# Electromagnetism on ice : from quantum spin ice to protons in water ice

nic shannon

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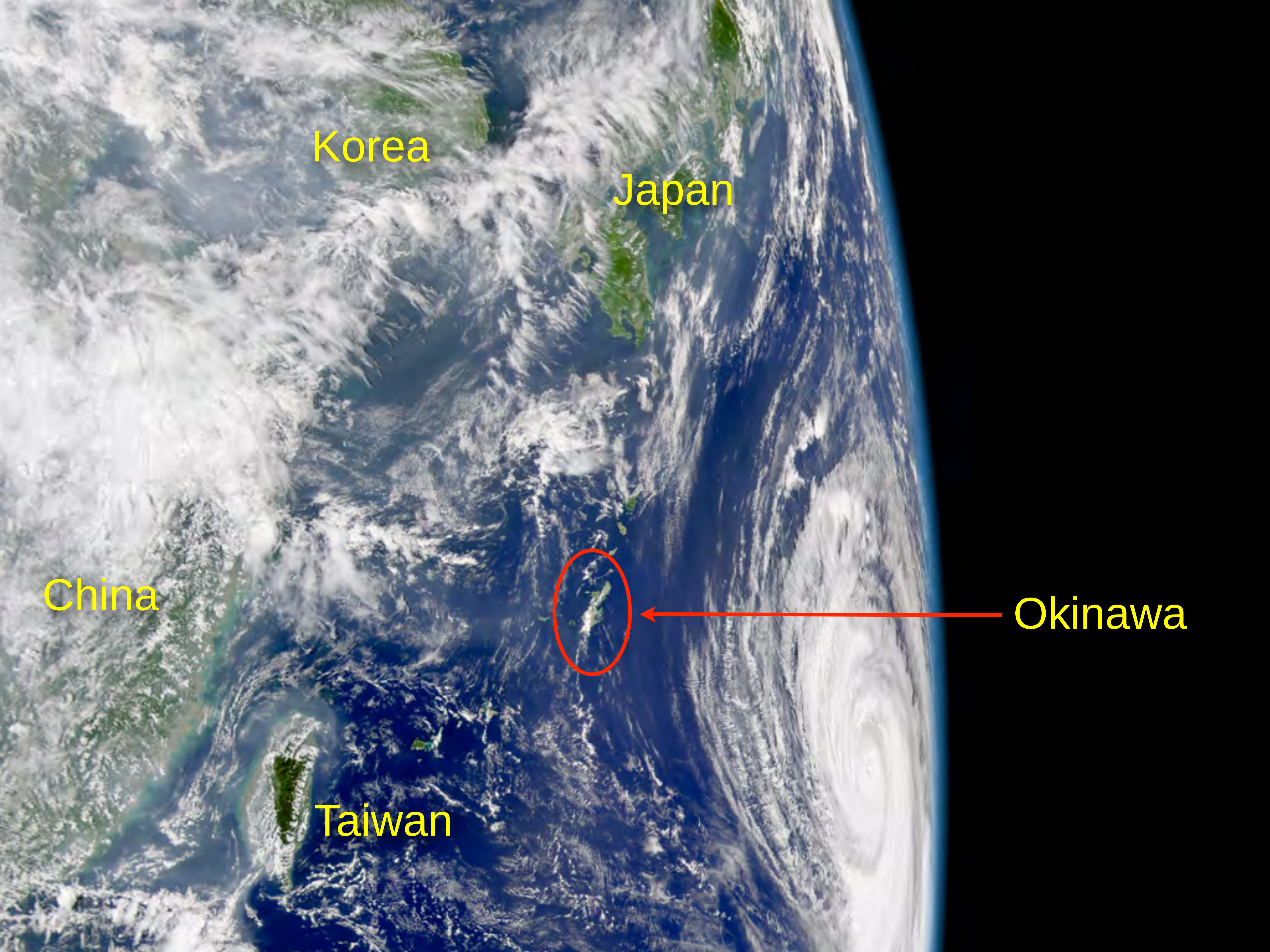
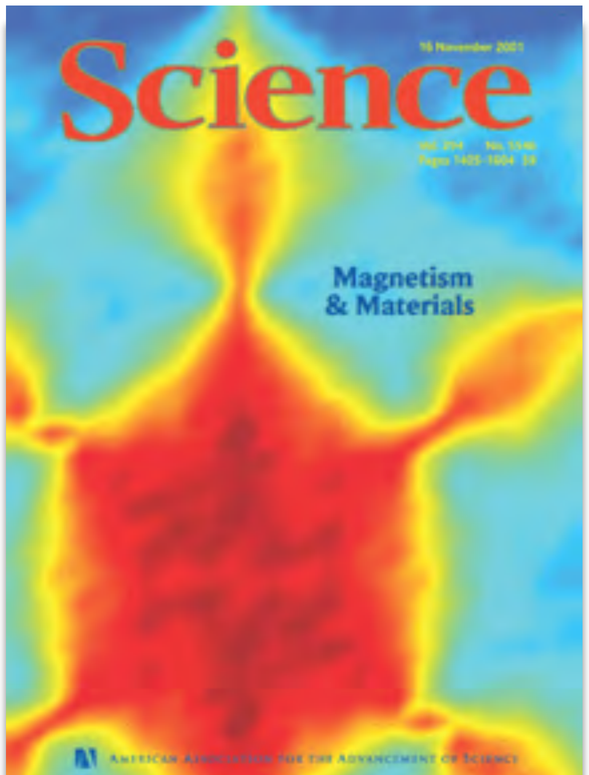

Korea
Japan
China
Okinawa
Taiwan


NQMP 2014
OIST, May 2014.
McMaster

# spin ice and its monopoles...






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

PRL 106, 187202 (2011) PHYSICAL REVIEW LETTERS 6 MAY 2011

**Rods of Neutron Scattering Intensity in $Yb_2Ti_2O_7$: 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]

PHYSICAL REVIEW X 1, 021002 (2011)

**Quantum Excitations in Quantum Spin Ice**

Kate A. Ross,[1] Lucile 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 $Yb_2Ti_2O_7$**

Lieh-Jeng Chang[1,2], Shigeki Onoda[3], Yixi Su[4], Ying-Jer Kao[5], Ku-Ding Tsuei[6], Yukio Yasui[7,8], Kazuhisa Kakurai[2] & Martin Richard Lees[9]

PRL 109, 097205 (2012) PHYSICAL REVIEW LETTERS week ending 31 AUGUST 2012

**Vindication of $Yb_2Ti_2O_7$ 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]

ARTICLE

Received 4 Apr 2014 | Accepted 12 Aug 2014 | Published 18 Sep 2014 DOI: 10.1038/ncomms5970

**Low-energy electrodynamics of novel spin excitations in the quantum spin ice $Yb_2Ti_2O_7$**

LiDong Pan[1], Se Kwon Kim[1], A. Ghosh[1], Christopher M. Morris[1], Kate A. Ross[1,2], Edwin Kermarrec[3], Bruce D. Gaulin[3,4,5], S.M. Koohpayeh[1], Oleg Tchernyshyov[1] & N.P. Armitage[1]

PRL **106,** 187202 (2011) PHYSICAL REVIEW LETTERS 6 MAY 2011

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Kazuhisa Kakurai[2] & Martin Richard Lees[9]

# spin ice goes quantum...

ETTERS week ending 31 AUGUST 2012

ange 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]

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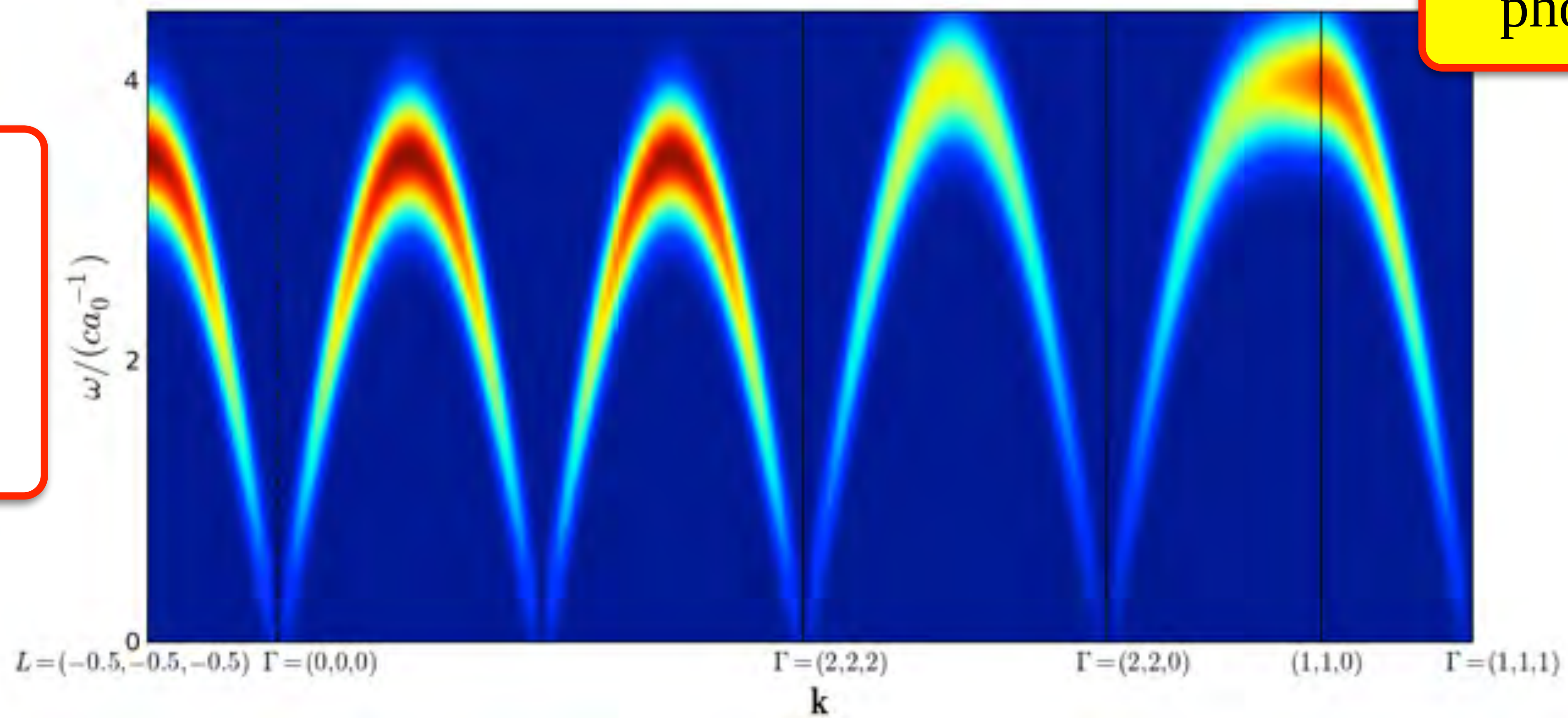
# why is this exciting ?

photons !

prediction for inelastic neutron scattering :




quantum tunnelling between different spin configurations can convert classical spin ice into a quantum spin liquid described by a U(1) lattice gauge theory !


M. Hermele *et al.*, Phys. Rev. B **69**, 064404 (2004)
A. Banerjee *et al.* Phys. Rev. Lett. **100**, 047208 (2008)
N. Shannon *et al.* Phys. Rev. Lett. **108**, 067204 (2012)
O. Benton *et al.*, Phys. Rev. B. **83**, 075174 (2012) ❦
Y. Kato and S. Onoda, arXiv:1411.1918v2

# in the beginning…

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)

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

PRL **103,** 165901 (2009) PHYSICAL REVIEW LETTERS week ending 16 OCTOBER 2009

## Anomalous Proton Dynamics in Ice at Low Temperatures

L. E. Bove,[1] S. Klotz,[1] A. Paciaroni,[2] and F. Sacchetti[2]
[1]IMPMC, CNRS UMR 7590, Université P&M Curie, F-75252

PRL **112,** 148302 (2014) PHYSICAL REVIEW LETTERS 11 APRIL 2014

## Quantum Simulation of Collective Proton Tunneling in Hexagonal Ice Crystals

Christof Drechsel-Grau* and Dominik Marx†

nature physics LETTERS
PUBLISHED ONLINE: 16 FEBRUARY 2015 | DOI: 10.1038/NPHYS3225

## Direct visualization of concerted proton tunnelling in a water nanocluster

Xiangzhi Meng[1†], Jing Guo[1†], Jinbo Peng[1†], Ji Chen[1], Zhichang Wang[1], Jun-Ren Shi[1,2], Xin-Zheng Li[3,2], En-Ge Wang[1,2*] and Ying Jiang[1,2*]

## Coordinated proton tunnelling in a cyclic network of four hydrogen bonds in the solid state

Dermot F. Brougham*, Roberto Caciuffo† & Anthony J. Horsewill*

PRL 103, 165901 (2009) PHYSICAL REVIEW LETTERS week ending 16 OCTOBER 2009

**Anomalous Proton Dynamics in Ice at Low Temperatures**

L. E. Bove,[1] S. Klotz,[1] A. Paciaroni,[2] and F. Sacchetti[2]

PRL 112, 14830

Quan

IL 2014

# growing evidence that quantum effects matter in water ice

nature physics

**Direct visualization of concerted proton tunnelling in a water nanocluster**

Xiangzhi Meng[1†], Jing Guo[1†], Jinbo Peng[1†], Ji Chen[1], Zhichang Wang[1], Jun-Ren Shi[1,2], Xin-Zheng Li[3,2], En-Ge Wang[1,2★] and Ying Jiang[1,2★]

**nnelling**

**four hydrogen bonds in the solid state**

Dermot F. Brougham*, Roberto Caciuffo† & Anthony J. Horsewill*

OIST OKINAWA INSTITUTE OF SCIENCE AND TECHNOLOGY GRADUATE UNIVERSITY

# could water ice be a "spin liquid" ?!

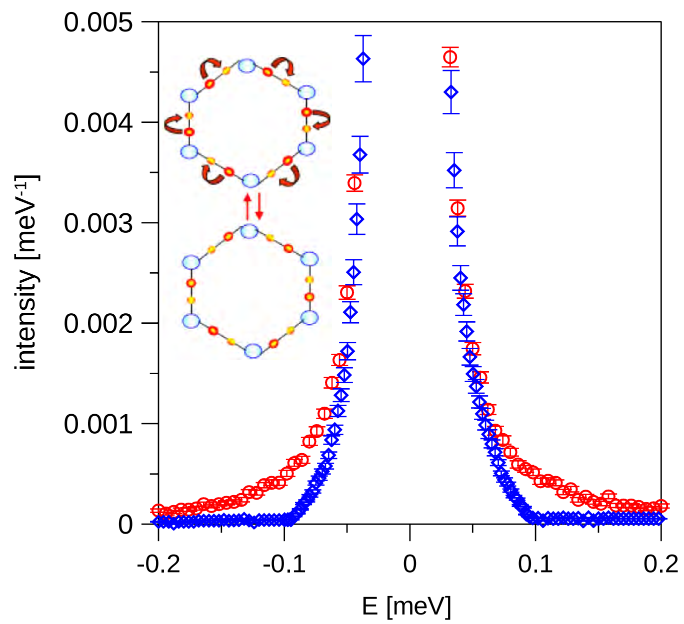
evidence for collective excitations of protons from inelastic neutron scattering

+

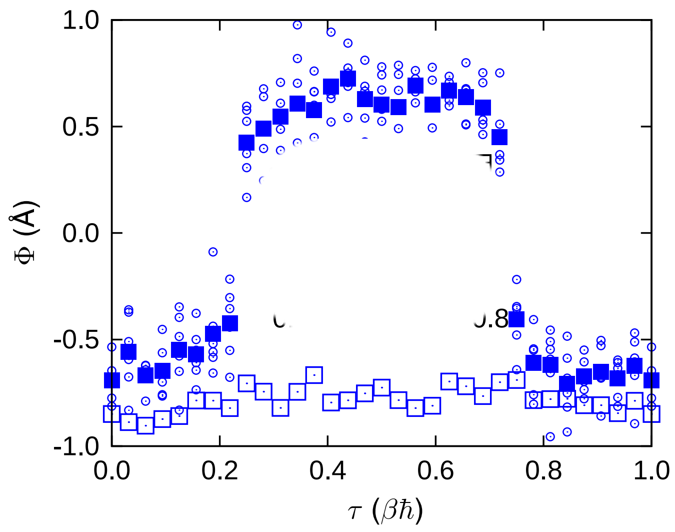
evidence for collective tunnelling of protons from ab initio calculations

=

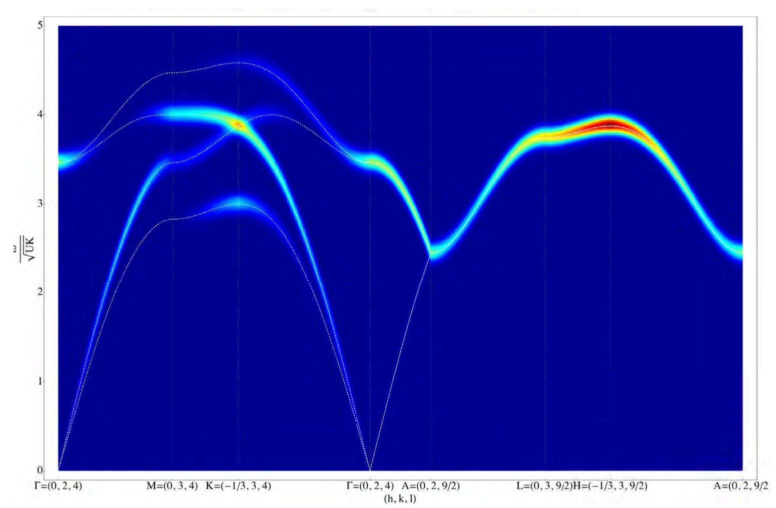
lattice gauge theory with bi-refringent photons




L.E. Bove *et al.*,
Phys. Rev. Lett **103**, 165901 (2009)




C. Drechsel-Grau and D. Marx,
Phys. Rev. Lett **112**, 148302 (2014)



O. Benton, O. Sikora and NS,
arXiv:1504.04158v1

# wouldn't have happened without…

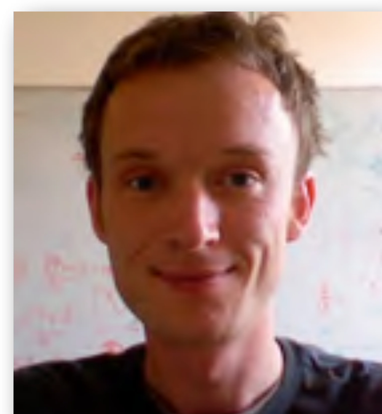
**Owen Benton**
Bristol/OIST

**Peter Fulde**
MPI-PKS

**Paul McClarty**
RAL

**Roderich Moessner**
MPI-PKS

**Karlo Penc**
Budapest



**Frank Pollmann**
MPI-PKS

**Olga Sikora**
NTU

EPSRC Engineering and Physical Sciences Research Council

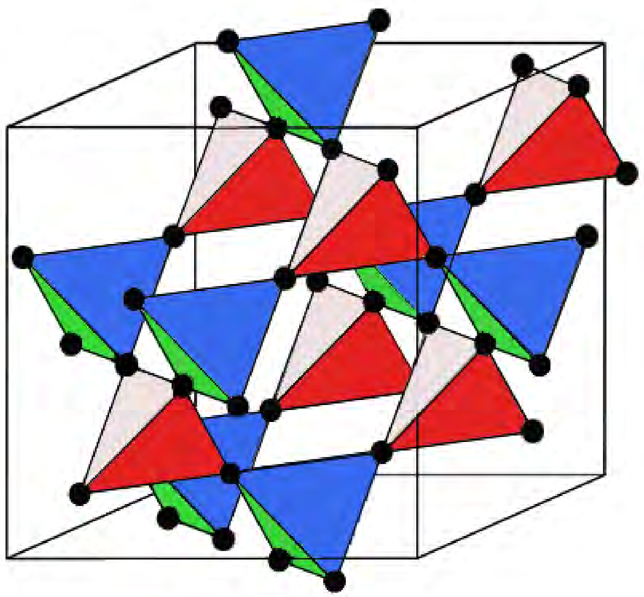
OTKA

OIST OKINAWA INSTITUTE OF SCIENCE AND TECHNOLOGY GRADUATE UNIVERSITY

# what is spin ice ?

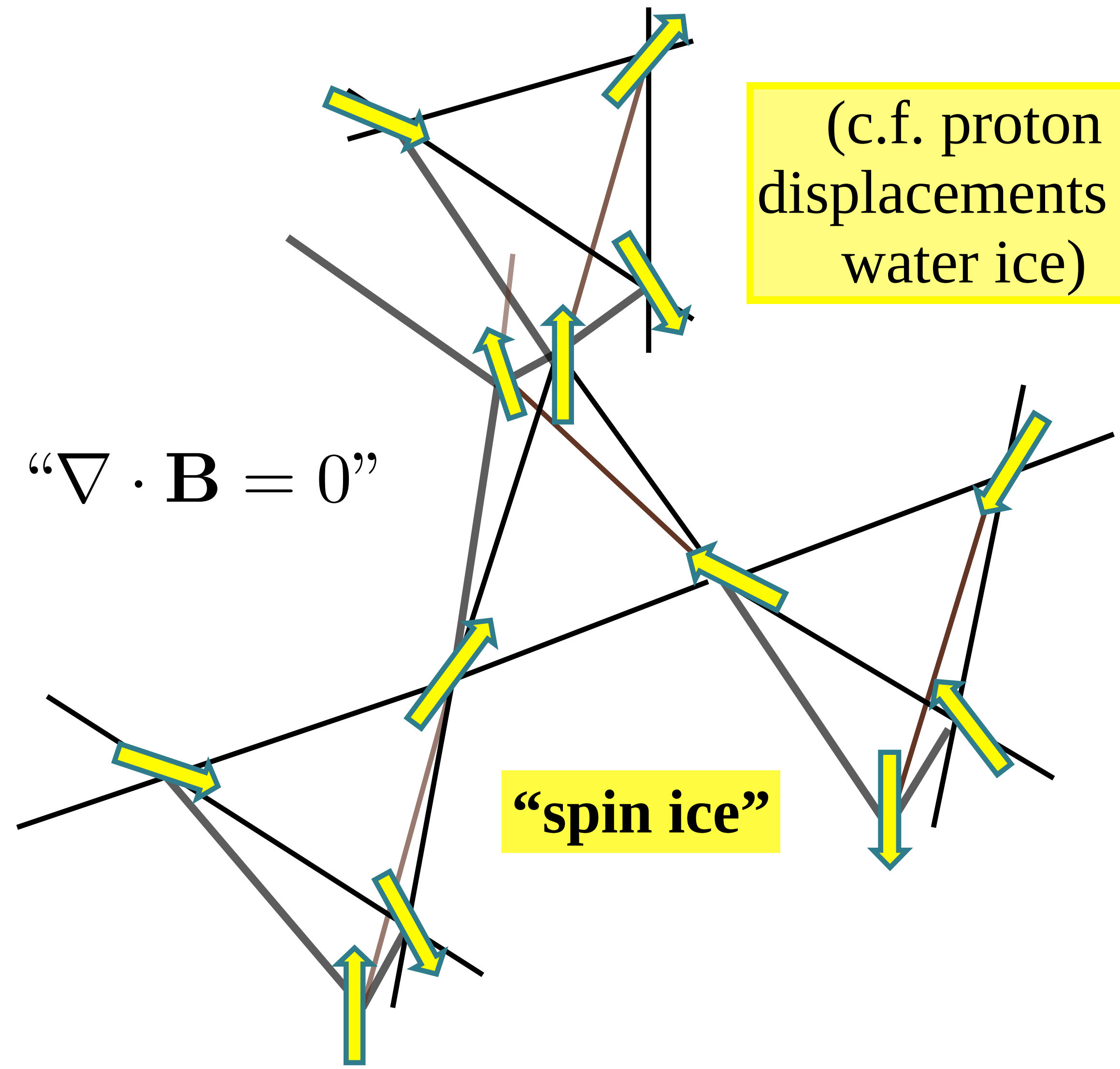
$Ho_2Ti_2O_7$
$Dy_2Ti_2O_7$


**ferromagnetic nearest-neighbour interactions** select an **extensive** number of states with **two in and two out** spins per tetrahedron



