Title: Approaching the ground state of classical dipolar spin ice $Dy_2Ti_2O_7$: a renewed study with neutron scattering

Date: Jun 08, 2017 11:20 AM

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Abstract: The true magnetic ground state in thermally equilibrated classical spin ice compounds such as $Dy_2Ti_2O_7$ remains an important but yet to be settled issue. In this talk, I will present our recent neutron scattering study of static and dynamic magnetic correlations in isotope-enriched $^{162}Dy_2Ti_2O_7$ single-crystal samples. Implications within the context of possible quantum effects in dipolar spin ice based on our neutron results will be discussed as well.

Approaching the ground state of classical dipolar spin ice $Dy_2Ti_2O_7$

Yixi Su

Juelich Centre for Neutron Science JCNS at MLZ
Forschungszentrum Jueich
Garching, Germany

MLZ is a cooperation between:

7-9th June 2017, Waterloo, Canada

Co-workers and collaborators

Viviane Pecanha-Antonio, Erxi Feng, Xiao Wang
(JCNS-MLZ, Garching)

Michel Gingras, Jeff Rau (Univ. of Warterloo)

G. Balakrishnan, M.C. Hatnean, M.R. Lees (Warwick Univ.)

Th. Brueckel (JCNS)

A. Sazonov, V. Hutanu (RWTH Aachen and MLZ)

J. Ollivier (ILL)

F. Demmel (ISIS)

V. Pomjakushin (PSI)

and many more …

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and many more

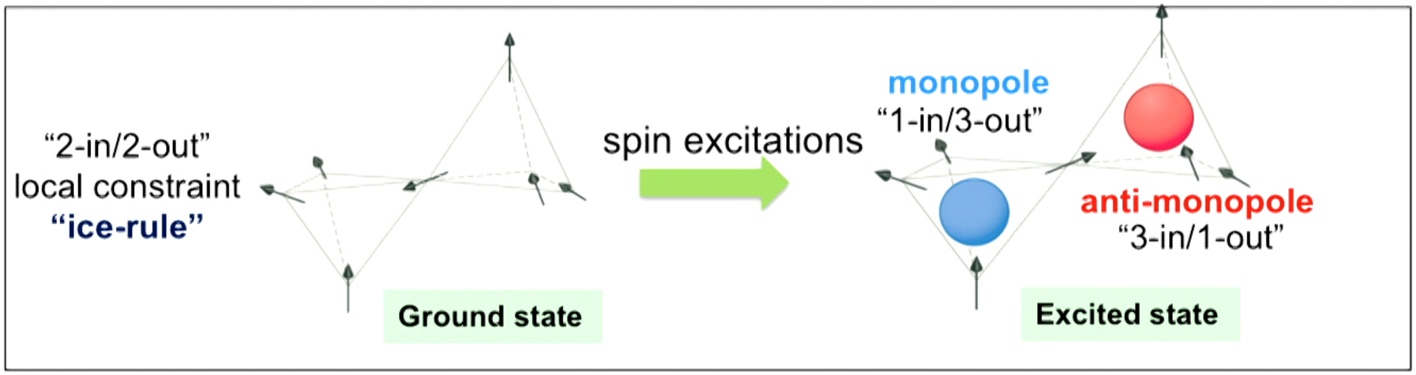
1. Introduction:
spin ice – from classical to quantum

Classical dipolar spin ice

Huge success with the dipolar spin ice physics

$$H_{DSI} = -J\sum_{\langle ij\rangle}\mathbf{S}_i\cdot\mathbf{S}_j + Da^3\sum_{j>i}\left[\frac{\mathbf{S}_i\cdot\mathbf{S}_j}{|\mathbf{r}_{ij}|^3} - \frac{3(\mathbf{S}_i\cdot\mathbf{r}_{ij})(\mathbf{S}_j\cdot\mathbf{r}_{ij})}{|\mathbf{r}_{ij}|^5}\right]$$

- Realization of emergent magnetic monopole and gauge structure
- Highly entropic ground state
- Spin freezing and non-equilibrium physics
- Monopole dynamics and possible Wien effect



"2-in/2-out"
local constraint
"ice-rule"

spin excitations

Ground state

monopole
"1-in/3-out"

anti-monopole
"3-in/1-out"

