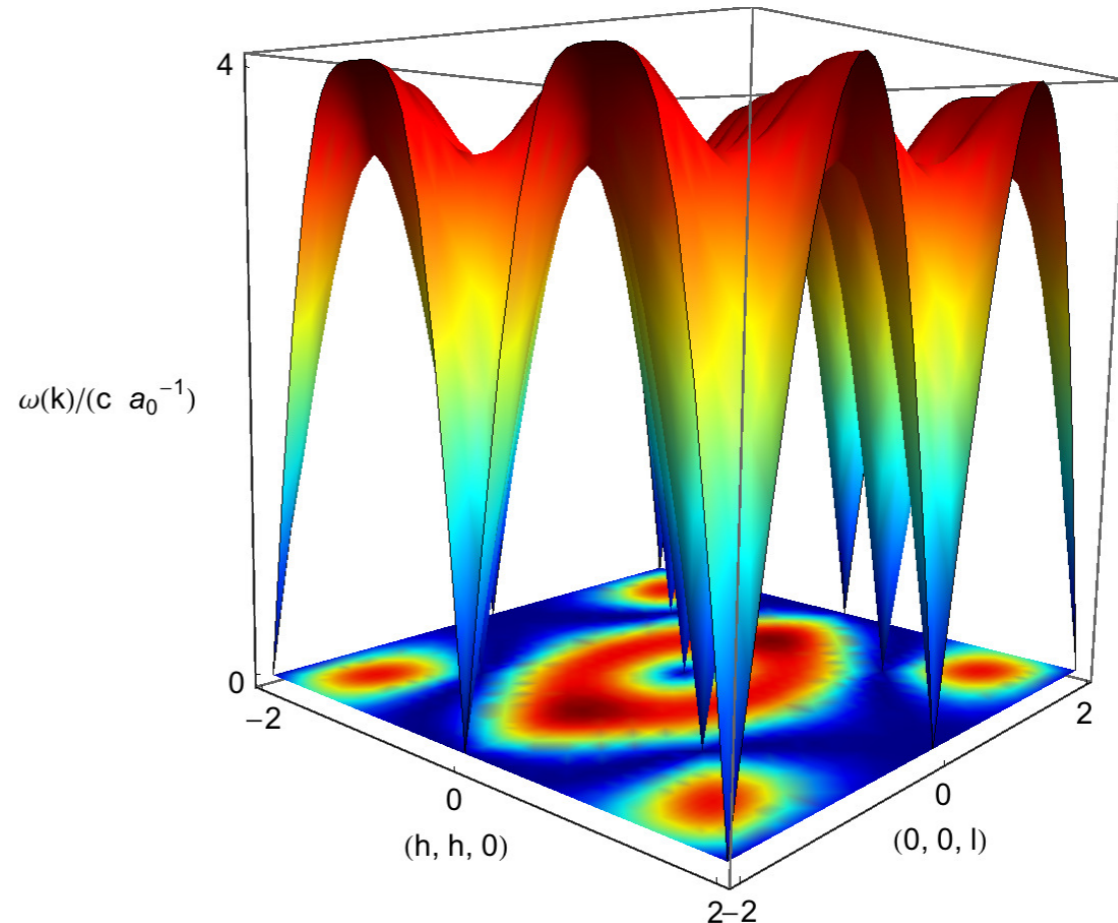


where did the pinch-points go ?

quasi-elastic scattering
given by energy-integral of
dynamical structure factor

$$S^{\alpha\beta}(\mathbf{q}) = \int d\omega S^{\alpha\beta}(\mathbf{q}, \omega)$$

vanishing spectral weight in
photons at low energies
 \Rightarrow loss of pinch-points



cf. M. Hermele, L. Balents and M. Fisher, Phys. Rev. B **69**, 064404 (2004)
L. Savary and L. Balents. Phys. Rev. Lett. **108**, 037202 (2012)



what happens at finite temperature ?

photons can be thermally excited, just like phonons...

$$S^{\alpha\beta}(\mathbf{q}) \propto \langle \mathcal{B}_\alpha(-\mathbf{q})\mathcal{B}_\beta(\mathbf{q}) \rangle = \frac{8\pi^4 q}{c} \left[\delta_{\alpha\beta} - \frac{q_\alpha q_\beta}{q^2} \right] \coth\left(\frac{cq}{2T}\right)$$

...how does this change the scattering ?