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

“$\nabla \cdot \mathbf{B} = 0$”




(c.f. proton displacements in water ice)

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

M.J. Harris et al., Phys. Rev. Lett. **79**, 2554 (1997)

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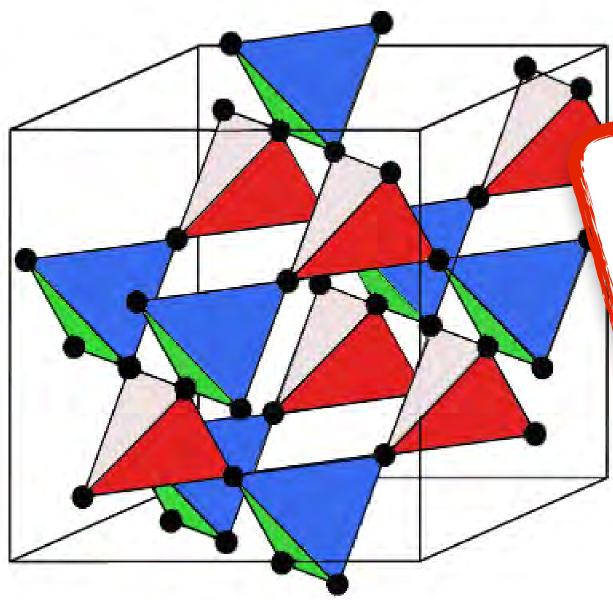
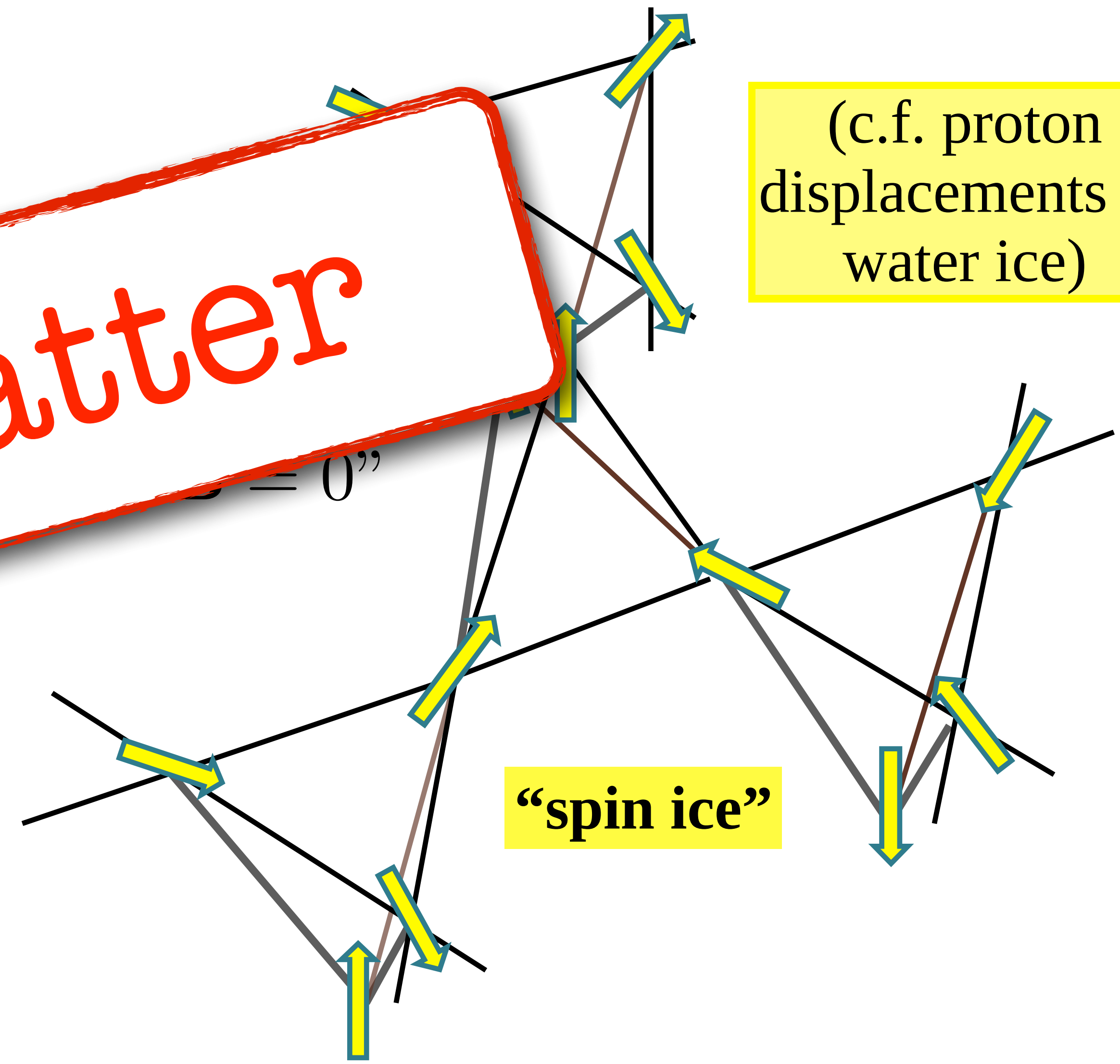


**...lore lattice**

= 0”

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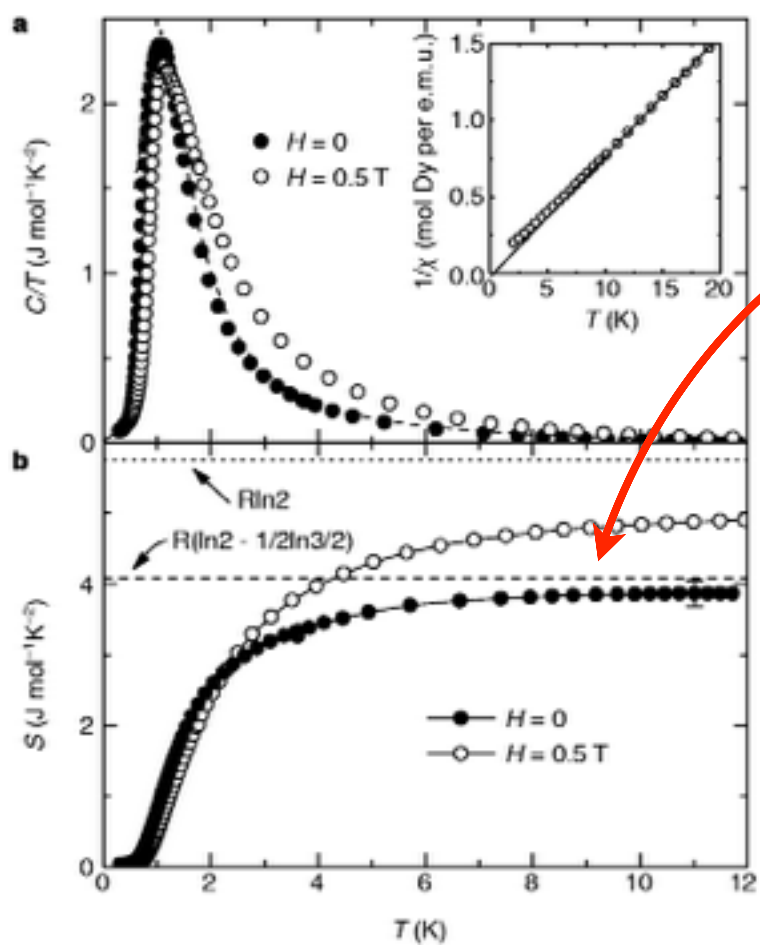
# why should you believe in spin ice ?

heat capacity of $Dy_2Ti_2O_7$

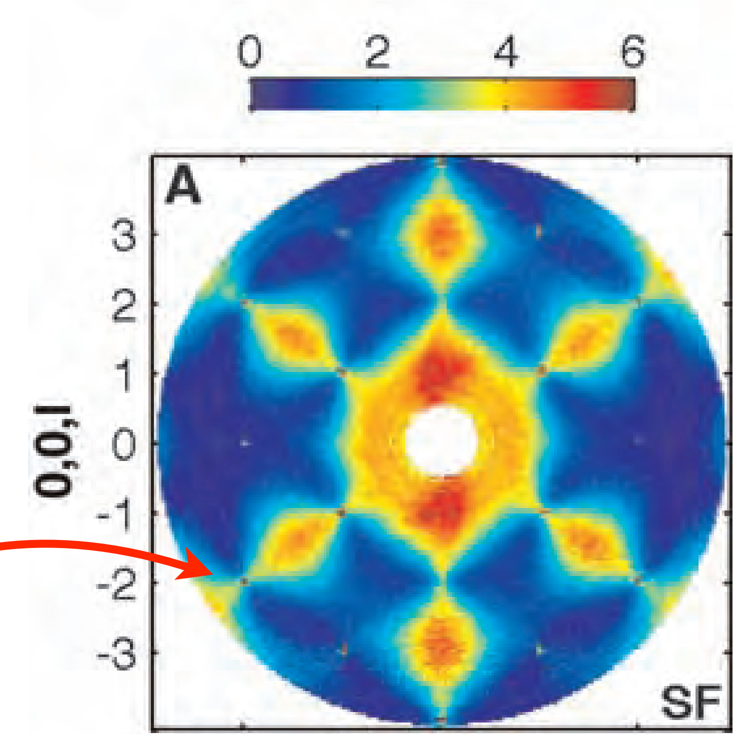



A.P. Ramirez et al.,
Nature **399**, 333 (1999)

Pauling Ice entropy

pinch point
“$\nabla \cdot \mathbf{B} = 0$”

neutron scattering on $Ho_2Ti_2O_7$




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

direct (thermodynamic) and indirect (scattering) evidence for extensive ground state manifold

PRL 106, 187202 (2011) PHYSICAL REVIEW LETTERS 6 MAY 2011

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PHYSICAL REVIEW X 1, 021002 (2011)

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PRL 109, 097205 (2012) PHYSICAL REVIEW LETTERS week ending 31 AUGUST 2012

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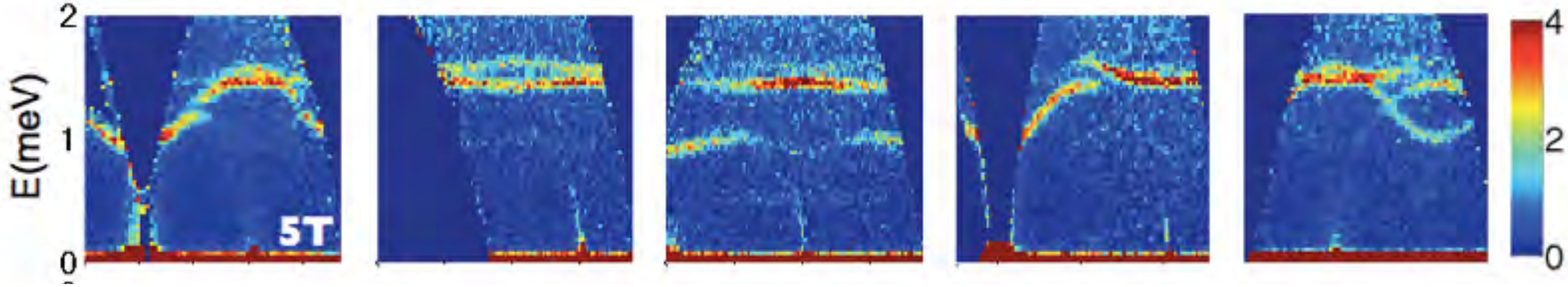
PRL 109, 017201 (2012) PHYSICAL REVIEW LETTERS 6 JULY 2012

**Power-Law Spin Correlations in the Pyrochlore Antiferromagnet $Tb_2Ti_2O_7$**

T. Fennell,[1,*] M. Kenzelmann,[2] B. Roessli,[1] M. K. Haas,[3] and R. J. Cava[3]

# what makes $Yb_2Ti_2O_7$ a “quantum spin ice” ?

inelastic neutron scattering in magnetic field sees dispersing excitations




these are well-described by a model with anisotropic exchange interactions

$$
\begin{aligned}
\mathcal{H}_{\text{ex}} \;=\; \sum_{\langle ij\rangle} \Big\{ & J_{zz}\mathsf{S}_i^z\mathsf{S}_j^z - J_{\pm}(\mathsf{S}_i^+\mathsf{S}_j^- + \mathsf{S}_i^-\mathsf{S}_j^+) \\
& + J_{\pm\pm}\left[\gamma_{ij}\mathsf{S}_i^+\mathsf{S}_j^+ + \gamma_{ij}^*\mathsf{S}_i^-\mathsf{S}_j^-\right] \\
& + J_{z\pm}\left[\mathsf{S}_i^z(\zeta_{ij}\mathsf{S}_j^+ + \zeta_{ij}^*\mathsf{S}_j^-) + i \leftrightarrow j\right] \Big\}
\end{aligned}
$$

$$
\begin{aligned}
J_{zz} &= 0.17 \\
J_{\pm} &= 0.05 \\
J_{\pm\pm} &= 0.05 \\
J_{z\pm} &= -0.14
\end{aligned}
$$

dominant term is an Ising-like interaction - cf. spin ice

Kate A. Ross *et al.*, Phys. Rev. X **1**, 021002 (2011)

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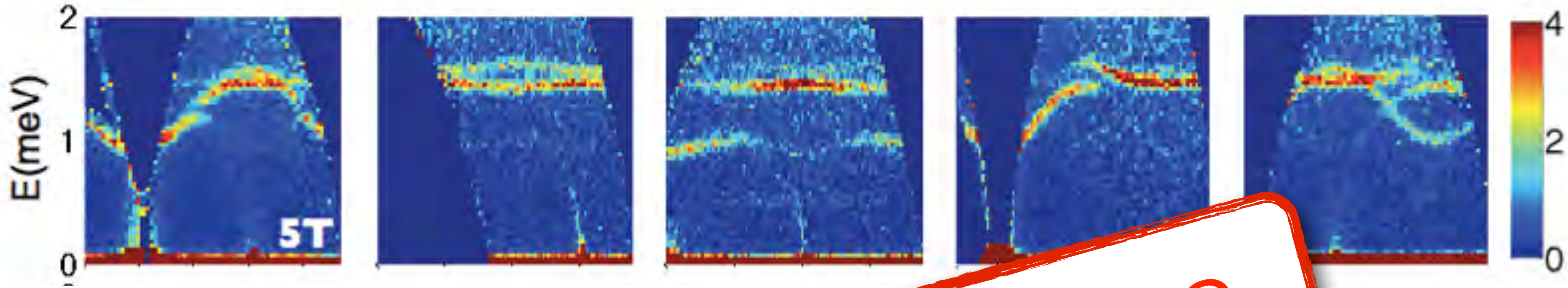



these are well-described by a ... eractions

(LS Matter)$^2$

$$\mathcal{H}_{\text{ex}} = \sum_{\langle ij \rangle} \Big\{ J_{zz} \ldots$$

$$+J_{\pm\pm} \left[ \gamma_{ij} \ldots \right]$$

$$+J_{z\pm} \left[ \mathsf{S}_i^z (\zeta_{ij} \mathsf{S}_j^+ + \zeta_{ij}^* \mathsf{S}_j^-) + i \leftrightarrow j \right] \Big\}$$

$$J_{zz} = 0.17$$
$$J_{\pm} = 0.05$$
$$J_{\pm\pm} = 0.05$$
$$J_{z\pm} = -0.14$$

dominant term is an Ising-like interaction - cf. spin ice

Kate A. Ross *et al.*, Phys. Rev. X **1**, 021002 (2011)

PHYSICAL REVIEW B **69**, 064404 (2004)

# 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 Balents[1]
[1]*Department of Physics, University of California, Santa Barbara, California 93106-9530, USA*
[2]*Kavli Institute for Theoretical Physics, University of California, Santa Barbara, California 93106-4030, USA*

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

$$\mathcal{H}_{\mathsf{XXZ}} = J_{zz} \sum_{\langle ij \rangle} \mathsf{S}_i^z \mathsf{S}_j^z - J_\pm \sum_{\langle ij \rangle} (\mathsf{S}_i^+ \mathsf{S}_j^- + \mathsf{S}_i^- \mathsf{S}_j^+)$$

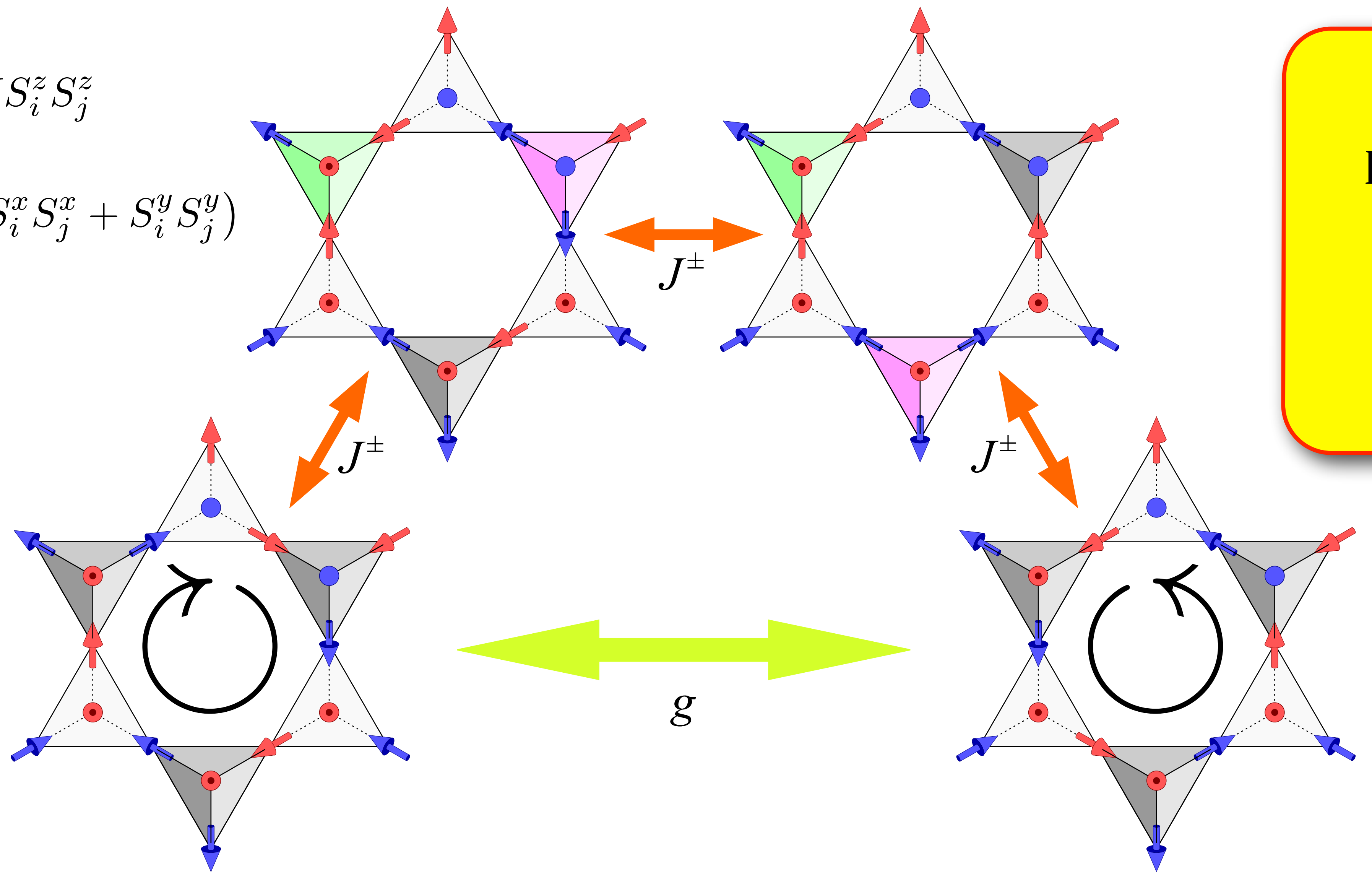
strong anisotropy

$$J_{zz} \gg J_\pm$$

selects ice manifold

# how does quantum tunnelling work ?

$$\mathcal{H}_{\mathsf{xxz}} = J_{zz} \sum_{\langle ij \rangle} J S_i^z S_j^z - J_{\pm} \sum_{\langle ij \rangle} \left( S_i^x S_j^x + S_i^y S_j^y \right)$$




degenerate perturbation theory

$$g = \frac{12 J_{\pm}^3}{J_{zz}^2}$$

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

PHYSICAL REVIEW B **69**, 064404 (2004)

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Michael Hermele,[1] Matthew P. A. Fisher,[2] and Leon Balents[1]
[1]*Department of Physics, University of California, Santa Barbara, California 93106-9530, USA*
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S=1/2 easy-axis magnet on a pyrochlore lattice...

$$\mathcal{H}_{\mathsf{XXZ}} = J_{zz} \sum_{\langle ij \rangle} \mathsf{S}_i^z \mathsf{S}_j^z - J_{\pm} \sum_{\langle ij \rangle} (\mathsf{S}_i^+ \mathsf{S}_j^- + \mathsf{S}_i^- \mathsf{S}_j^+)$$

strong anisotropy

$$J_{zz} \gg J_{\pm}$$

selects ice manifold

Hamiltonian acting on ice states...

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

degenerate perturbation theory

$$g = \frac{12 J_{\pm}^3}{J_{zz}^2}$$

...extra term makes model exactly soluble for μ=g

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PHYSICAL REVIEW B **69**, 064404 (2004)

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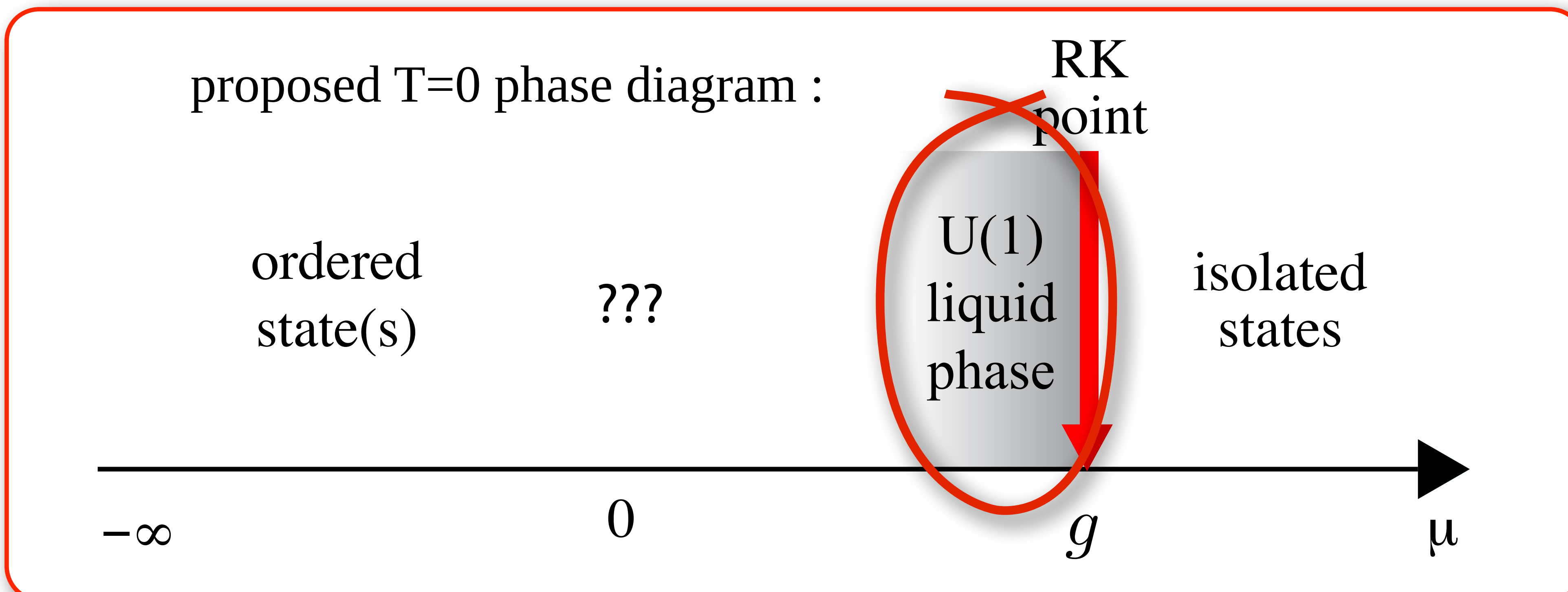
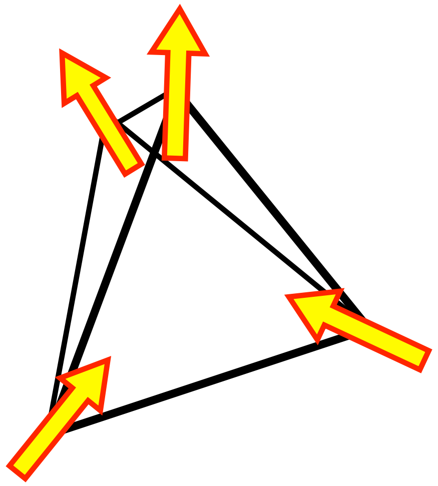
$$\mathcal{H}_\mu = -g \sum_{\hexagon} |\circlearrowright\rangle\langle\circlearrowleft| + |\circlearrowleft\rangle\langle\circlearrowright| + \mu \sum_{\hexagon} |\circlearrowright\rangle\langle\circlearrowright| + |\circlearrowleft\rangle\langle\circlearrowleft|$$

…argue for U(1)-liquid phase, based on properties of **exactly soluble point** μ=g




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

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



$\nabla . \mathbf{B} = 0$ ...by **explicit construction**

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

quantum ice has **local dynamics** : $\mathcal{H} = -g \sum_{\hexagon} |\circlearrowright\rangle\langle\circlearrowleft| + |\circlearrowleft\rangle\langle\circlearrowright|$

tunneling between ice states ⇒ gauge field varies in time

simplest guess for effective field theory in a liquid phase is **Maxwell** action :

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

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

PRL **100**, 047208 (2008) PHYSICAL REVIEW LETTERS week ending 1 FEBRUARY 2008

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

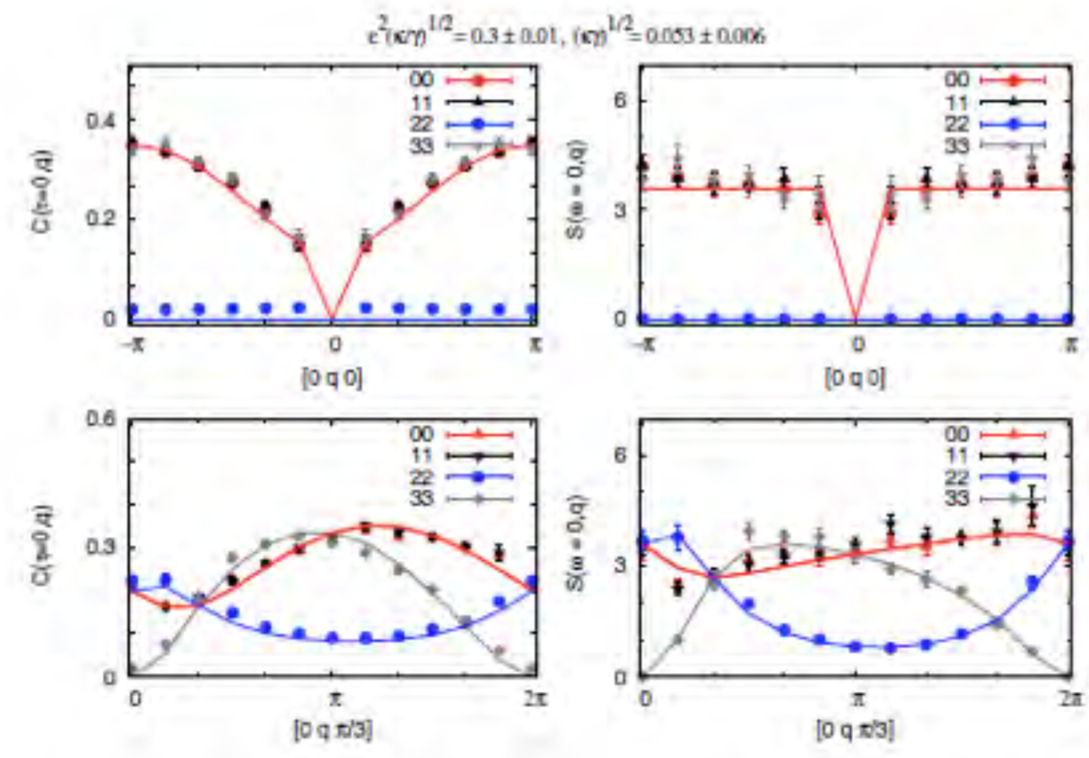
Argha Banerjee,[1] Sergei V. Isakov,[2] Kedar Damle,[1] and Yong Baek Kim[2]

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

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

quantum charge ice with tunneling

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




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

PRL **100,** 047208 (2008) PHYSICAL REVIEW LETTERS week ending 1 FEBRUARY 2008

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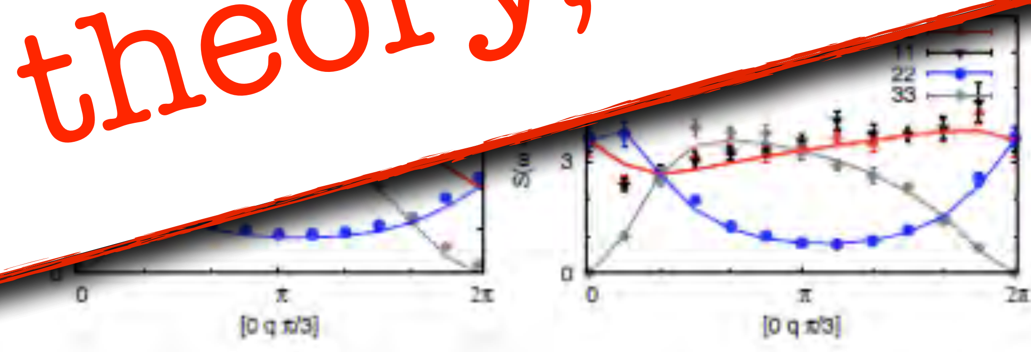
consider hard-core Bosons with ...
strong nearest nei...
inte...

q... ...ce with tunneling

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

quantitive agreement with prediction of U(1) gauge theory, T>0




finite-temperature correlation functions, calculated using QMC [T ≈ g], and compared to the predictions of a U(1) gauge theory

OIST OKINAWA INSTITUTE OF SCIENCE AND TECHNOLOGY GRADUATE UNIVERSITY

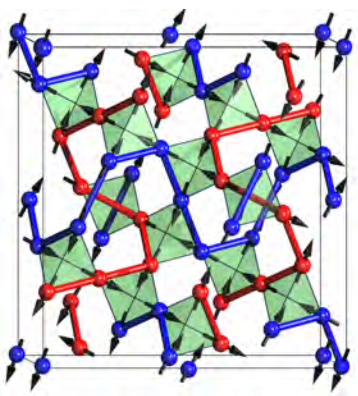
PRL **108,** 067204 (2012) PHYSICAL REVIEW LETTERS week ending 10 FEBRUARY 2012

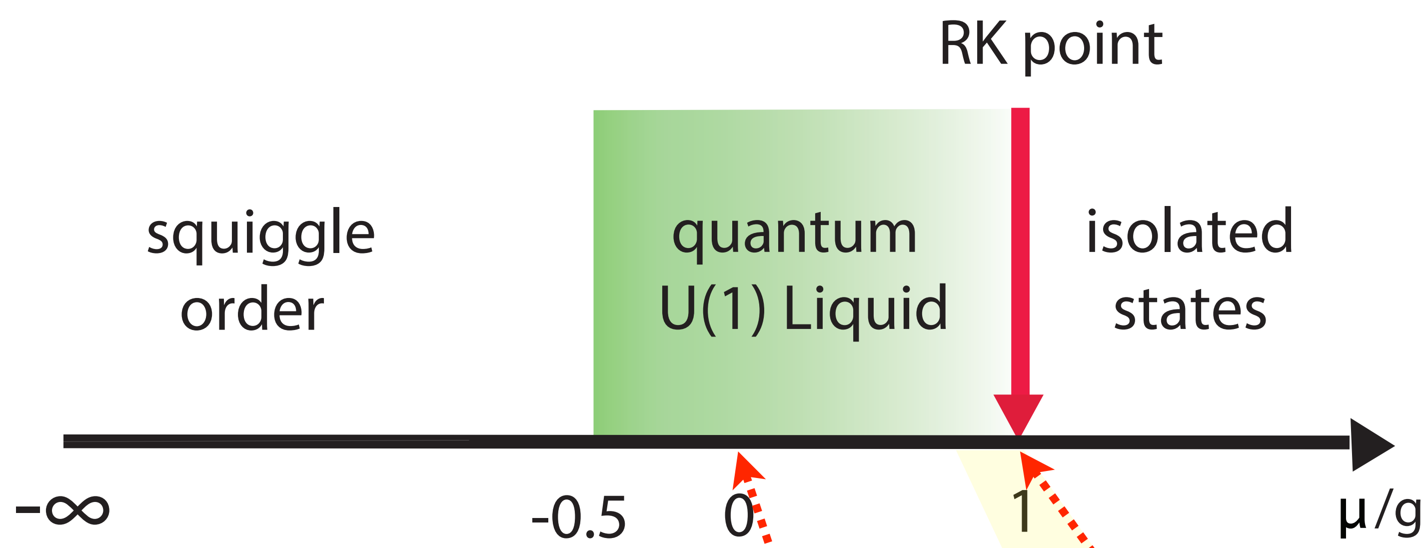
**Quantum Ice: A Quantum Monte Carlo Study**

Nic Shannon,[1] Olga Sikora,[1] Frank Pollmann,[2] Karlo Penc,[3] and Peter Fulde[2,4]

consider minimal model with RK-point, acting on spin-ice states…

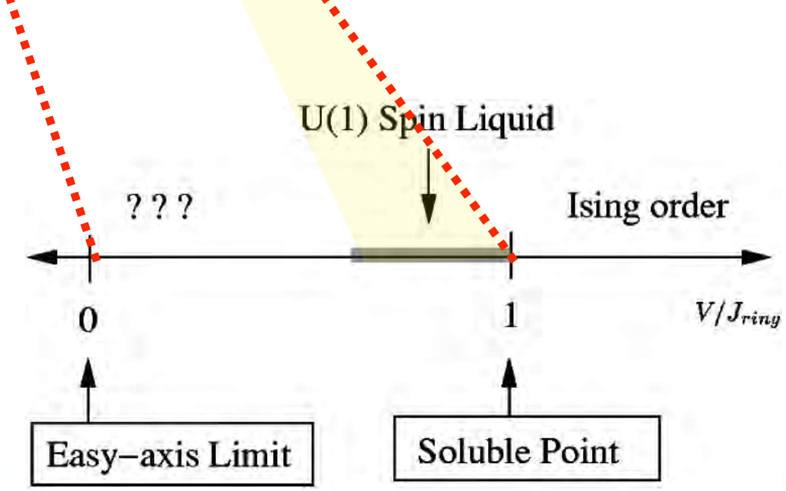
$$\mathcal{H}_\mu = -g \sum_{\hexagon} |\circlearrowright\rangle\langle\circlearrowleft| + |\circlearrowleft\rangle\langle\circlearrowright| + \mu \sum_{\hexagon} |\circlearrowright\rangle\langle\circlearrowright| + |\circlearrowleft\rangle\langle\circlearrowleft|$$






ice entropy absorbed into unique quantum ground state

cf. M. Hermele *et al.*, Phys. Rev. B **69**, 064404 (2012)

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PRL **108,** 067204 (2012) PHYSICAL REVIEW LETTERS week ending 10 FEBRUARY 2012

**Quantum Ice: A Quantum Monte Carlo Study**

Nic Shannon,[1] Olga Sikora,[1] Frank Pollmann,[2] Karlo Penc,[3] and Peter Fulde[2,4]

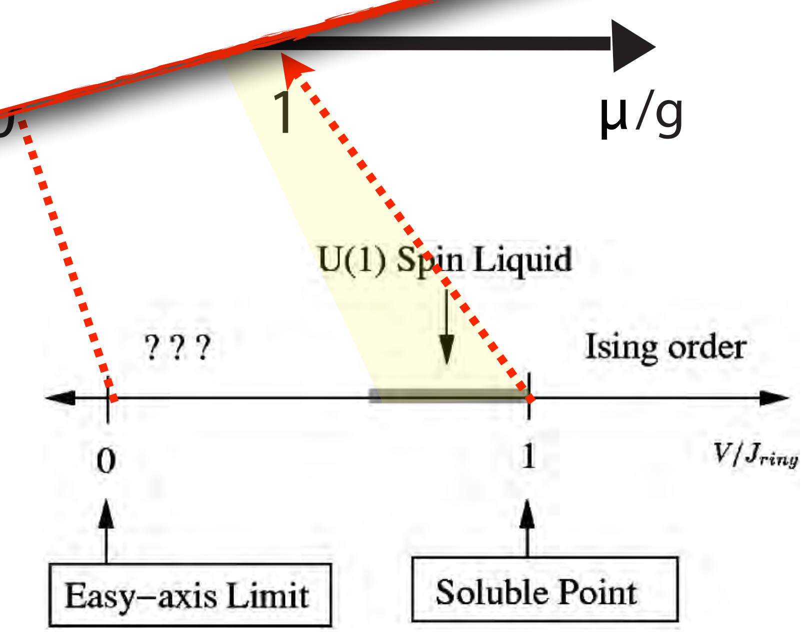
consider minimal model with RK-point

$$\mathcal{H}_\mu = -g \sum | \circlearrowright \rangle \langle$$

quantitive agreement with prediction of U(1) gauge theory, T=0

ic…
abs…
uniqu…
ground state

cf. M. Hermele *et al.*, Phys. Rev. B **69**, 064404 (2012)

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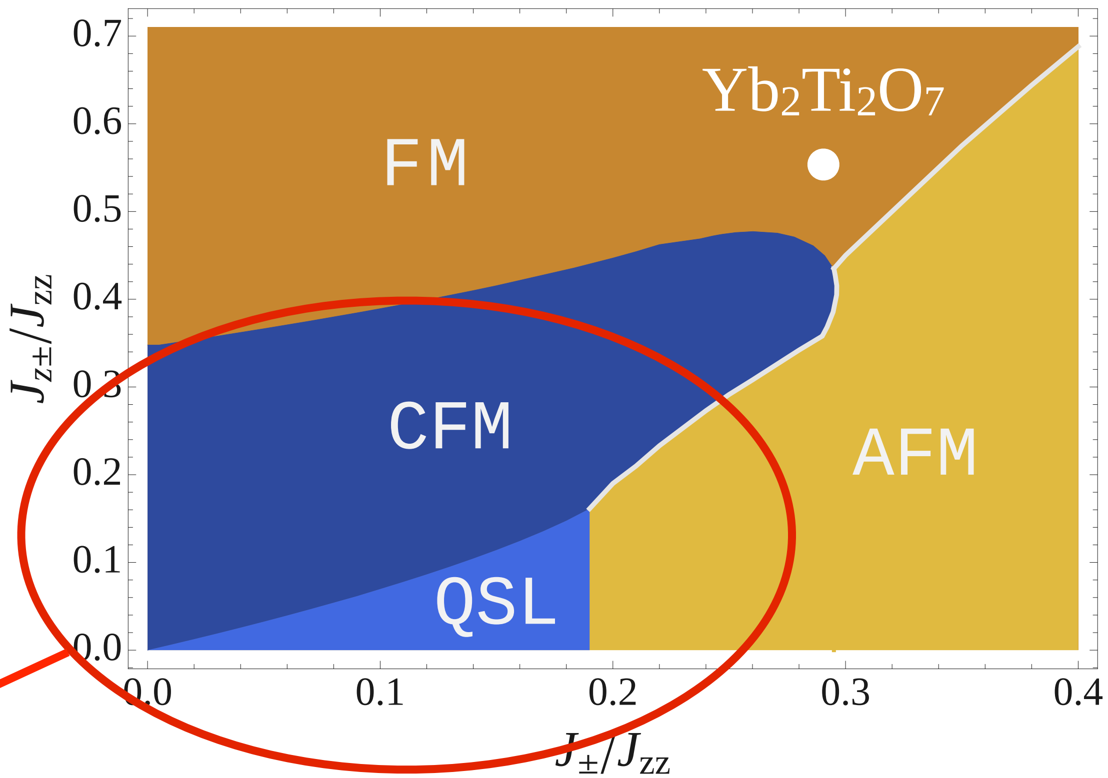
# valid for more general interactions ?

general model for exchange
between spin-1/2 ions
on a pyrochlore lattice has 4
independent parameters

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

phase diagram within
"Gauge mean field theory" :




quantum spin liquids derived
from spin ice

L. Savary and L. Balents,
Phys. Rev. Lett **108**, 037202 (2012)

N.B. see also :
S. Onoda *et al.*, Phys. Rev. B **83**, 094411 (2011)
S.-B. Lee *et al.*, Phys. Rev. B **86**, 104412 (2012)

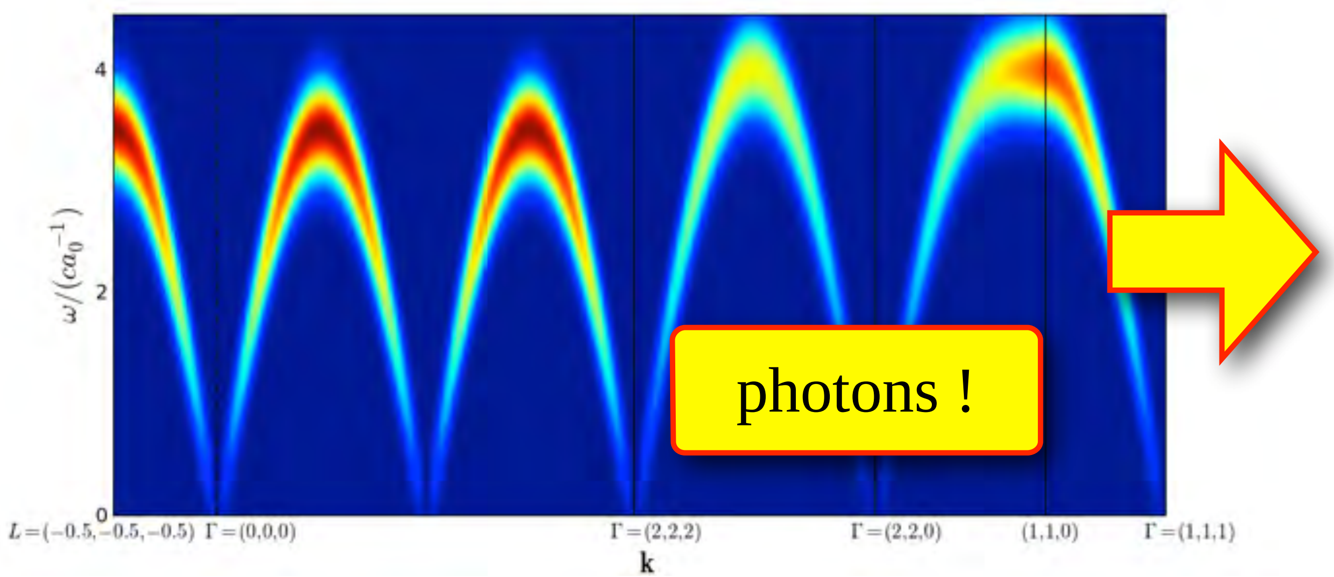
# what would this look like in experiment ?

consider minimal model for a quantum spin ice…

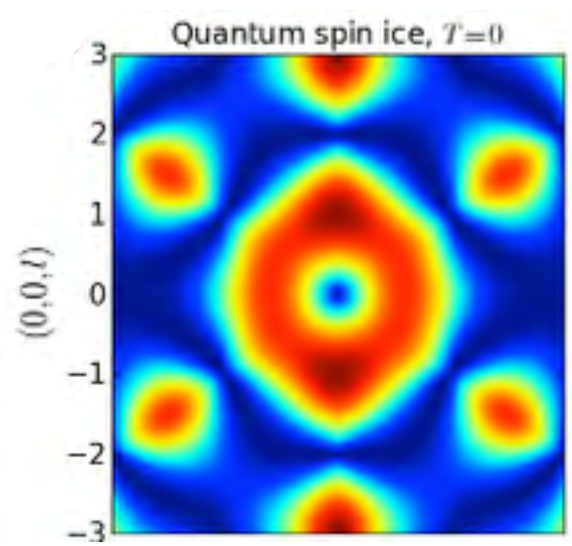
$$\mathcal{H}_{\text{tunneling}} = -g \sum_{\hexagon} |\circlearrowright\rangle\langle\circlearrowleft| + |\circlearrowleft\rangle\langle\circlearrowright|$$ (acting on spin-ice states)

…parameterize lattice gauge theory from quantum Monte Carlo simulation

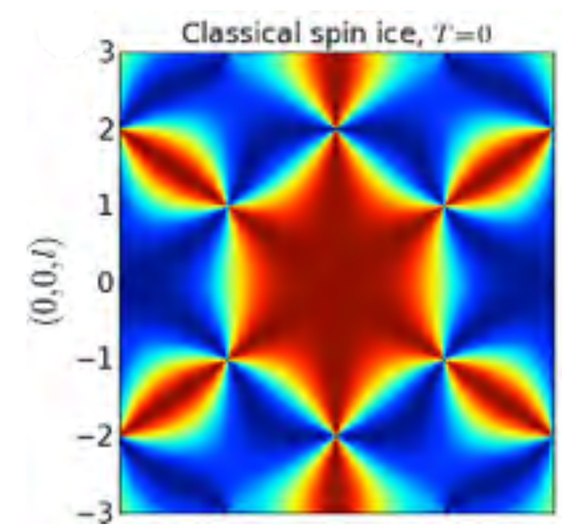
prediction for inelastic neutron scattering




prediction for quasi-elastic neutron scattering




v.




pinch points are suppressed !