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$$\lim_{q \rightarrow 0} \coth\left(\frac{cq}{2T}\right) \propto \frac{T}{q} \Rightarrow$$

$$S^{\alpha\beta}(\mathbf{q} \approx 0) \propto T \left[\delta_{\alpha\beta} - \frac{q_\alpha q_\beta}{q^2} \right]$$

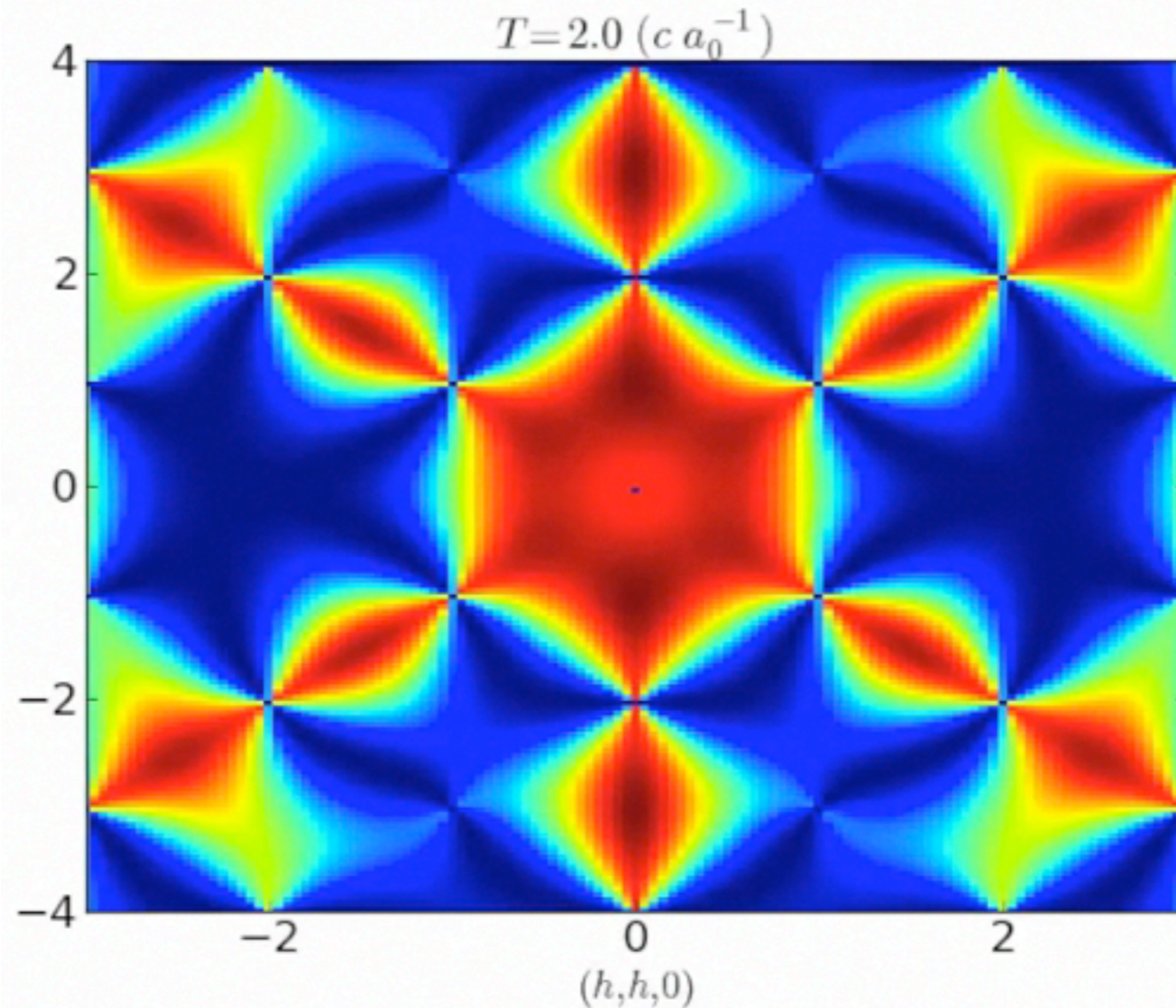
...i.e. pinch points vanish linearly with T

O. Benton *et al.*, Phys. Rev. B. **83**, 075174 (2012) 🐣



what happens at finite temperature ?