O. Benton *et al.*, Phys. Rev. B. **86**, 075174 (2012) ❦

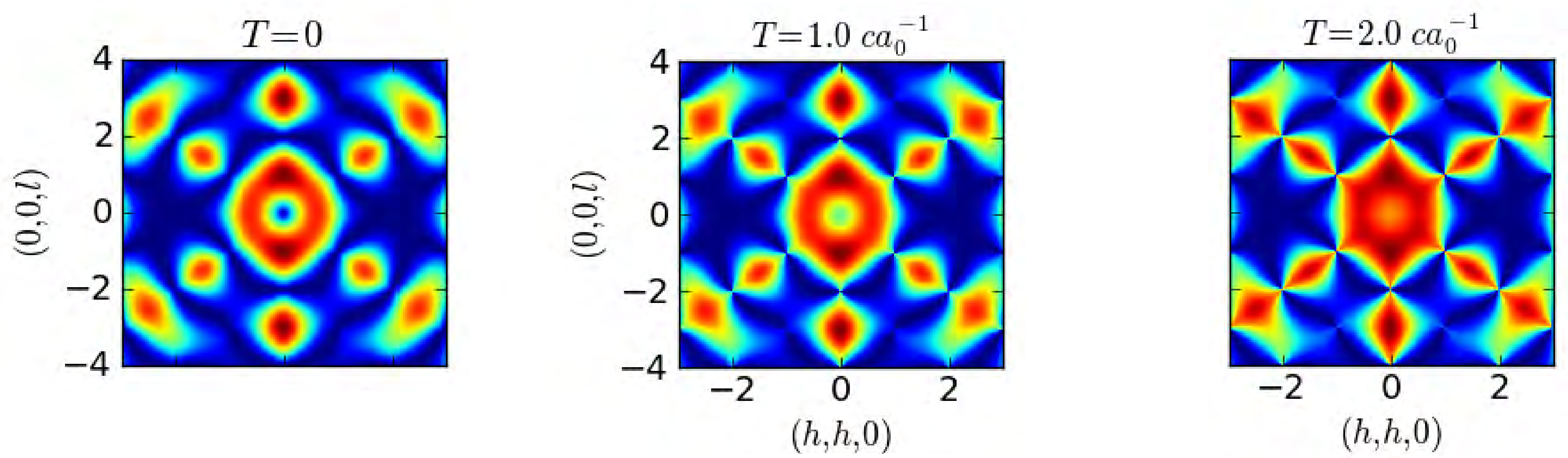
# how does this connect with (classical) spin-ice ?

simplest scenario is a **crossover**, controlled by the **thermal excitation of photons**

i.e. see classical correlations for $\mathbf{q} \ll \frac{1}{\lambda_T}$ where **thermal de Broglie wavelength** $\lambda_T = \frac{\pi c}{T}$

thermal correction to quantum correlations at low T :

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




...i.e. pinch-points are progressively restored at finite T

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

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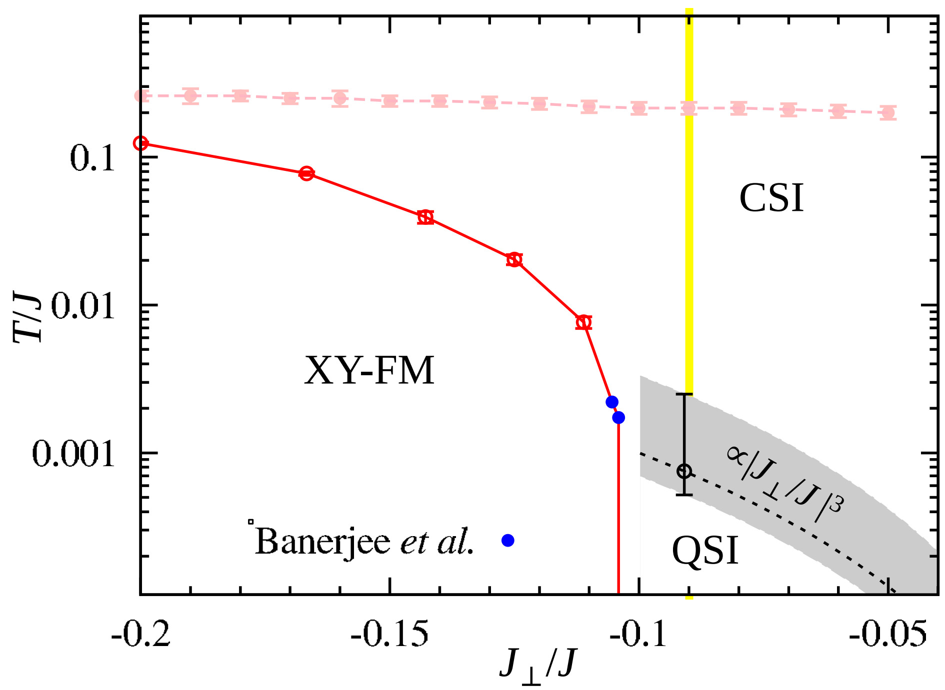
# is this seen in simulation ?

$$\mathcal{H} = \sum_{\langle ij \rangle} \left[ J S_i^z S_j^z + J_\perp \left( S_i^x S_j^x + S_i^y S_j^y \right) \right]$$

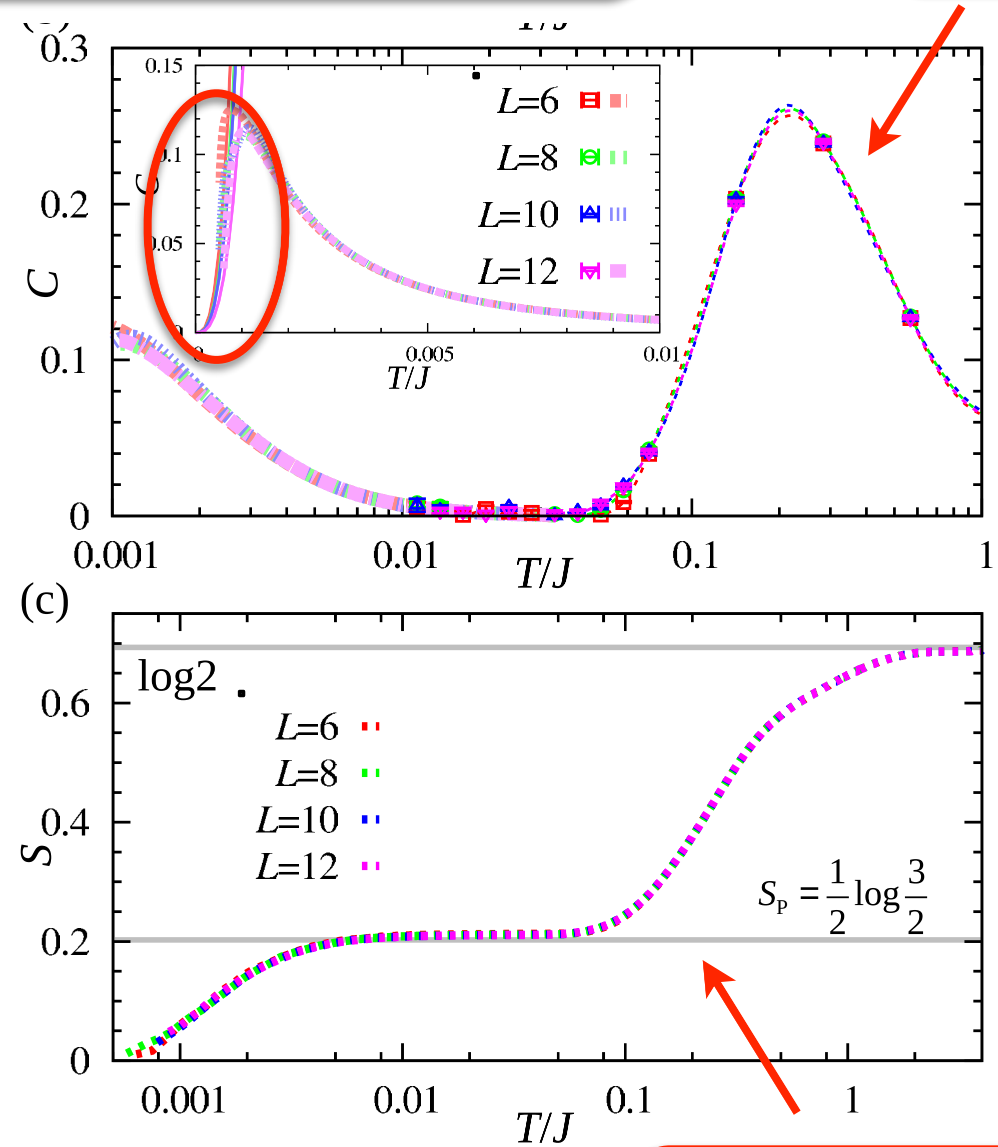
…finite-temperature QMC




$J_\perp/J = -1/11$

T^3 at low-T ⇒ photons ✓

spin-ice peak

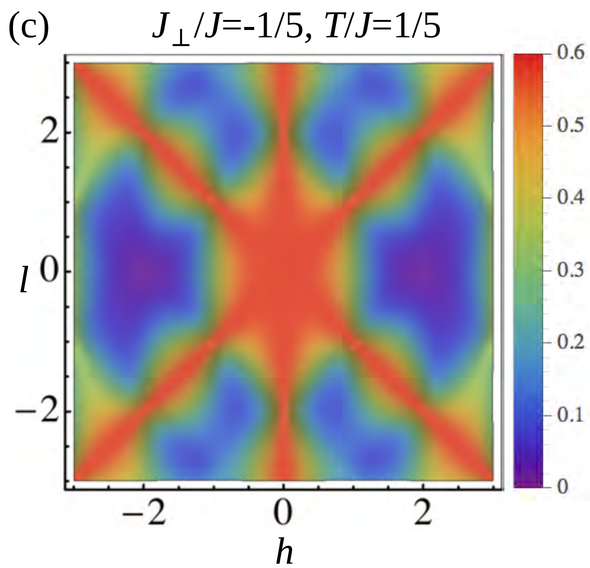


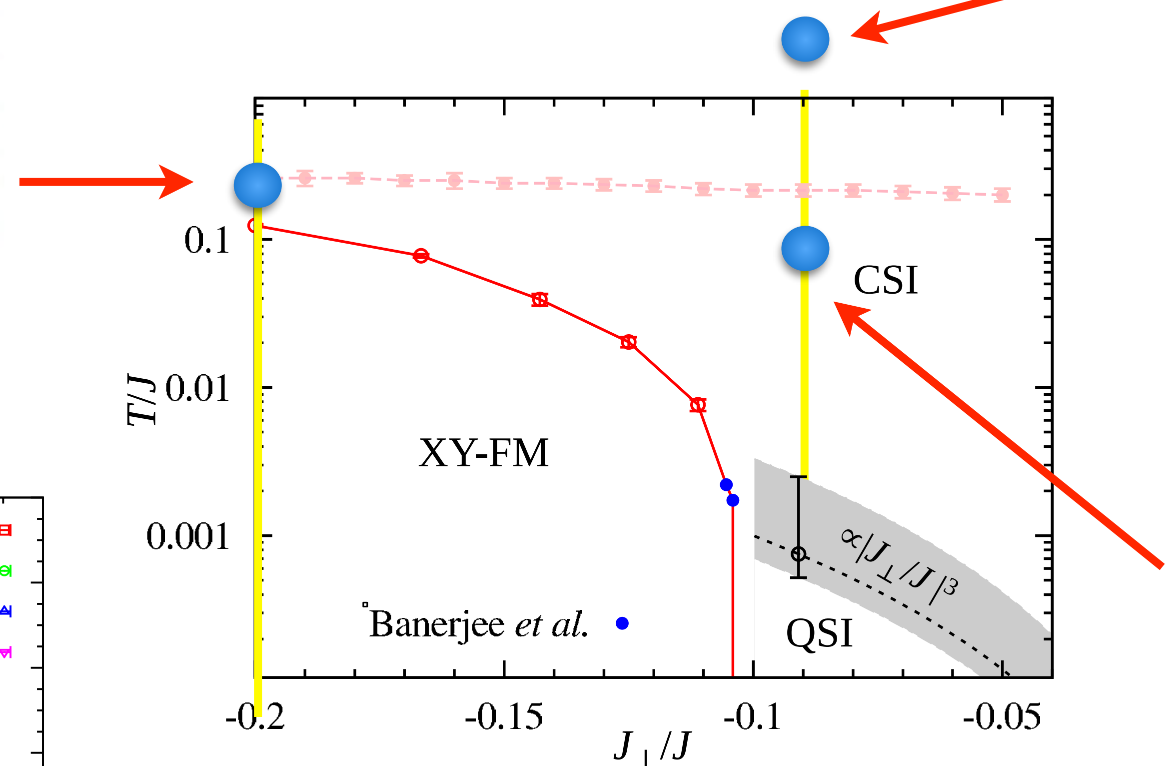

spin-ice entropy

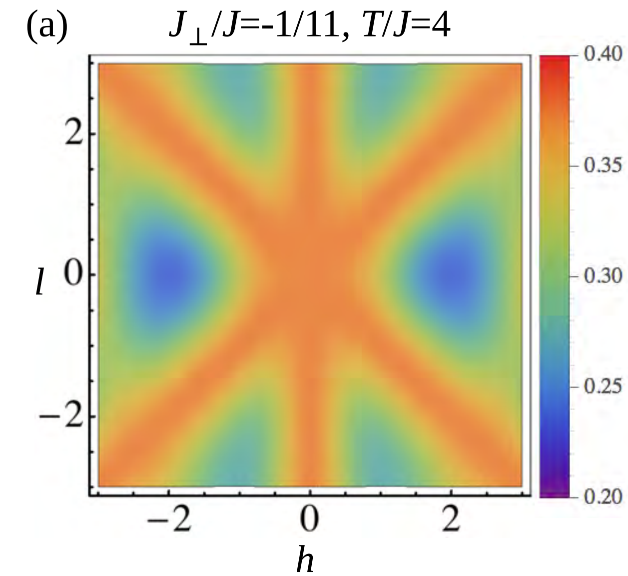
Y. Kato and S. Onoda, arXiv:1411.1981v1

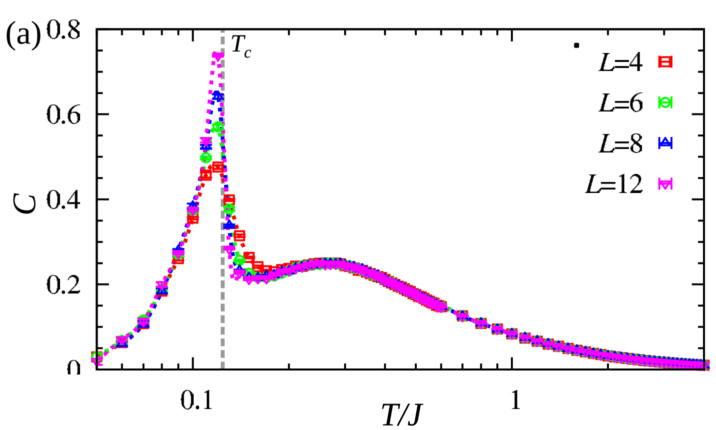
cf. Banerjee *et al*. Phys. Rev. Lett. **100**, 047208 (2008)

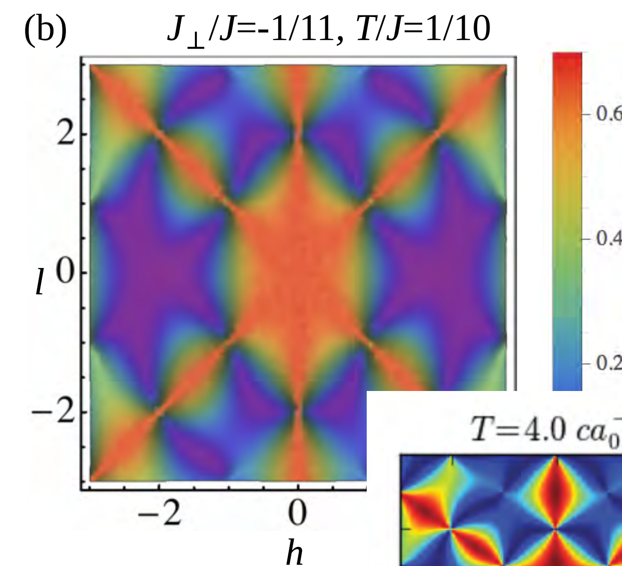
# is this seen in simulation ?

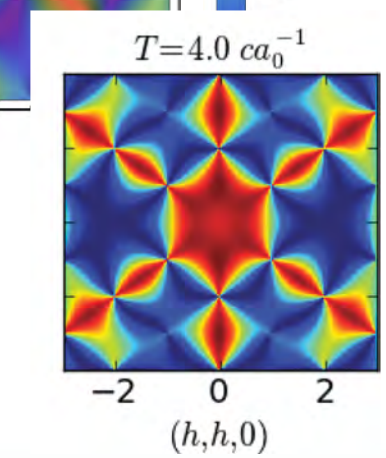


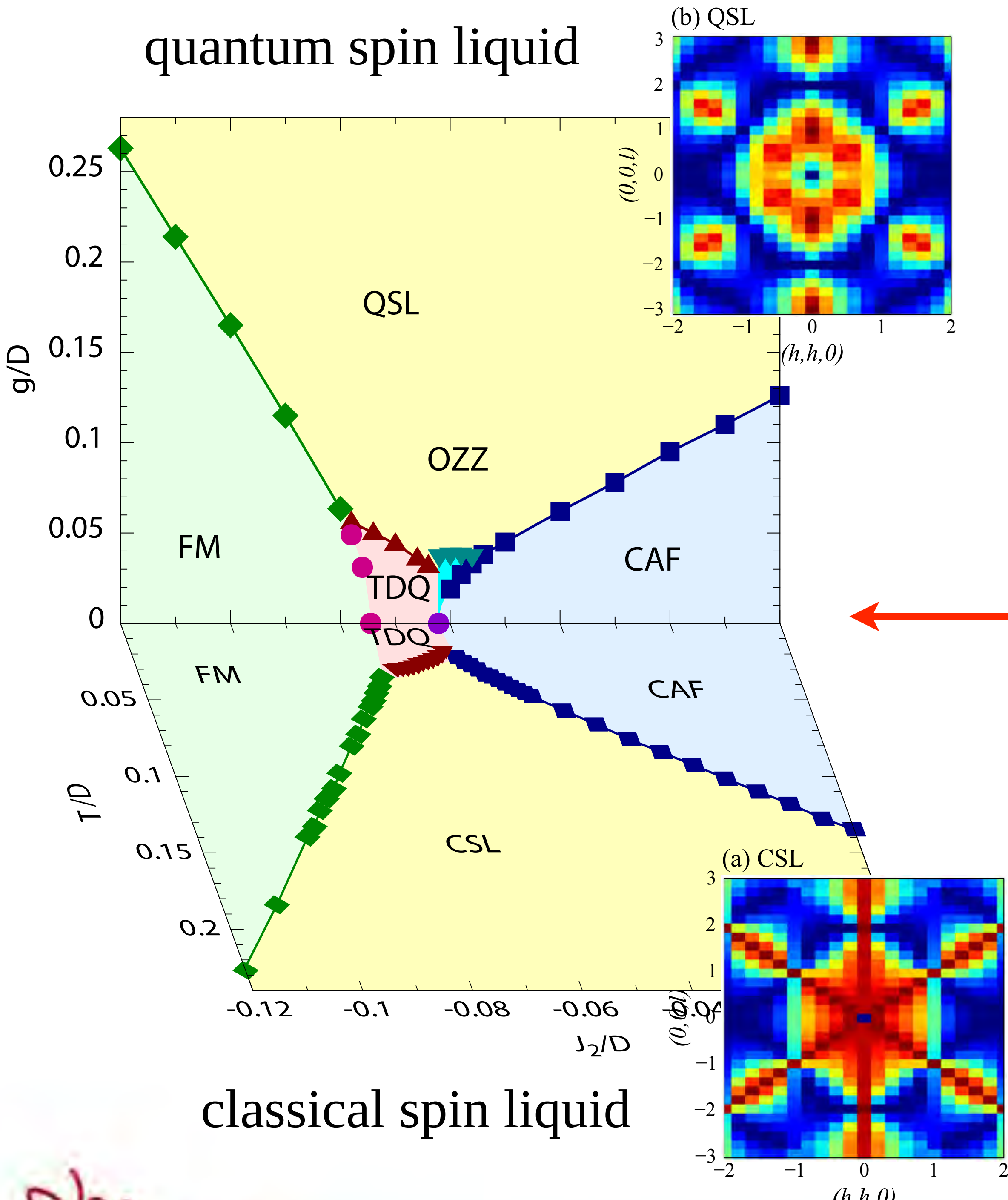



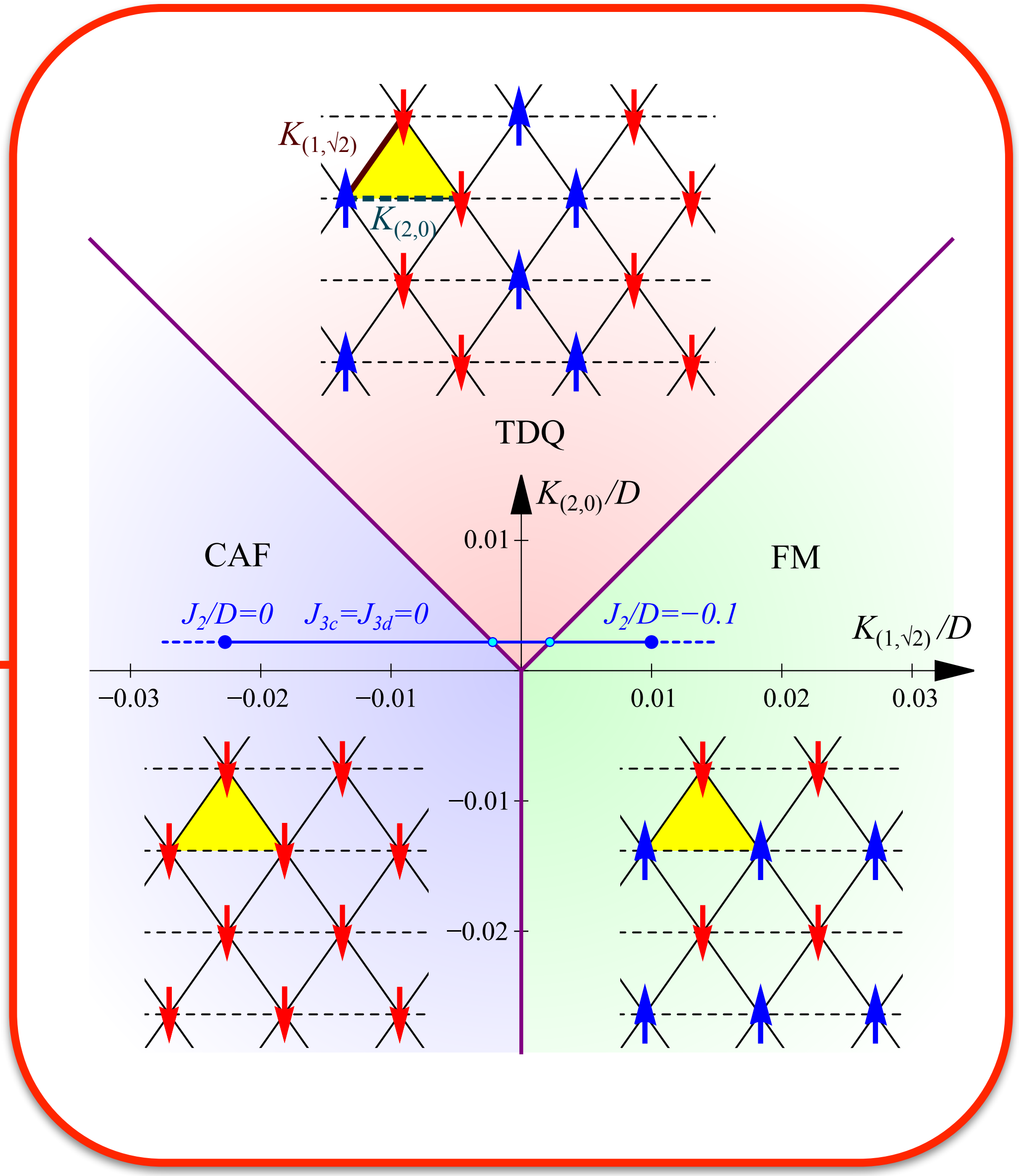

Y. Kato and S. Onoda, arXiv:1411.1981v1

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

# what about dipolar interactions ?




ordered ground states




T=0, g=0

P. McClarty *et al.*, arXiv.1410.0451v1

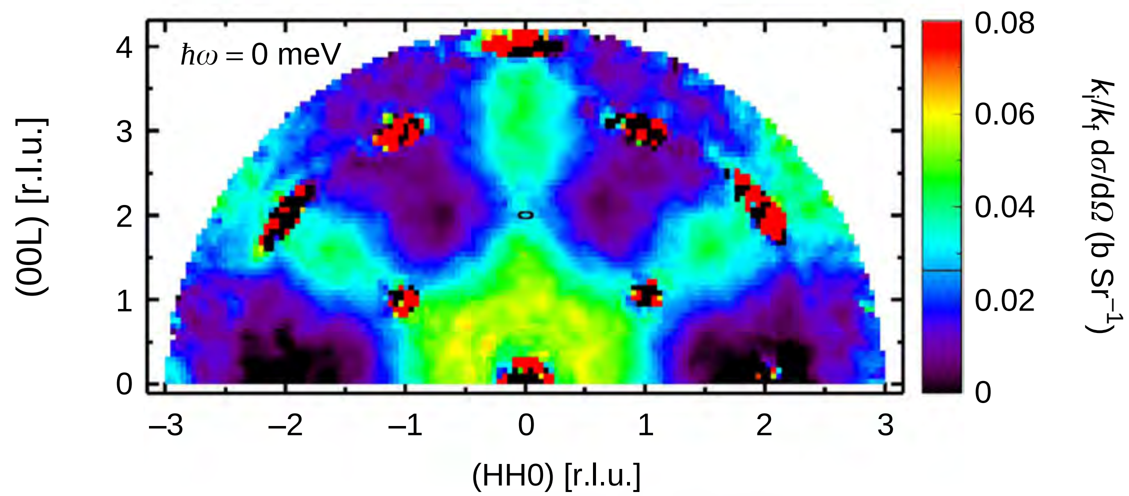
ARTICLE

Received 20 Sep 2012 | Accepted 18 Apr 2013 | Published 17 Jun 2013

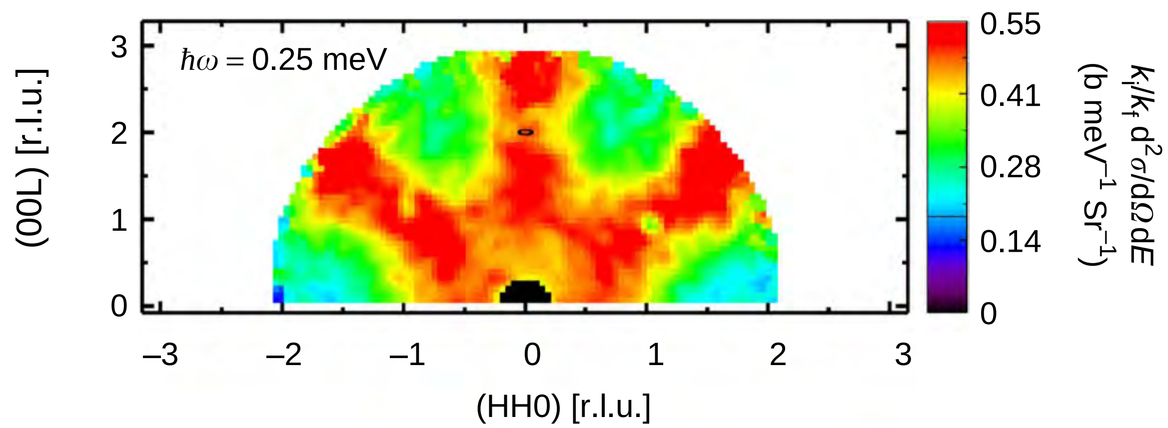
DOI: 10.1038/ncomms2914

# Quantum fluctuations in spin-ice-like $Pr_2Zr_2O_7$

K. Kimura[1], S. Nakatsuji[1,2], J.-J. Wen[3], C. Broholm[3,4,5], M.B. Stone[5], E. Nishibori[6] & H. Sawa[6]




elastic scattering suggests spin-ice correlations




but most scattering is inelastic - evidence of quantum monopole-dynamics ?

K. Kimura *et al*., Nat. Communs. **4**, 1934 (2013)

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ARTICLE

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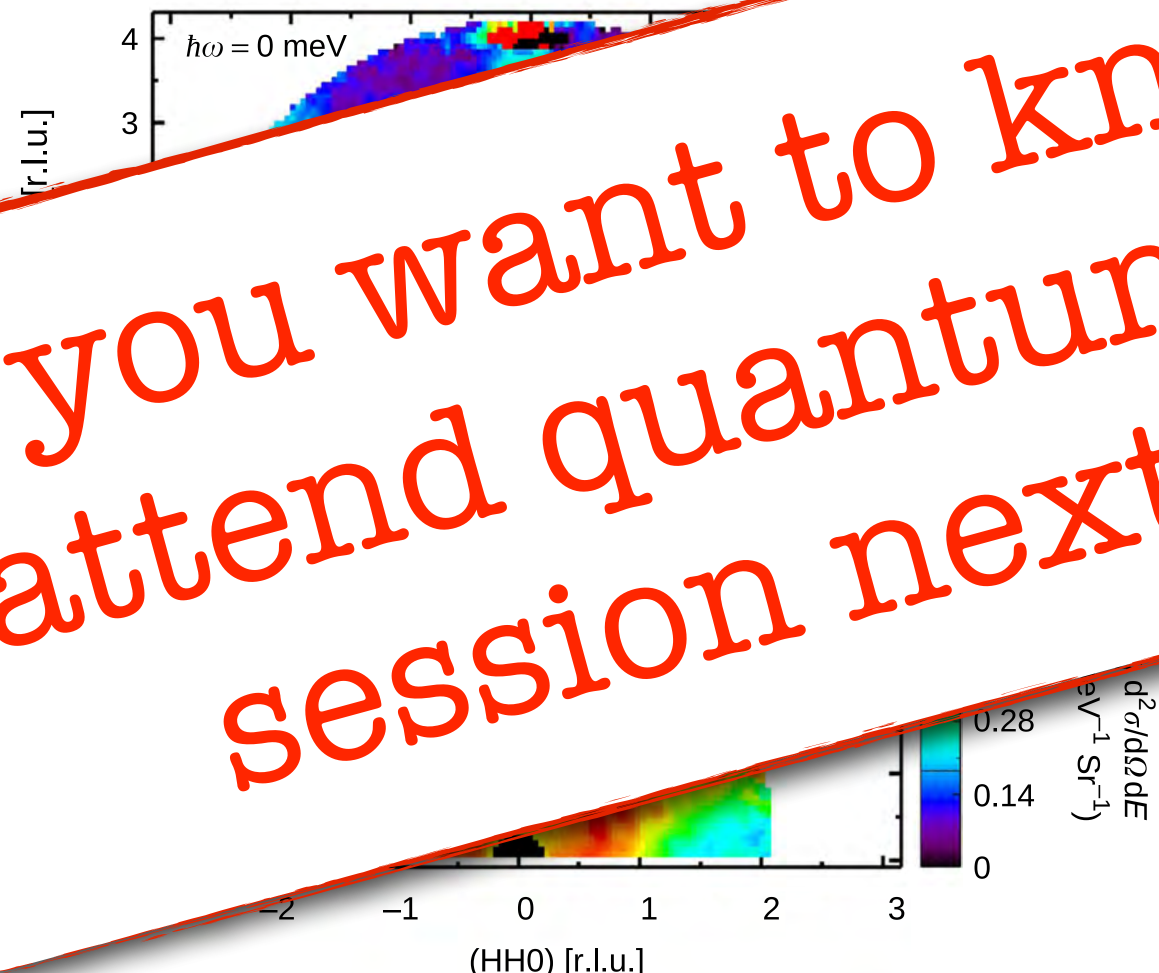
# Quantum fluctuations in spin-ice-like $Pr_2Zr_2O_7$

K. Kimura[1], S. Nakatsuji[1,2], J.-J. Wen[3], C. Broholm[3,4,5], M.B. Stone[5], E. Nishib...




if you want to know more, attend quantum spin ice session next week !

but most scattering is inelastic - evidence of quantum monopole-dynamics ?

K. Kimura *et al*., Nat. Communs. **4**, 1934 (2013)

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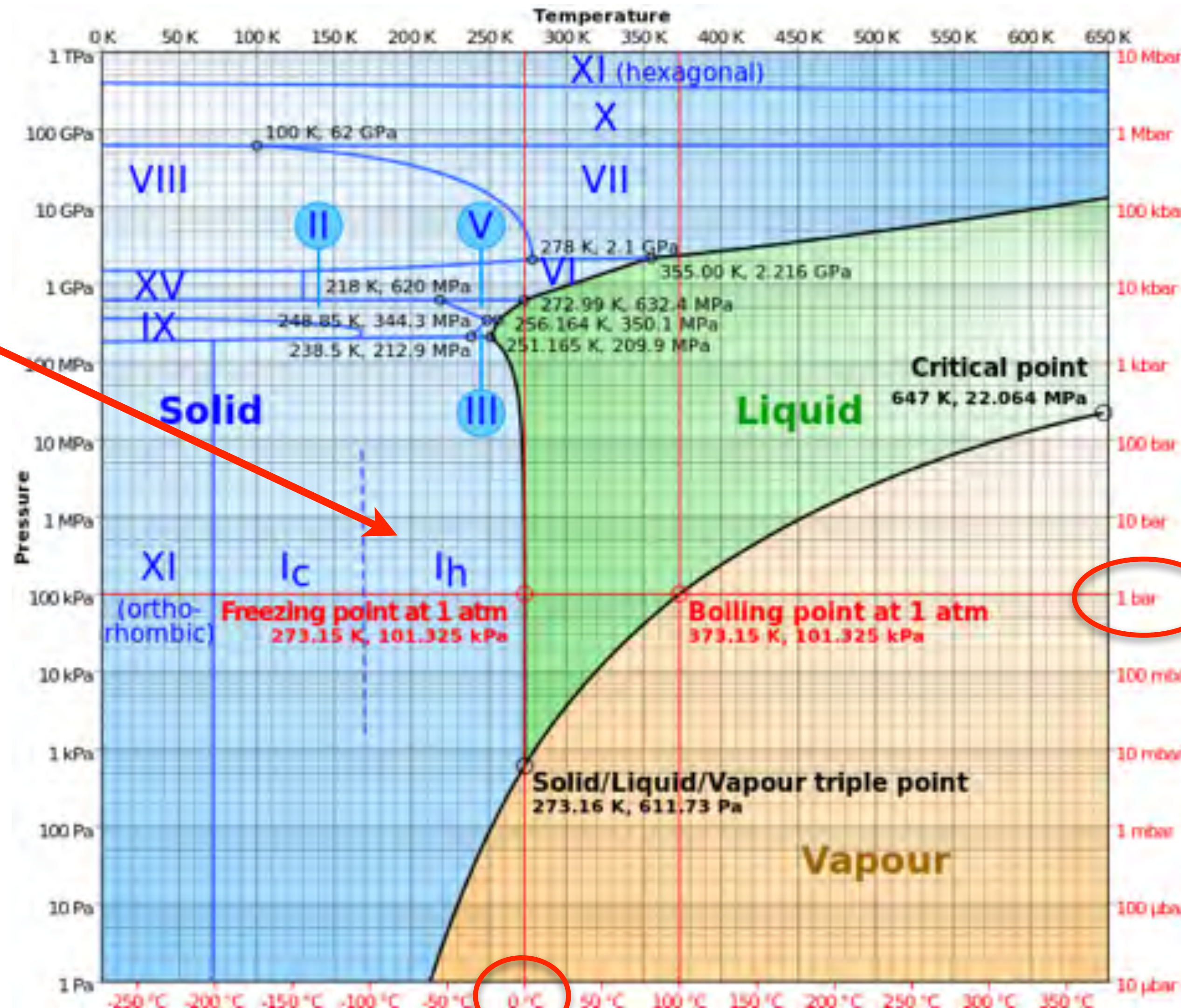
could something like this
happen in water ice ?

# how many forms of ice ?!!!

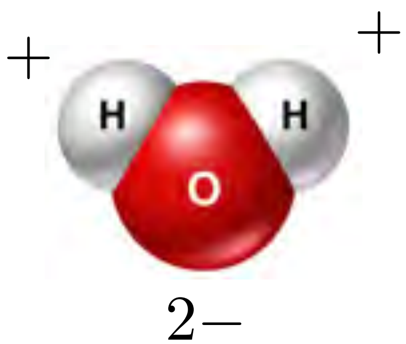



atomespheric pressure

zero celcius

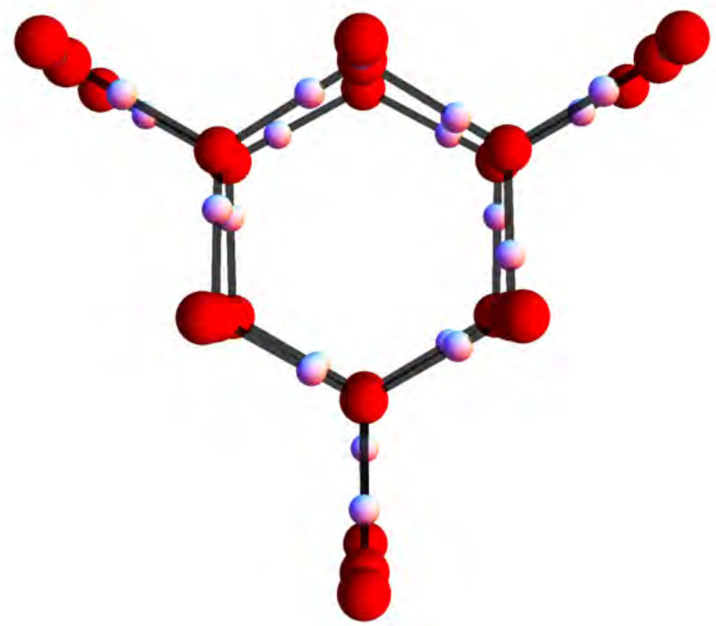
…the 17 phases of ice known to Wikipedia

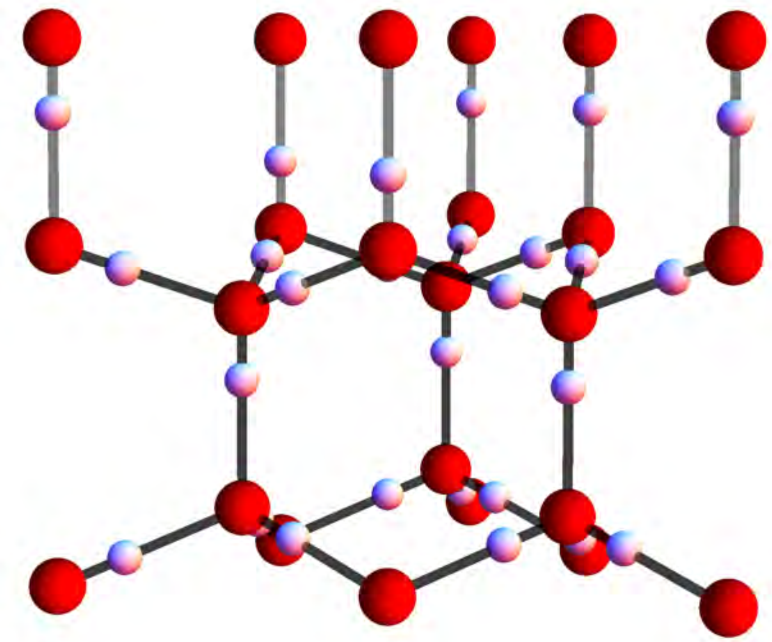
# what about common (Ih) water ice ?




cf.

O$^{2-}$ form a hexagonal crystal lattice





H$^{+}$ do not order

$$\left(\frac{3}{2}\right)^{N/2}$$

different proton configurations satisfying the ice-rules

J. D. Bernal and R. H. Fowler, J. Chem. Phys. **1**, 515 (1933)
L. Pauling, J. Am. Chem. Soc. **27**, 2680 (1935)

# why should you believe this ?

heat capacity of water ice

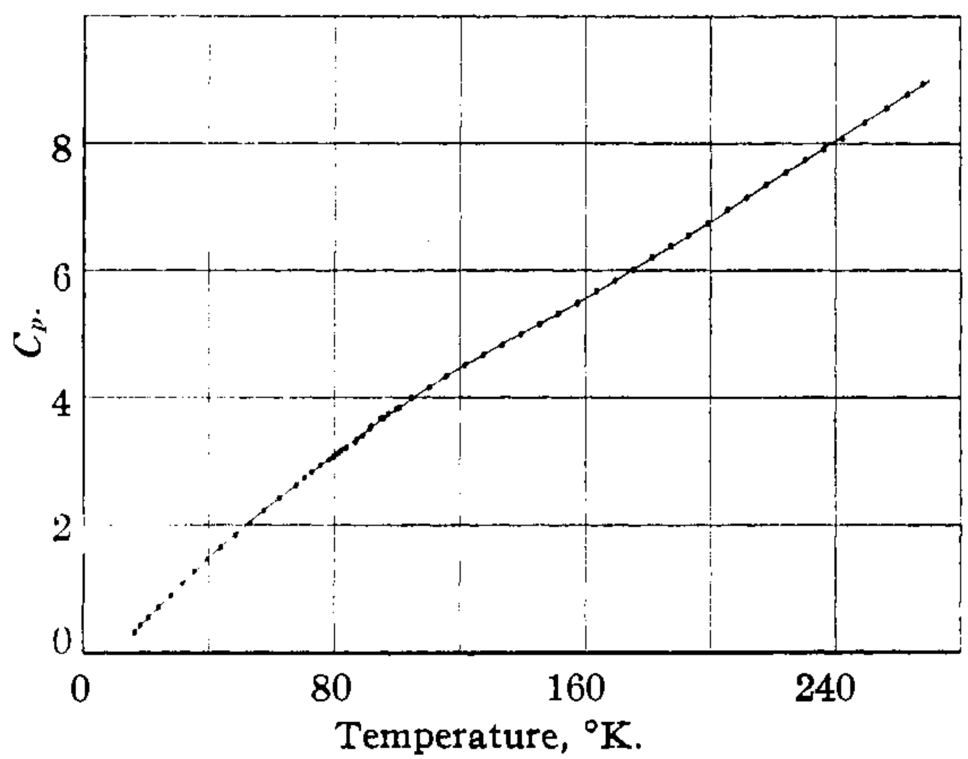
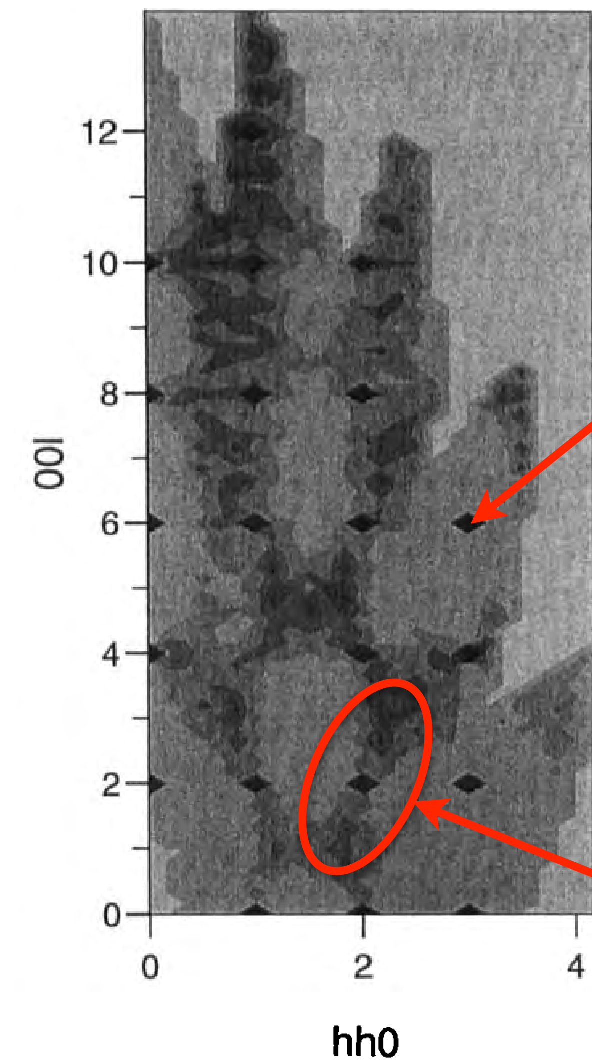



Fig. 2.—Heat capacity in calories per degree per mole of ice.