pinch points “wash out” as system is cooled to $T=0$

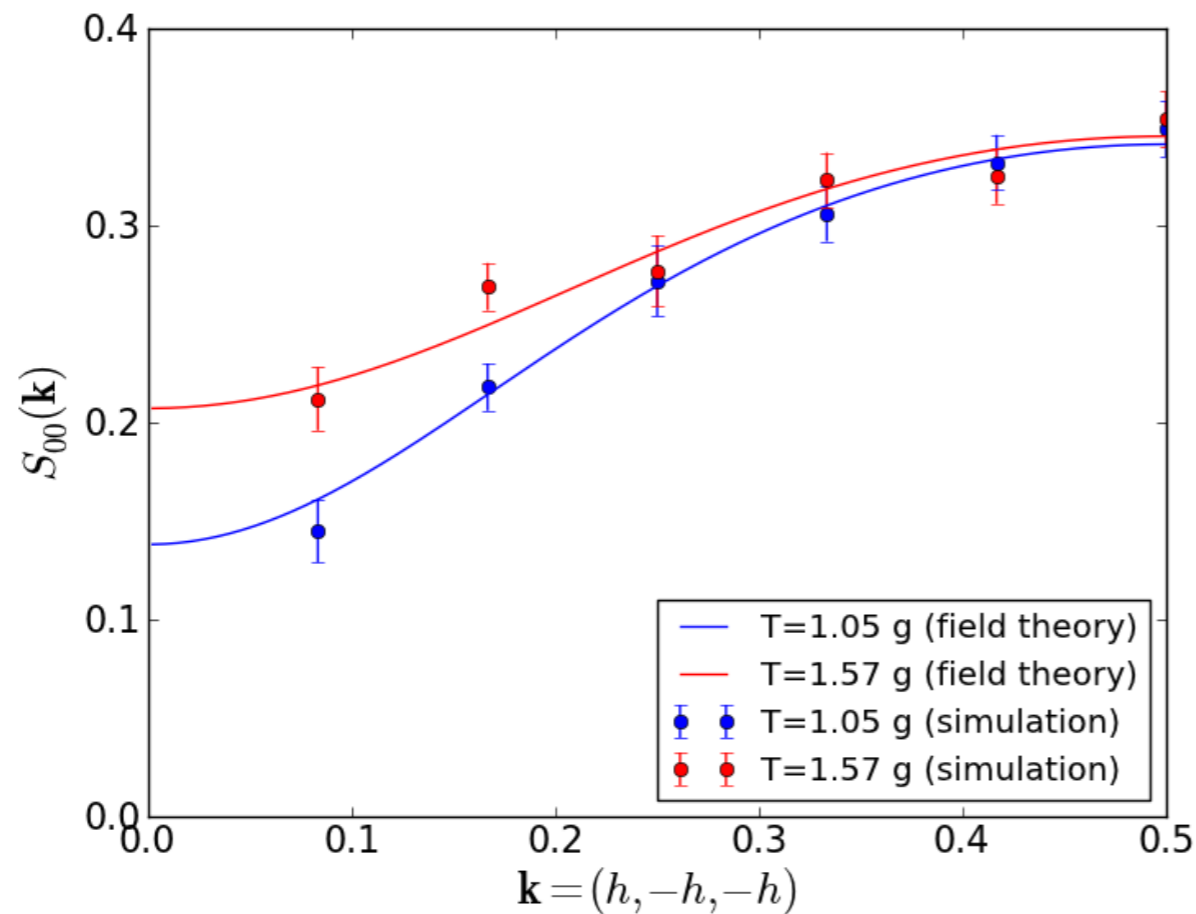


...predictions of lattice gauge theory

O. Benton *et al.*, Phys. Rev. B. **83**, 075174 (2012) 




what about simulation ?



points : finite temperature
QMC simulation

A. Banerjee *et al.*,
Phys. Rev. Lett. **100**, 047208 (2008)

lines : U(1) lattice gauge theory
at finite temperature

O. Benton *et al.*,
Phys. Rev. B. **83**, 075174 (2012) 



does this have anything to
do with real materials ?



can we hope to see photons in $\text{Yb}_2\text{Ti}_2\text{O}_7$?

estimate of tunnelling matrix element
between ice states in $\text{Yb}_2\text{Ti}_2\text{O}_7$ from
parameters obtained by Ross et al.