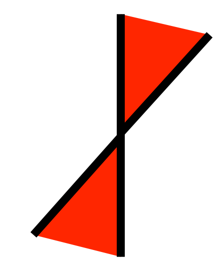
recover Pauling residual “ice entropy” for T➛0

W.F. Giauque and J.W. Stout, Am. J. Chem. Phys.**58**, 114 (1936)

neutron scattering from $D_20$ @ 20 K




nuclear Bragg peak ($O^{2-}$)



“pinch -point”

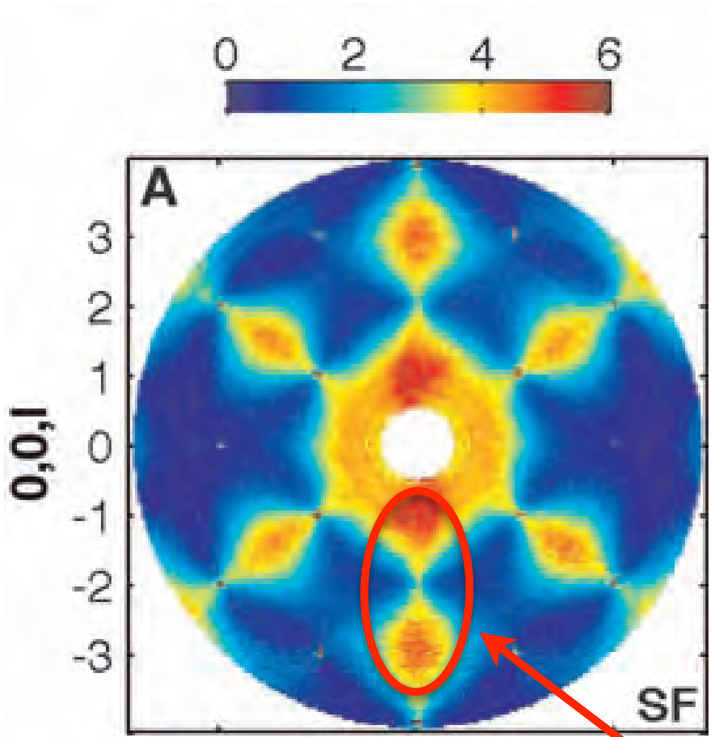
J.C. Li et al., Phil. Mag. B 69, 1173 (1994)

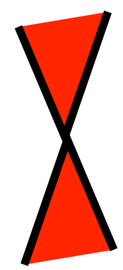
direct (thermodynamic) and indirect (scattering) evidence for an extensive manifold of states

N.B. recent theoretical study : S. Isakov *et al.*, Phys. Rev. B **91**, 245152 (2015) ❦

# is that all ?

$Ho_2Ti_2O_7$ @ 1.7 K

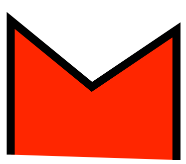


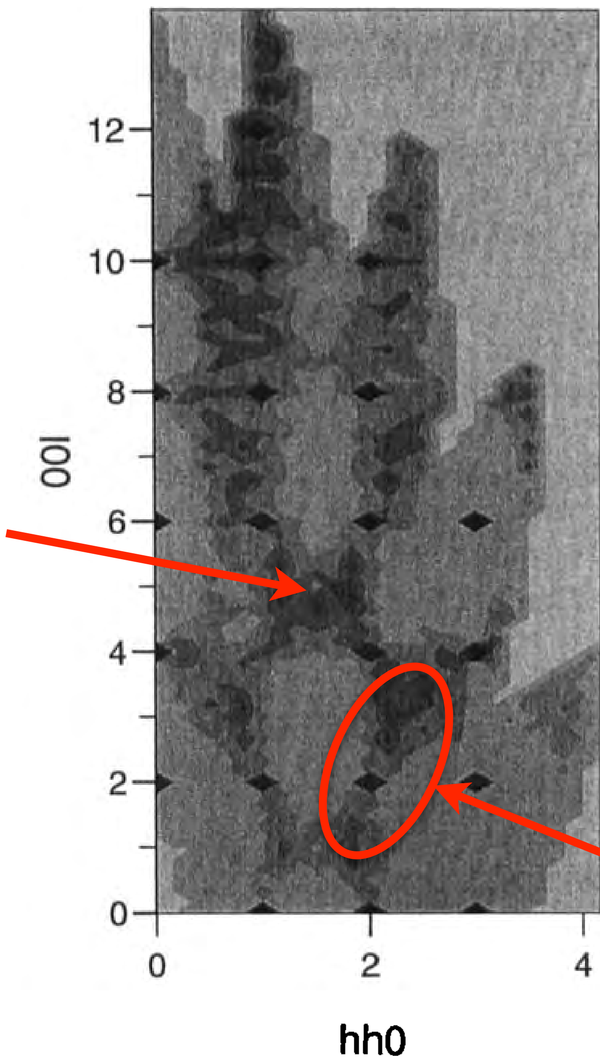



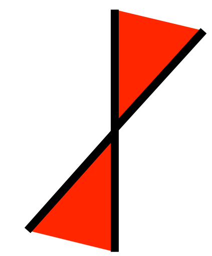
“pinch -point”

D$_2$0 @ 20 K

broad assymetric feature





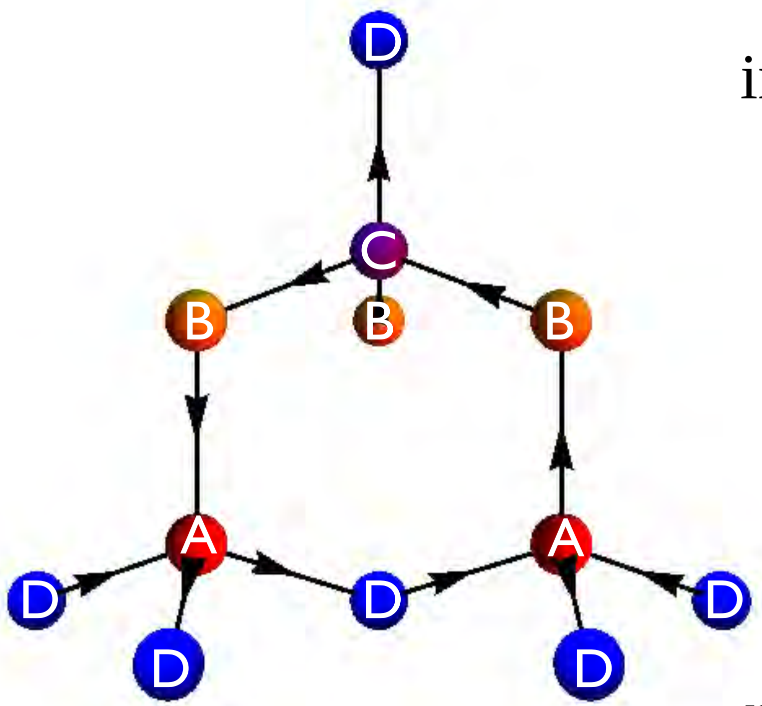
“pinch -point”

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

J.C. Li *et al*., Phil. Mag. B **69**, 1173 (1994)

…the ice rules in ice Ih do not just lead to pinch points

# where do extra features come from ?



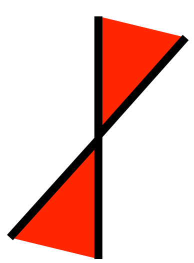

near reciprocal lattice vector **Q**, expand correlations of protons in terms of uniform polarisation **P**$^+$ and staggered polarisation **P**$^-$ :

$$S^{\text{diffuse}}_{\text{proton}}(\mathbf{Q}+\tilde{\mathbf{q}}) \approx \sum_{\upsilon=\pm} F^{\text{proton}}_{\upsilon}(\mathbf{Q})\langle|\hat{\boldsymbol{\lambda}}^{\text{proton}}_{\mathbf{Q},\upsilon}\cdot\mathbf{P}_\upsilon(\tilde{\mathbf{q}})|^2\rangle$$

form factor for protons

unit-vector specific to **Q**

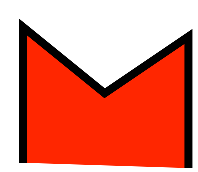
require fields **P**$_+$ and **P**$_-$ to be continuous $\Rightarrow$ **two** constraints



$$\nabla\cdot\mathbf{P}_+(\mathbf{r}) = 0$$

"pinch-point"

$$P^z_-(\mathbf{r}) + \frac{2}{3}a_0\nabla\cdot\mathbf{P}_-(\mathbf{r}) - a_0\partial_z P^z_-(\mathbf{r}) = 0$$



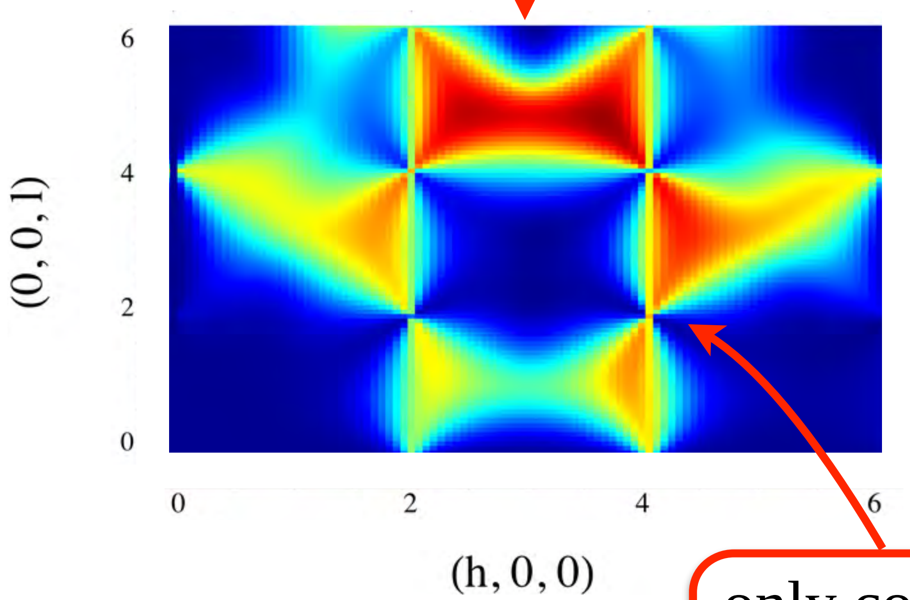
broad assymetric feature

O. Benton, O. Sikora and NS, arXiv:1504.04158v1

# what is ice trying to tell us ?

analytic theory based
on projection onto ice states

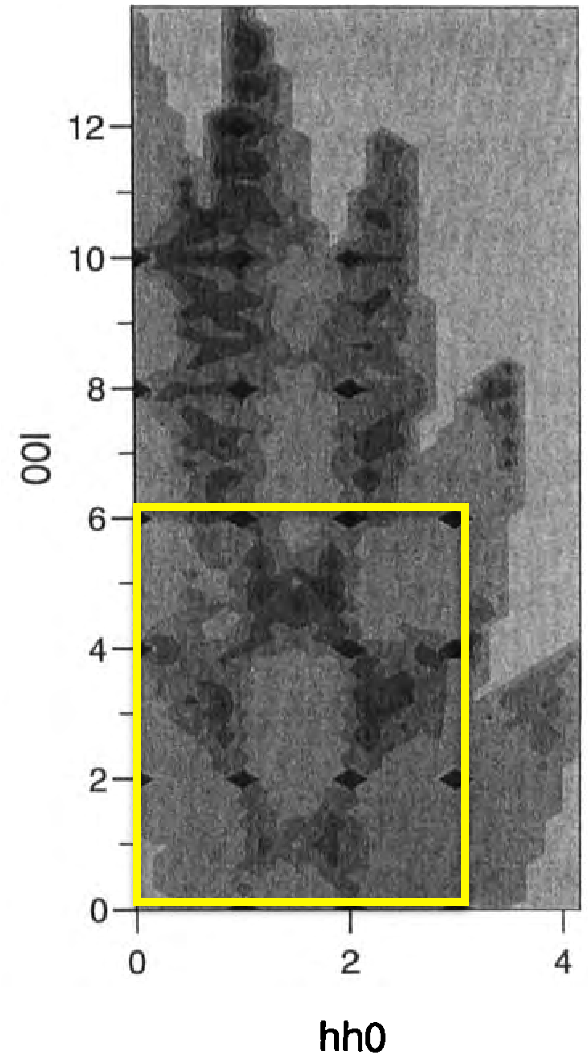
correlations of **P**$^+$ and **P**$^-$ contribute




only correlations of **P**$^+$ contribute

O. Benton, O. Sikora and NS, arXiv:1504.04158v1

$D_20$ @ 20 K




J.C. Li *et al.*, Phil. Mag. B **69**, 1173 (1994)

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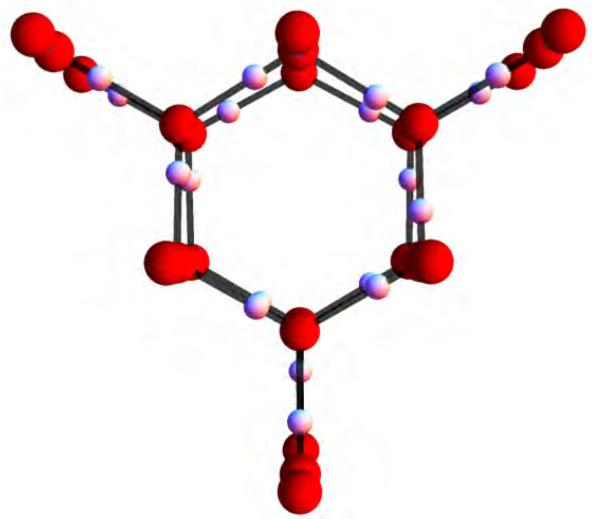
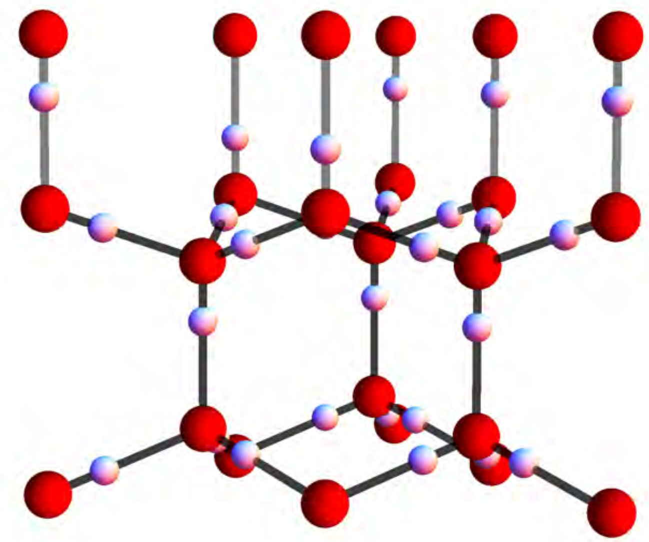
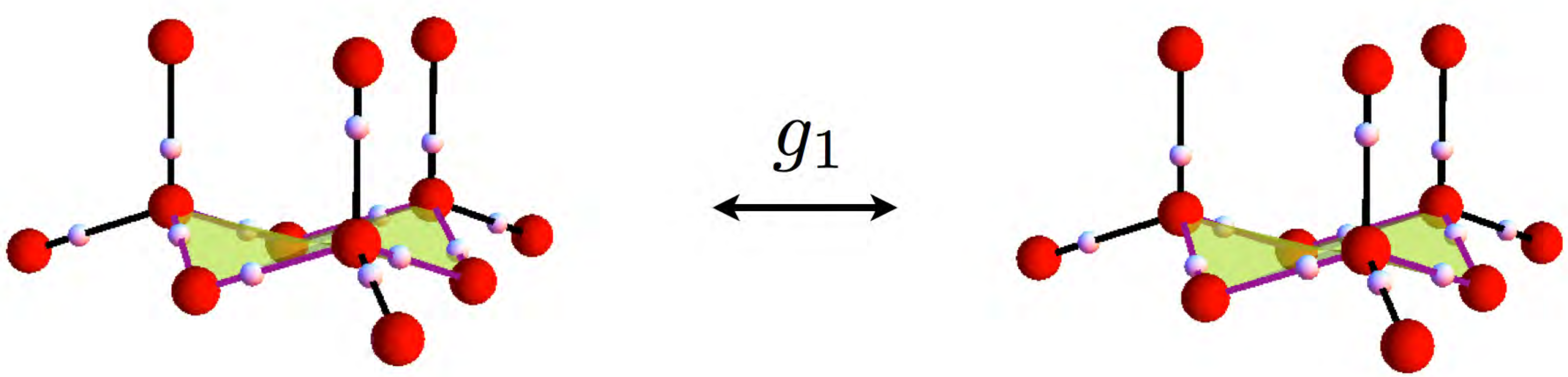
# can the protons move ?

in common (Ih) water ice, oxygen ions form lattice with hexagonal plaquettes





quantum tunnelling can occur on such a plaquette without violating the ice rules

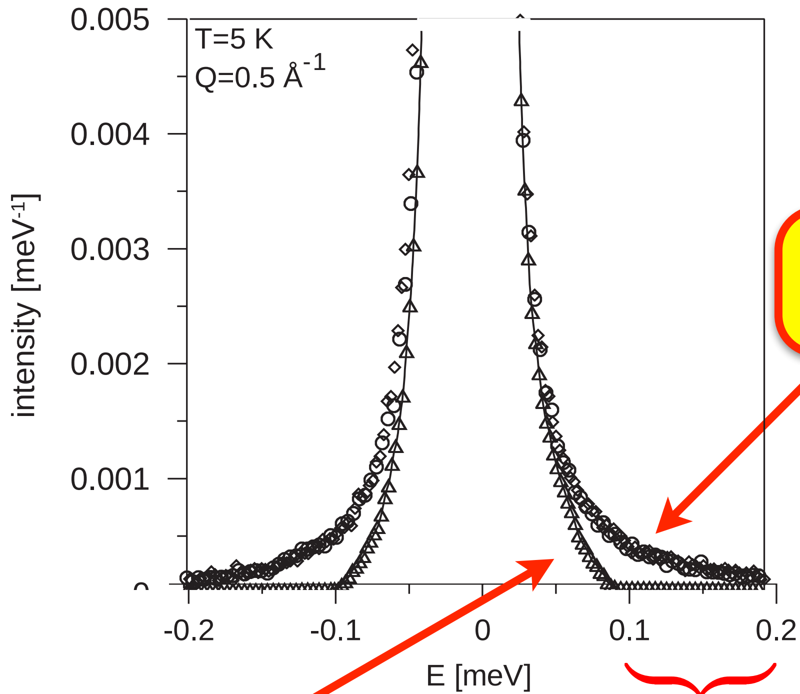



could this provide a route to quantum dynamics in water ice ?

# does this happen in water ice ?

incoherent inelastic neutron scattering experiments carried out at IRIS, RAL :

**ice Ih** (proton-disordered)
and **ice VIII** (proton-ordered) :

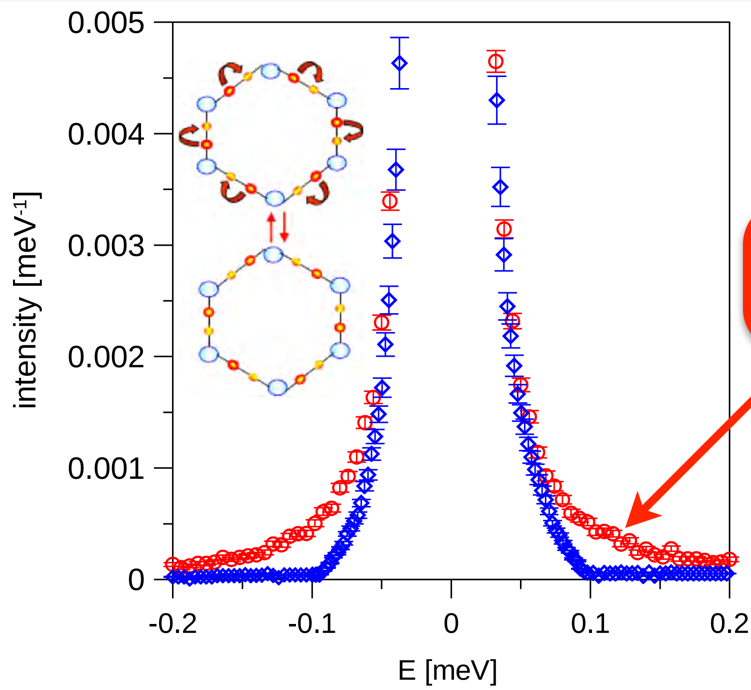



ice Ih

elastic line

inelastic “wing”

**ice Ih** (proton-disordered)
and **partially-deuterated ice Ih** (proton-ordered) :




ice Ih

inelastic signal does **not** have an activated temperature dependence, and form-factors suggest that it **is** associated with protons (not phonons)…

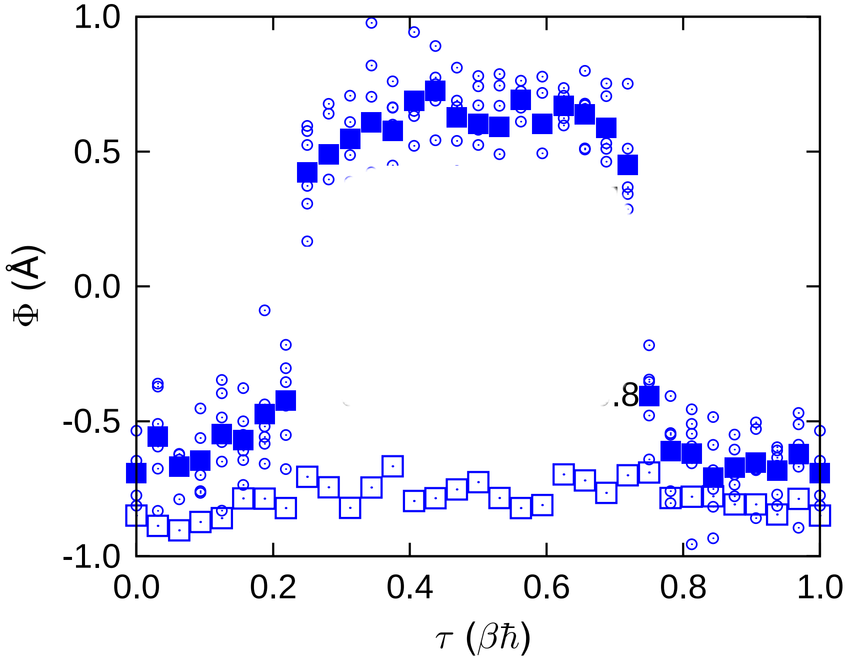
$\Rightarrow$

interpret as collective tunnelling of protons

L.E. Bove *et al.*, Phys. Rev. Lett **103**, 165901 (2009)