$$g_{\text{Yb}_2\text{Ti}_2\text{O}_7} \approx 0.05 \text{ meV.}$$

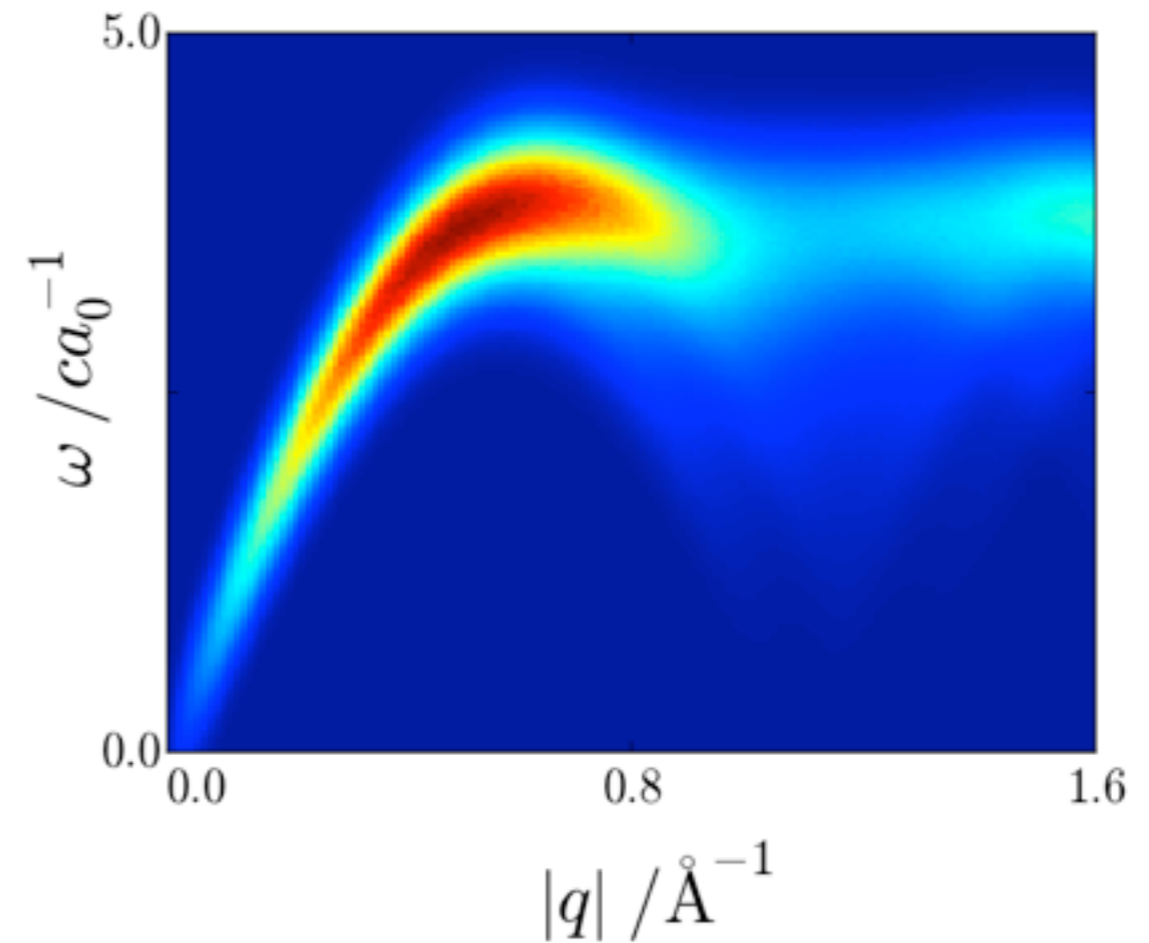
corresponding speed of light

$$c \sim 0.3 \text{ meV \AA} \sim 50 \text{ ms}^{-1}$$

photon bandwidth

$$\Delta\omega \sim 0.1 \text{ meV}$$

this can be resolved !



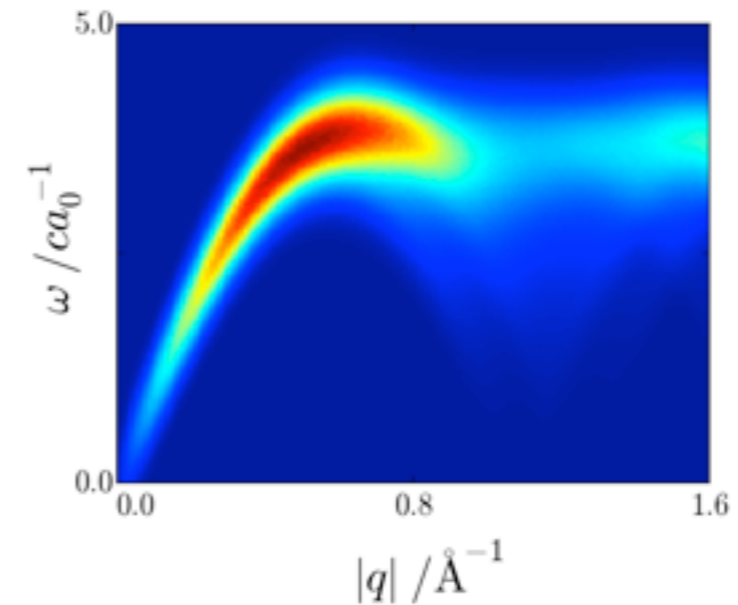
predictions for inelastic neutron
scattering on a powder sample of
 $\text{Yb}_2\text{Ti}_2\text{O}_7$

O. Benton *et al.*, Phys. Rev. B. **83**, 075174 (2012) ☞

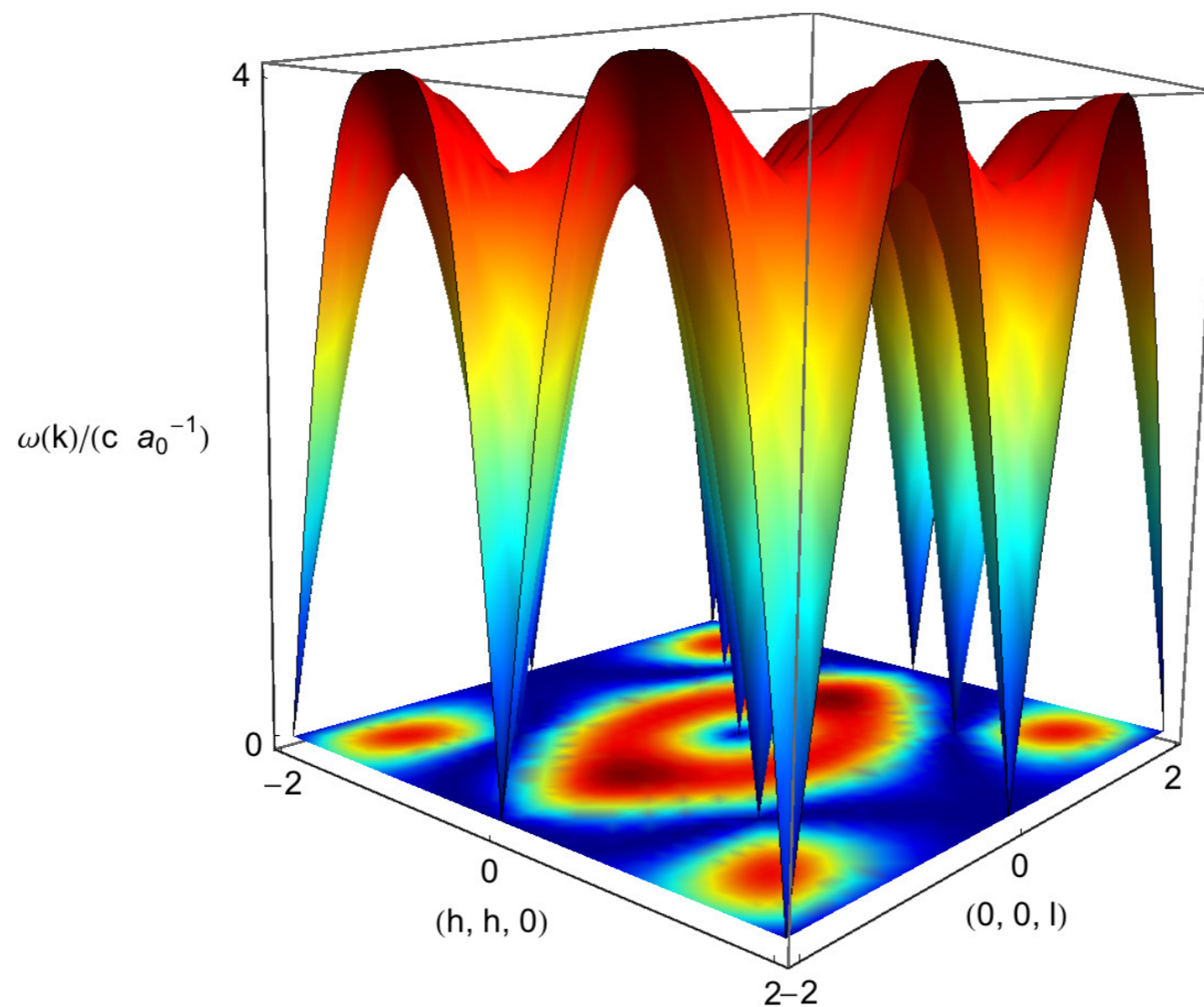


where does this leave us ?

- * **quantum ice** has a (spin) liquid ground state with excitations described by the Maxwell action of classical electromagnetism
- * this state support **photons** which should be visible in neutron scattering experiments
- * a number candidate '**quantum spin ice**' materials are now being intensively investigated...
- * lots of interesting open questions !



thanks for listening !



Owen Benton *et al.*, Phys. Rev. B. **83**, 075174 (2012) 
N.S. *et al.*, Phys. Rev. Lett. **108**, 067204 (2012)