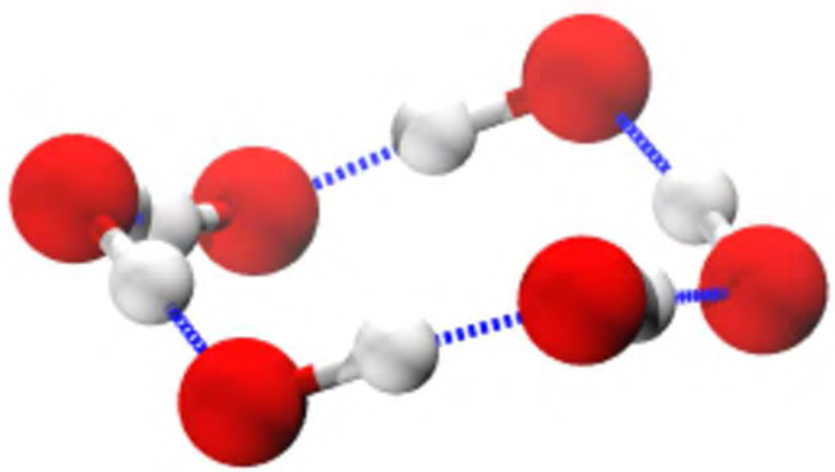
# does this interpretation make sense ?

path-integral quantum Monte Carlo used to explore the (imaginary) time-evolution of proton configurations within a cluster of 48 water molecules




simulation variables for group of protons

simultaneous, correlated tunnelling of 6 protons on an hexagonal plaquette



(see movie)

C. Drechsel-Grau and D. Marx, Phys. Rev. Lett **112**, 148302 (2014)

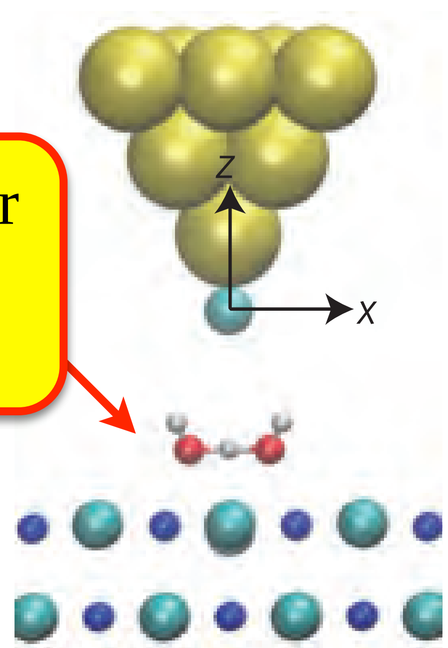
# collective tunnelling observed !

use STM to explore changes in proton configuration in a ring of water molecules on a surface

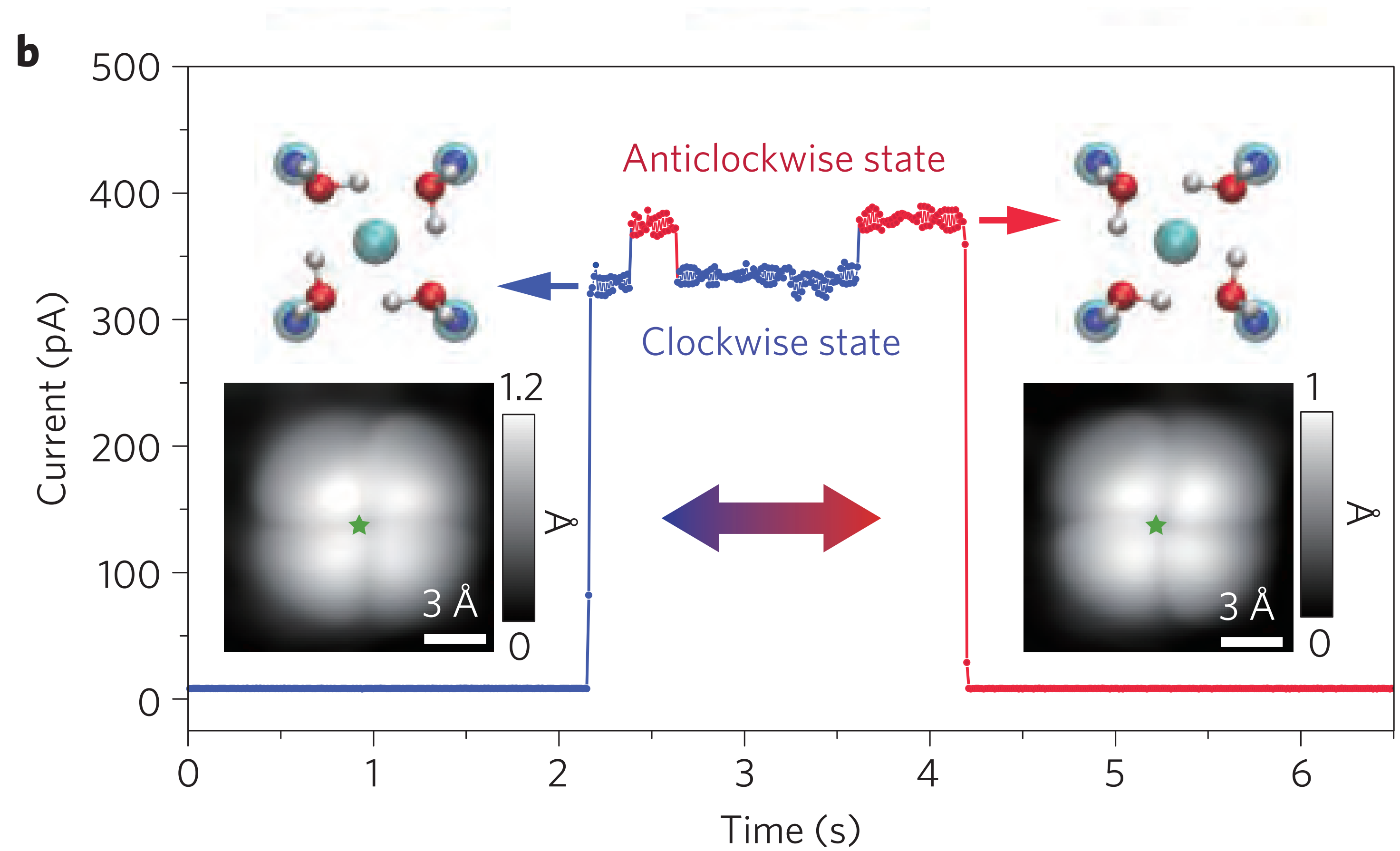
observe collective tunnelling of four protons between the four water molecules

STM tip

ring of four water molecules




NaCl




X. Meng *et al.*, Nature Physics **11**, 235 (2015) what is known (or believed) about the effect of quantum tunnelling in water ice ?

PHYSICAL REVIEW B 74, 024302 (2006)

# Ice: A strongly correlated proton system

A. H. Castro Neto,[1] P. Pujol,[2] and Eduardo Fradkin[3]

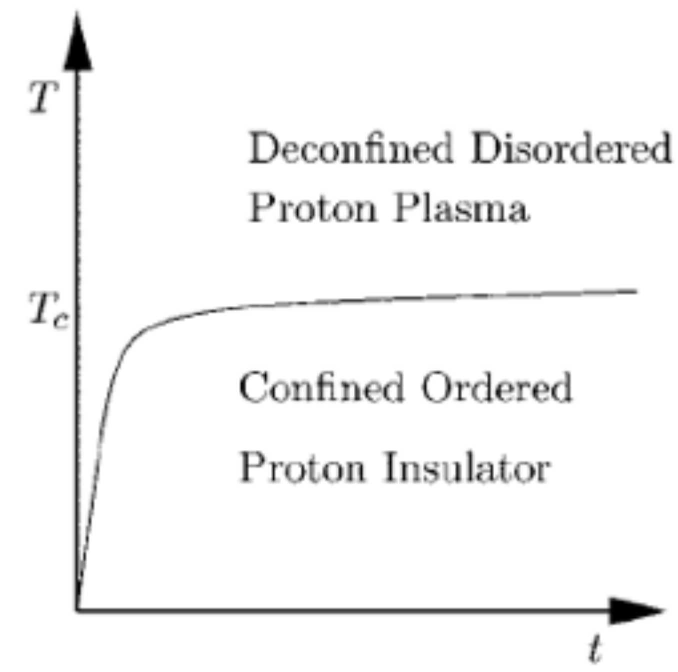
**2D planar ice**

quantum ice on checkerboard lattice
⇒ (compact) lattice gauge theory
⇒ continuum field theory

$$S_{\text{eff}} = \int d^2\vec{r}\, d\tau \left\{ \frac{K}{2}\left[(\nabla\chi_1)^2 + (\partial_\tau\chi_1)^2\right] - \gamma \cos(4\pi\chi_1) \right\},$$

RG ⇒ ordered ground state (cf. Polyakov)

temperature




proton hopping

ordered ground state in 2D ice model known from numerical simulations :

N. Shannon, G. Misguich and K. Penc, Phys. Rev. B **69**, 220403(R) (2004).
O. F. Syljusen and S. Chakravarty, Phys. Rev. Lett. **96**, 147004 (2006).
L. P. Henry and T. Roscilde, Phys. Rev. Lett. **113**, 027204, (2014).

PHYSICAL REVIEW B 74, 024302 (2006)

# Ice: A strongly correlated proton system

A. H. Castro Neto,[1] P. Pujol,[2] and Eduardo Fradkin[3]

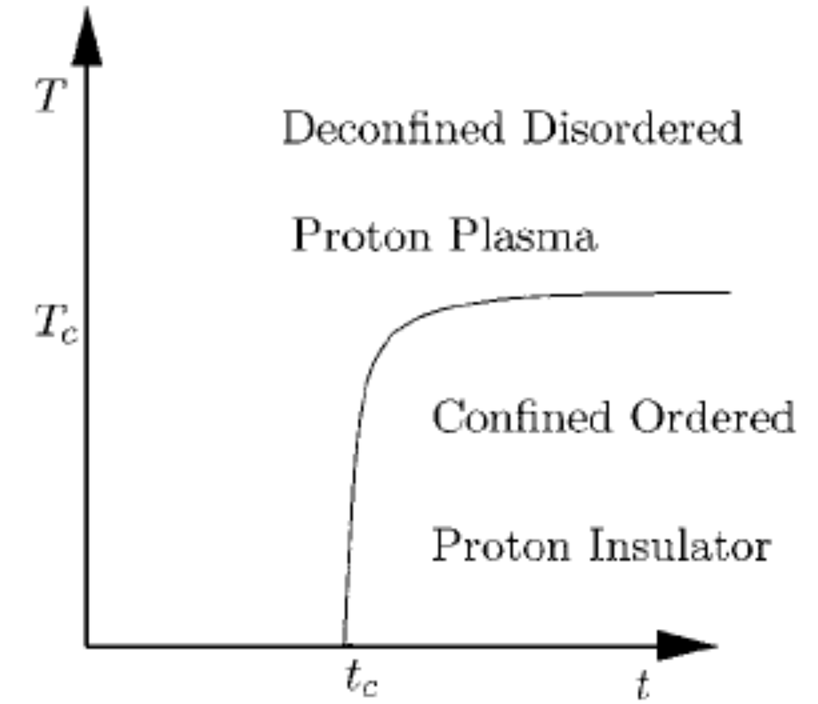
**3D, notionally cubic, ice**

anticipate T=0 confinement-transition

argue that quantum fluctuations could be controlled by doping K in common (Ih) water ice

connection with ice XI ?
(proton-ordered, ferroelectric…)

temperature




proton hopping

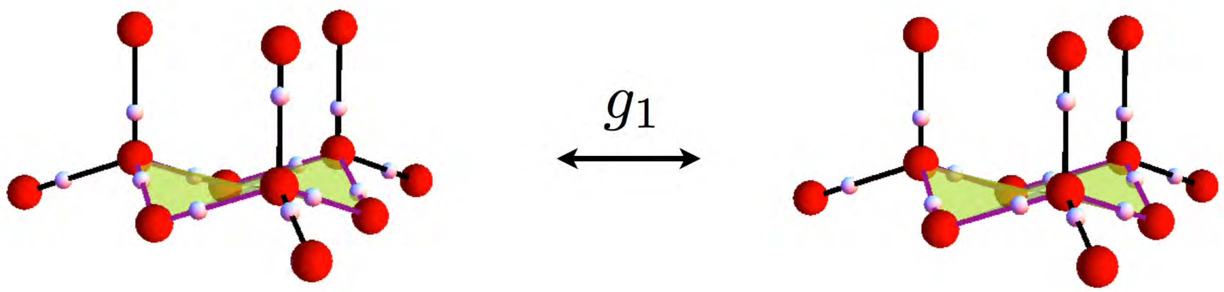
deconfined ground state in 3D quantum spin-ice model confirmed by numerical simulations :

M. Hermele *et al.*, Phys. Rev. B **69**, 064404 (2004); A. Banerjee *et al*. Phys. Rev. Lett. **100**, 047208 (2008)
N. Shannon *et al*. Phys. Rev. Lett. **108**, 067204 (2012); O. Benton *et al.*, Phys. Rev. B. **83**, 075174 (2012) ❦
Y. Kato and S. Onoda, arXiv:1411.1918v2

# what model do we need to solve ?

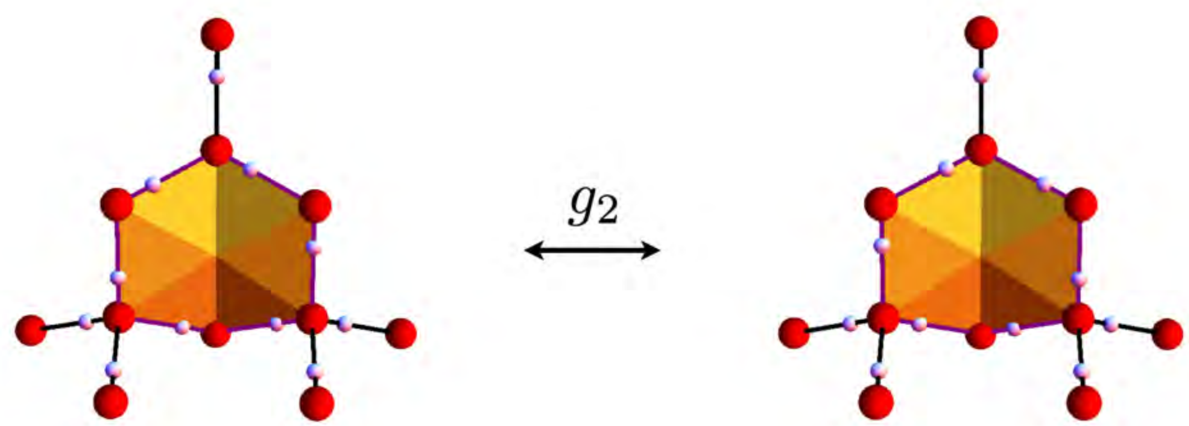
minimal model for quantum tunnelling between different
proton configurations obeying the ice rules

$$\mathcal{H}^{\text{hexagonal}}_{\text{tunnelling}} = -g_1 \sum_{\hexagon \in I} \left[ |\circlearrowright\rangle\langle\circlearrowleft| + |\circlearrowleft\rangle\langle\circlearrowright| \right] - g_2 \sum_{\hexagon \in II} \left[ |\circlearrowright\rangle\langle\circlearrowleft| + |\circlearrowleft\rangle\langle\circlearrowright| \right]$$




…tunnelling on plaquettes
within hexagonal planes

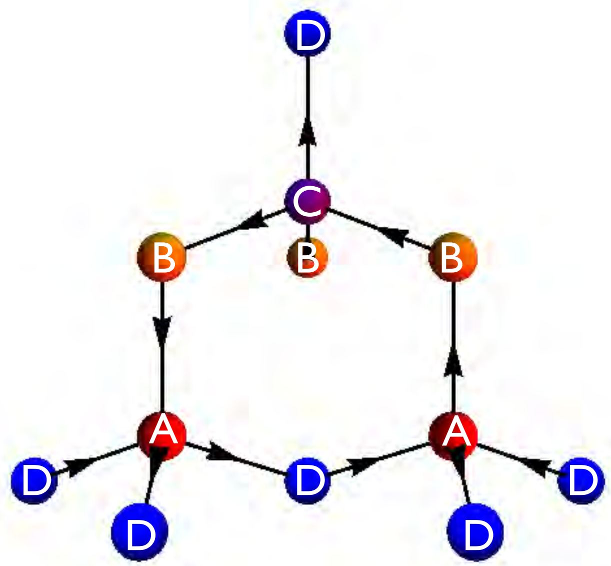
tunnelling on plaquettes
out of hexagonal planes…



# ice Ih as a lattice gauge theory…

minimal microscopic model for quantum tunnelling…

$$\mathcal{H}^{\text{hexagonal}}_{\text{tunnelling}} = -g_1 \sum_{\hexagon \in I} \left[ |\circlearrowright\rangle\langle\circlearrowleft| + |\circlearrowleft\rangle\langle\circlearrowright| \right] - g_2 \sum_{\hexagon \in II} \left[ |\circlearrowright\rangle\langle\circlearrowleft| + |\circlearrowleft\rangle\langle\circlearrowright| \right]$$




$$S^z_{\mathbf{rr'}} \to E_{\mathbf{rr'}} \ , \ S^{\pm}_{\mathbf{rr'}} \to e^{\pm i A_{\mathbf{rr'}}}$$

$$[E_{\mathbf{rr'}}, A_{\mathbf{r''r'''}}] = i\delta_{\mathbf{rr''}}\delta_{\mathbf{r'r'''}}$$

spin-1/2 representation of protons on bonds

$$\mathcal{H}_{\mathsf{U}(1)} = \frac{\mathcal{U}}{2} \sum_{\langle \mathbf{rr'} \rangle \in \mathsf{CS}} E^2_{\mathbf{rr'}} + \frac{\mathcal{U}'}{2} \sum_{\langle \mathbf{rr'} \rangle \in \mathsf{MS}} E^2_{\mathbf{rr'}} + \frac{\mathcal{K}}{2} \sum_{\hexagon \in I} [\nabla_{\hexagon} \times A]^2 + \frac{\mathcal{K}'}{2} \sum_{\hexagon \in II} [\nabla_{\hexagon} \times A]^2$$

…non-compact U(1) lattice gauge theory

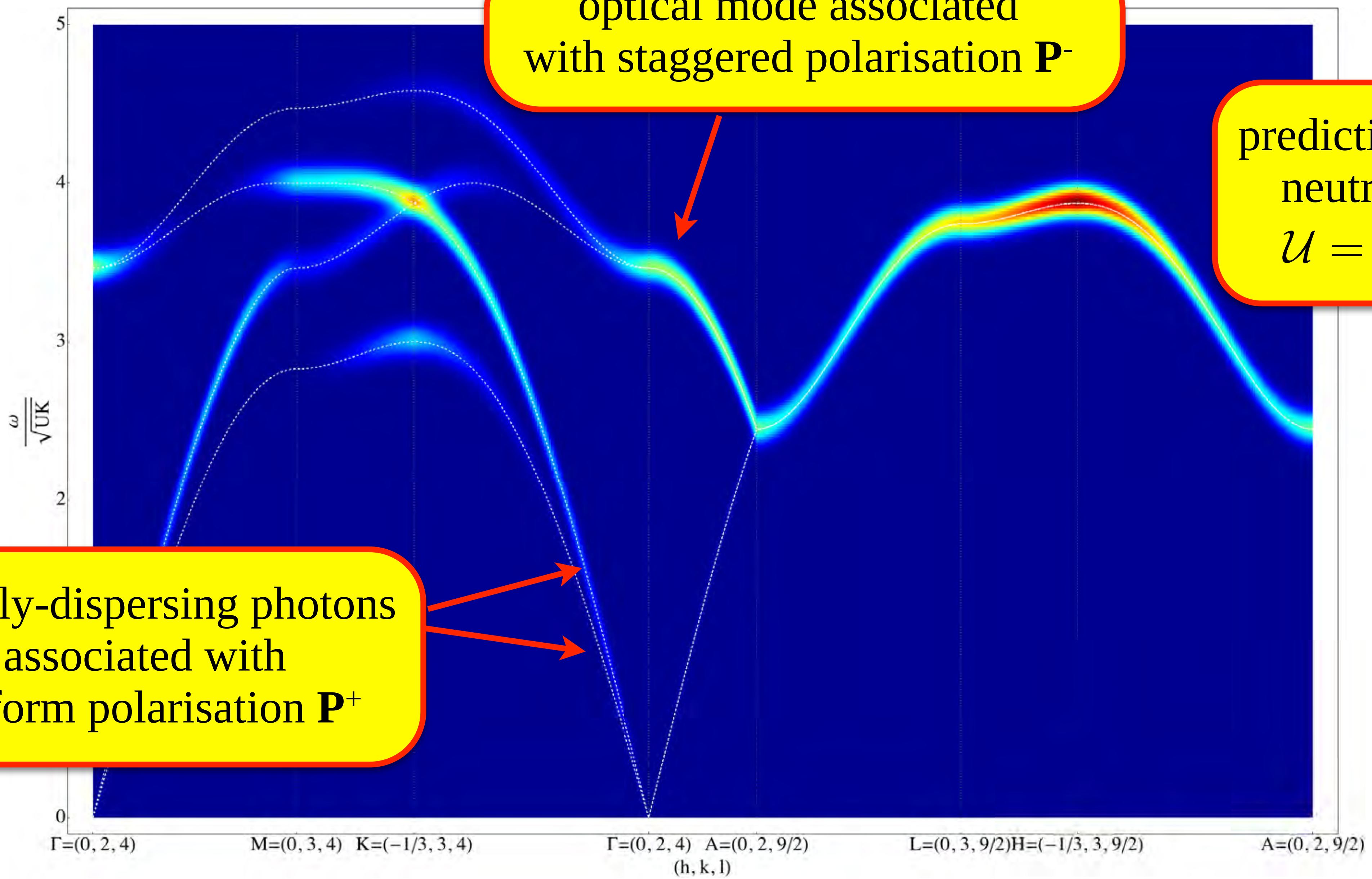
O. Benton, O. Sikora and NS, arXiv:1504.04158v1

# emergent photons…

O. Benton, O. Sikora and NS, arXiv:1504.04158v1

optical mode associated with staggered polarisation $\mathbf{P}^-$

prediction for inelastic neutron scattering
$\mathcal{U} = \mathcal{U}'$ , $\mathcal{K} = \mathcal{K}'$




linearly-dispersing photons associated with uniform polarisation $\mathbf{P}^+$

**emergent photons are birefringent**,
i.e. two distinct branches associated with different polarisations of “light”

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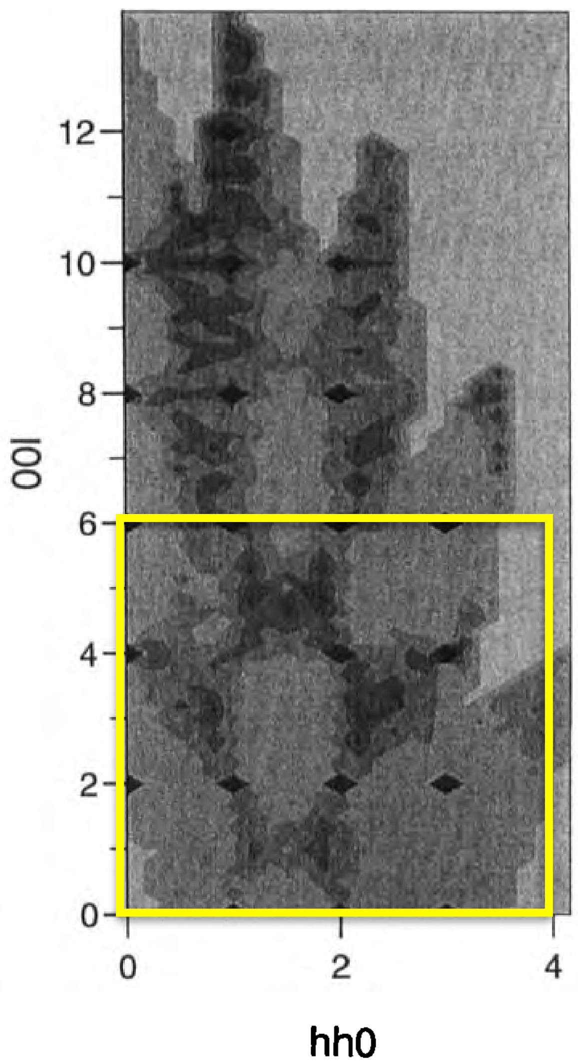
# what about diffuse scattering ?

diffuse neutron scattering from $D_20$ @ 20K

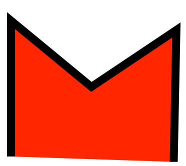
classical theory

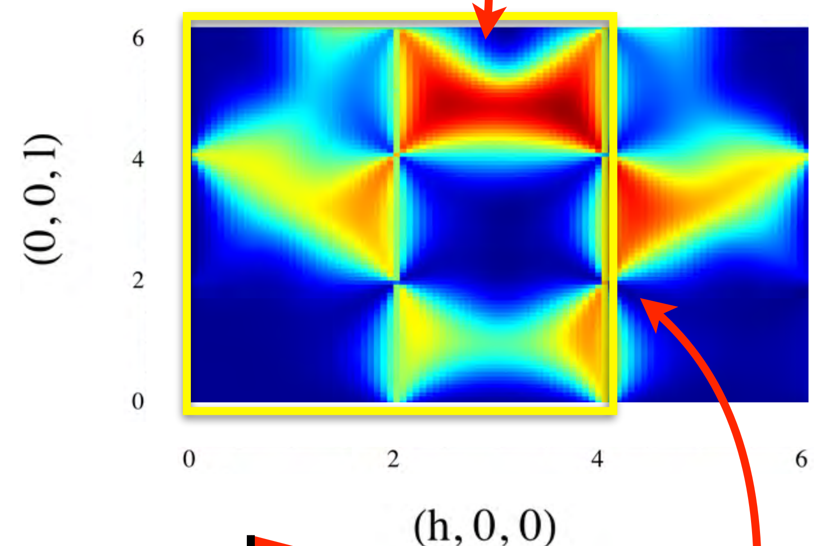
quantum theory

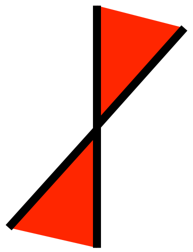



J.C. Li et al.,
Phil. Mag. B 69, 1173 (1994)

broad assymetric feature

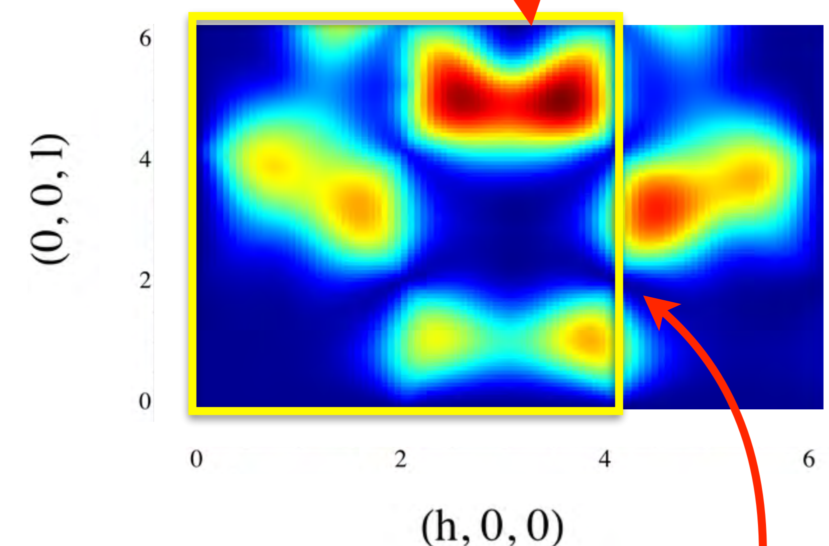




"pinch-point"

broad feature loses sharp edges

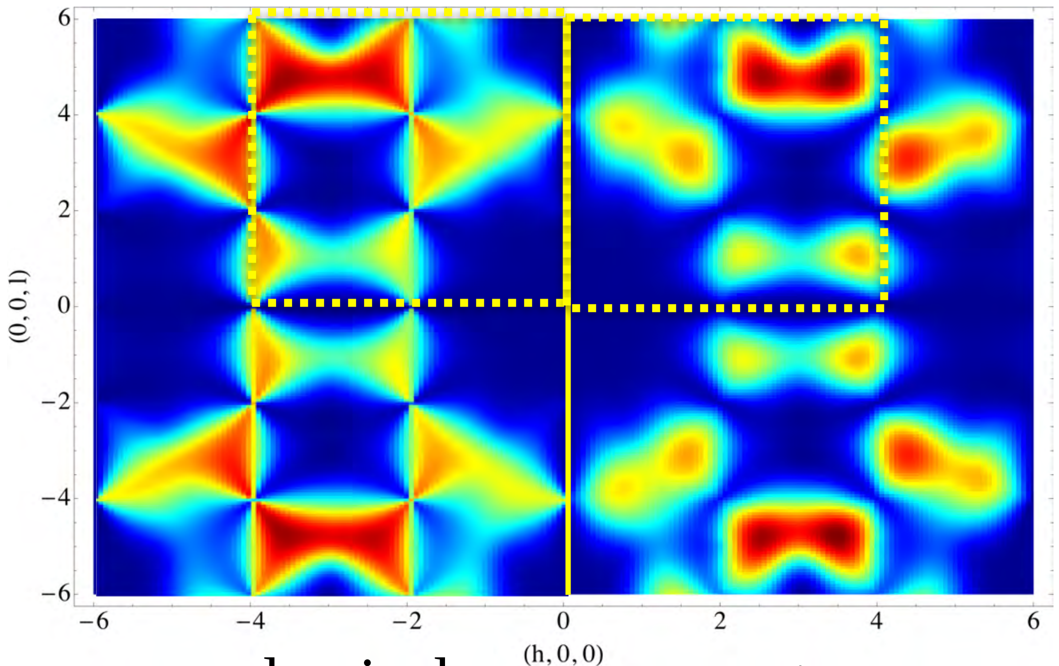



"pinch-point" washes out

O. Benton, O. Sikora and NS, arXiv:1504.04158v1

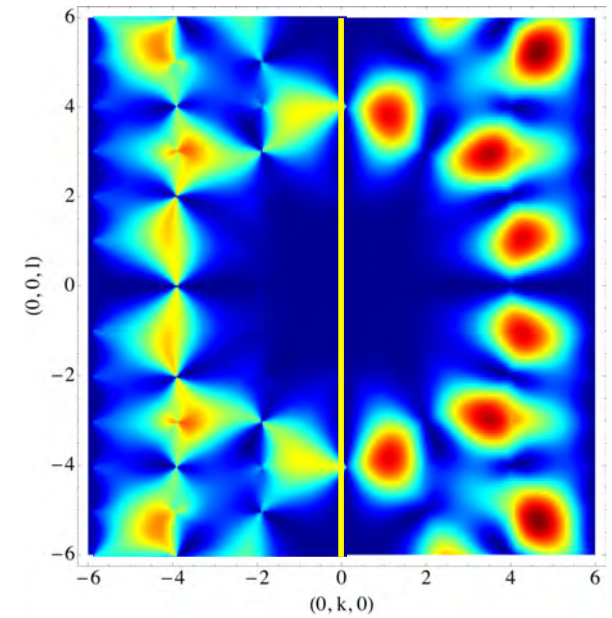
# what about diffuse scattering ?

h0l plane



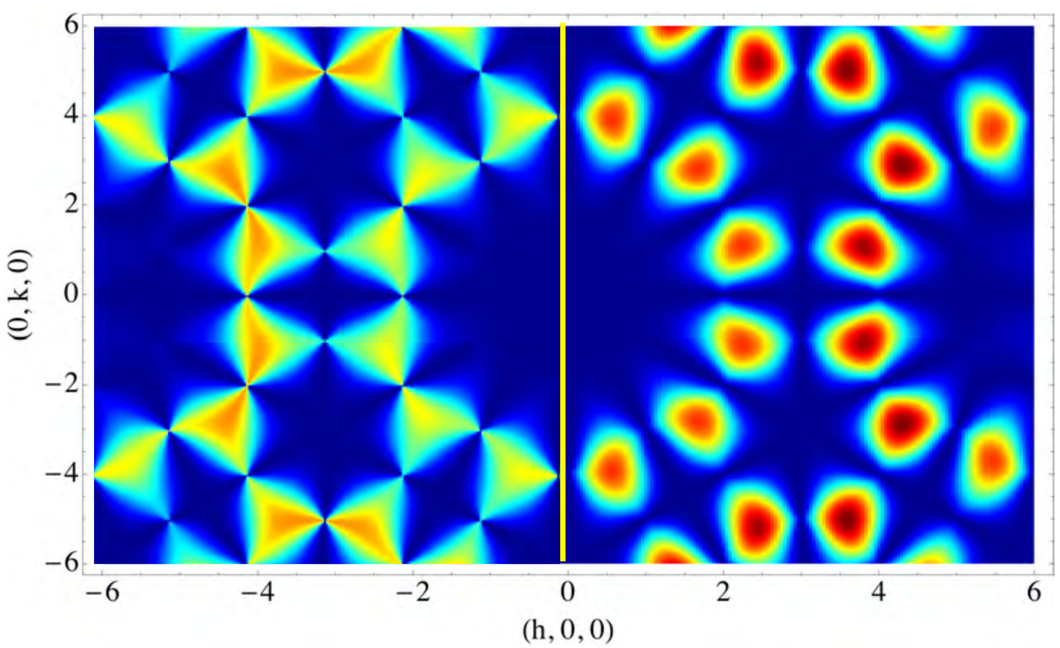

classical quantum

0kl plane




classical quantum

hk0 plane



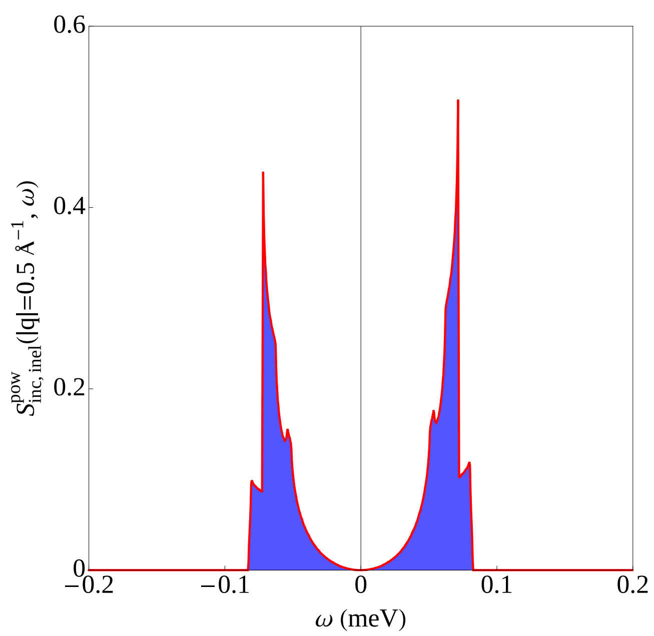

classical quantum

in both cases, results have been validated by comparison with classical/quantum Monte Carlo simulation (not shown)

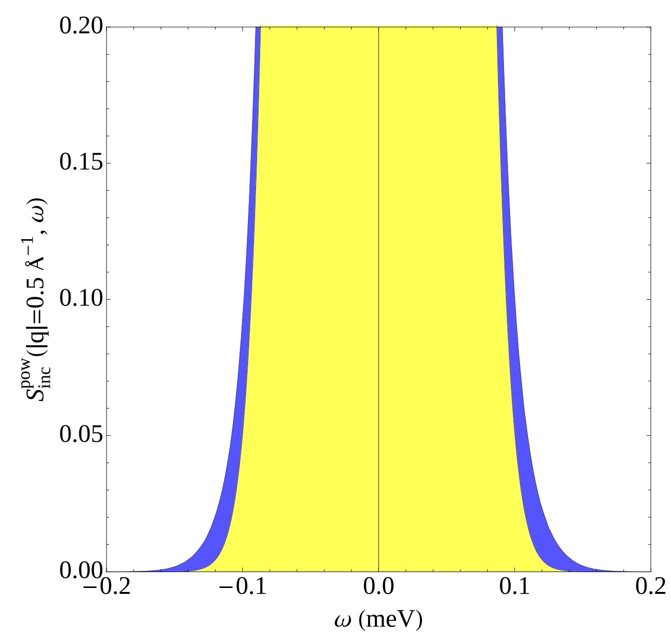
O. Benton, O. Sikora and NS, arXiv:1504.04158v1

# can we explain those “wings” ?

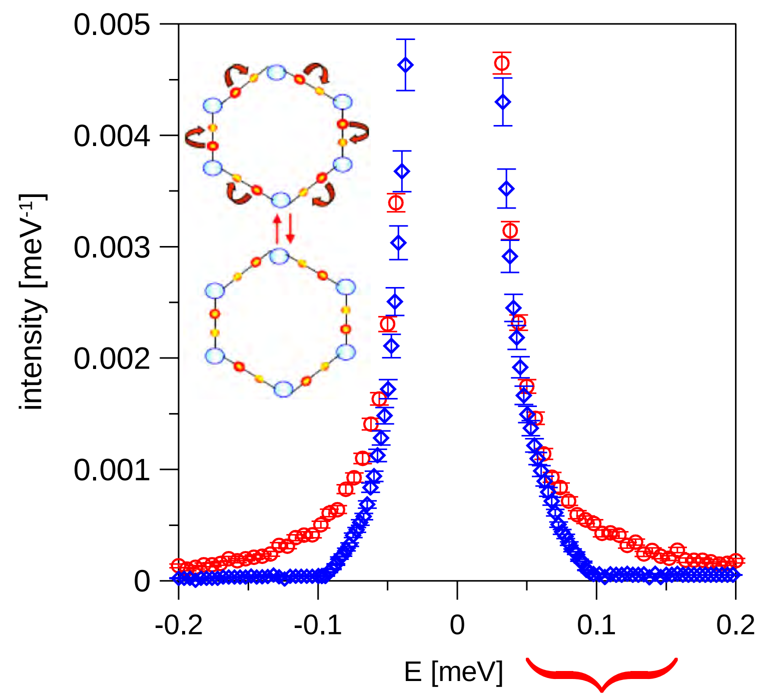
prediction for incoherent
inelastic neutron scattering




convoluted with Gaussian,
on top of elastic line




experimental data




inelastic “wing”

inelastic “wing”

$$\mathcal{U} = \mathcal{U}' \ , \ \mathcal{K} = \mathcal{K}'$$

$$\sqrt{\mathcal{U}\mathcal{K}} = 0.018 \, \mathrm{meV}$$

L.E. Bove *et al.*,
Phys. Rev. Lett **103**, 165901 (2009)

encouraging, but not conclusive - similar features, but a lot of parameters to play with…

O. Benton, O. Sikora and NS, arXiv:1504.04158v1

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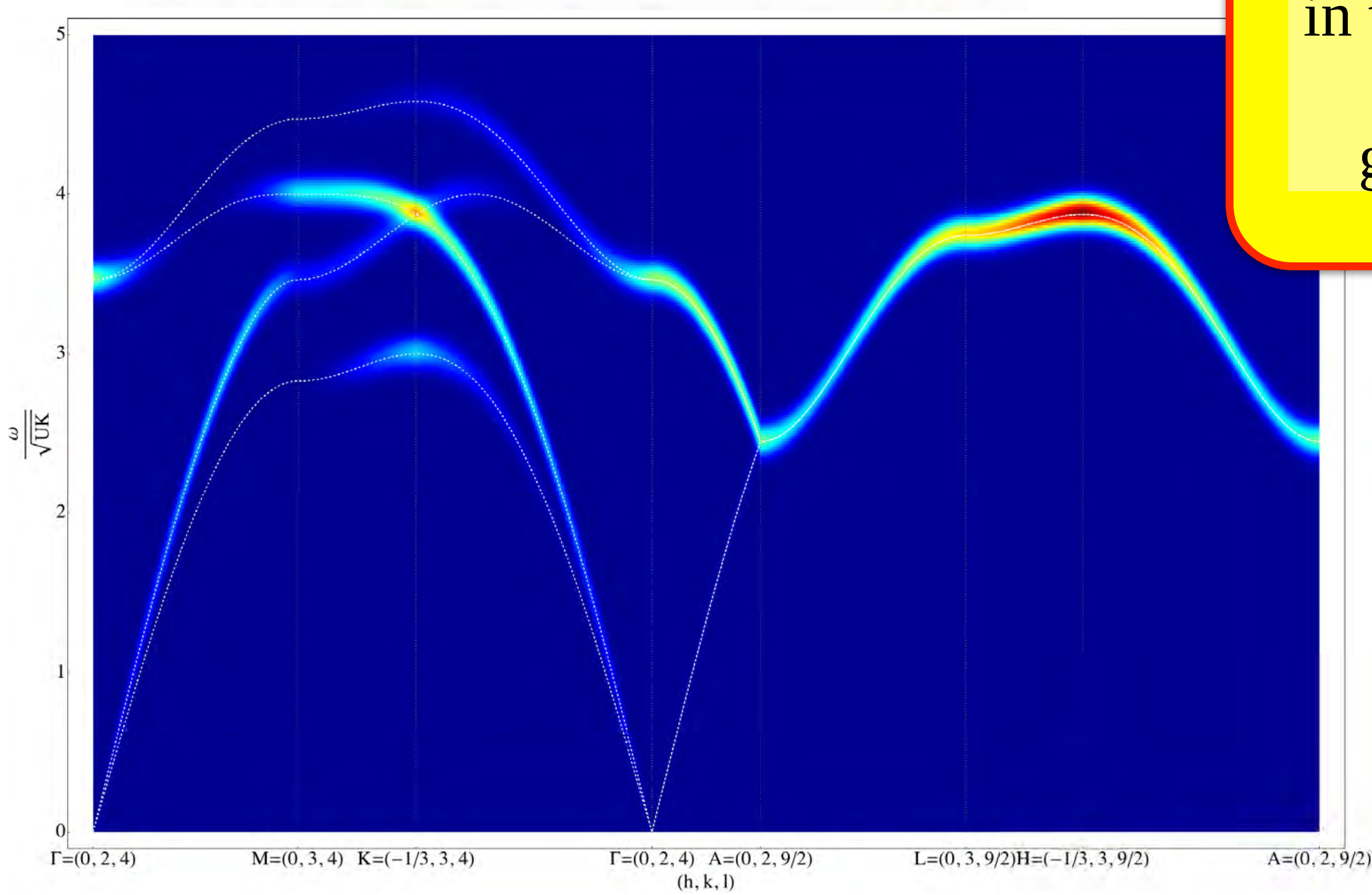
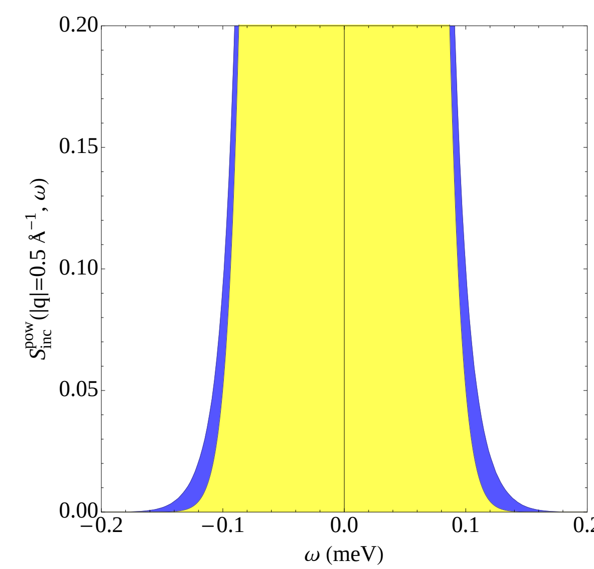
# where does that leave us ?

we have used a combination of analytic field theory and numerical simulation to explore the correlation between protons in classical, and quantum, models of hexagonal (Ih) water ice

in the case of quantum water ice, we obtain a description in terms of a U(1) lattice gauge theory with birefringent photons

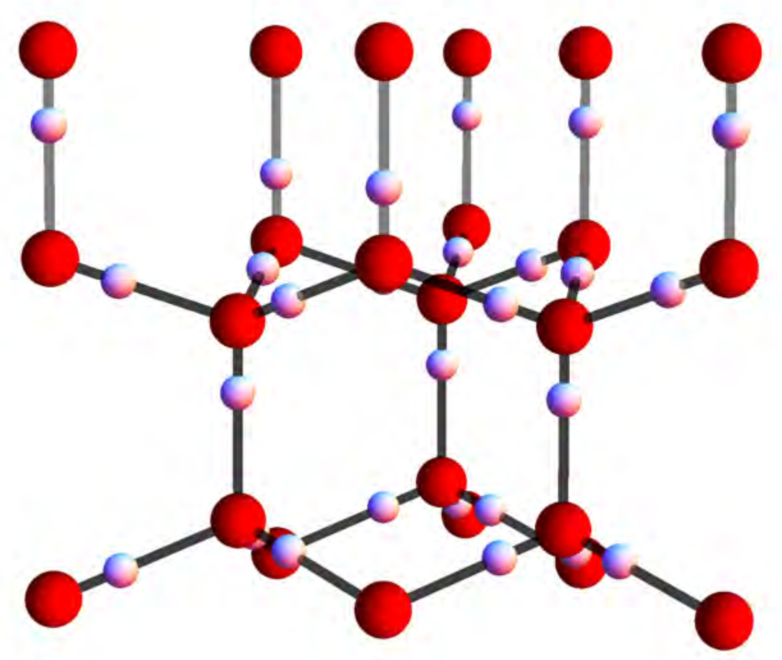



O. Benton, O. Sikora and NS, arXiv:1504.04158v1




this may explain the “wings” seen in incoherent inelastic neutron scattering on Ih water ice

# where next ?



water ice is a strange and beautiful substance and,
despite being studied for thousands of years,
is still at the frontiers of research in
chemistry, physics, life and earth sciences

proton correlations in water ice have been discussed
for almost 100 years, but there are still many issues
which are not understood

in many cases, modern theoretical and experimental techniques have yet to be applied

encouragingly, water ice is just one example of a proton-bonded system - there are lots of others,
many of which have interesting classical degeneracies, and potentially quantum dynamics


thanks for listening !