Excited state

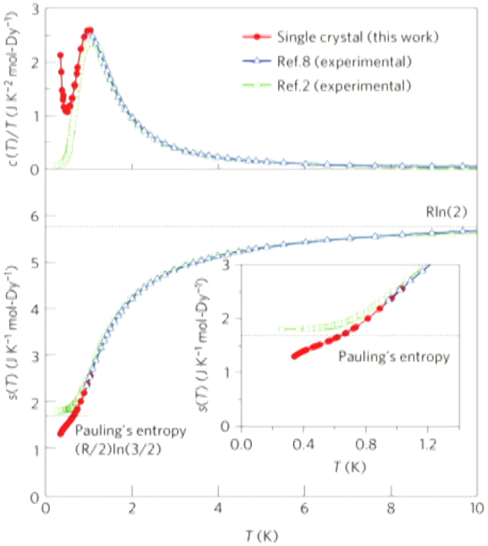
C. Catelnovo, *et al.*, Nature **451**, 42 (2008)
D.J.P. Morris, *et al.*, Science **326**, 411 (2009)

Classical dipolar spin ice

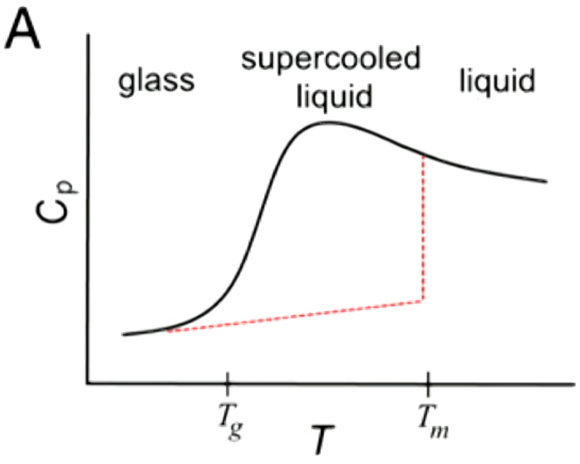
➢ However, controversies still exist

Absense of Pauling's entropy in thermally equilibrated $Dy_2Ti_2O_7$
- *possible quantum effect?*

“Glassiness” in spin ice
– *supercooled spin liquid?*



D. Pomaranski, *et al.*, Nat. Phys. **9**, 353 (2013)



E.R. Kassner, *et al.*, PNAS **112**, 8549 (2015)

Quantum spin ice

- Add quantum fluctuation in spin ice: ***anisotropic exchange Hamiltonian***

$$
\begin{aligned}
H = \sum_{\langle ij\rangle} \{ & J_{zz} S_i^z S_j^z - J_{\pm}(S_i^+ S_j^- + S_i^- S_j^+) \\
& + J_{\pm\pm}(\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^-) \\
& + J_{z\pm}[S_i^z(\zeta_{ij} S_j^+ + \zeta_{ij}^* S_j^-) + (\zeta_{ij} S_i^+ + \zeta_{ij}^* S_i^-) S_j^z]\}
\end{aligned}
$$

Transverse exchange interaction terms: $J_{\pm}$, $J_{\pm\pm}$ and $J_{z\pm}$

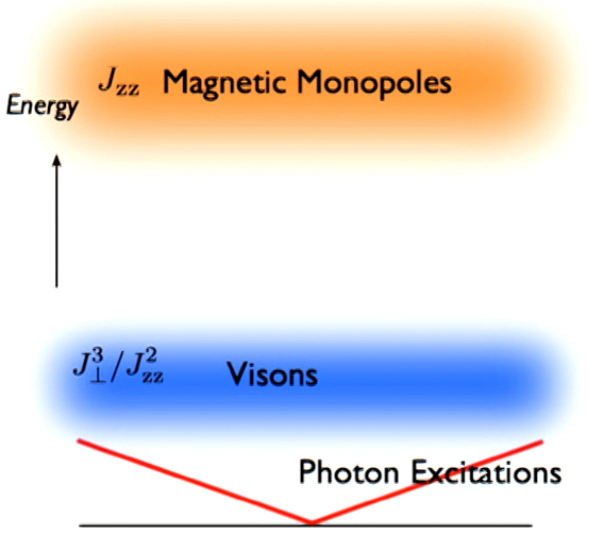
	$Ho_2Ti_2O_7$	$Yb_2Ti_2O_7$
CEF: 1st excited state	240 K	680 K
Ground state	Non-Kramers	Kramers doublet: effective S=½
g-tensor	$g_{//}\approx 19$, $g_{\perp}\approx 0$	$g_{//}\approx 1.78$, $g_{\perp}\approx 4.28$
anisotropy	Ising easy-axis anisotropy	XY easy-plane anisotropy
Localized moment	~10 μB	~3 μB

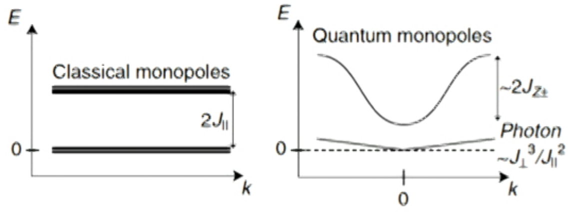
In real materials: Yb-, Pr-based pyrochlore compounds are strong candidates for QSI

Quantum spin ice

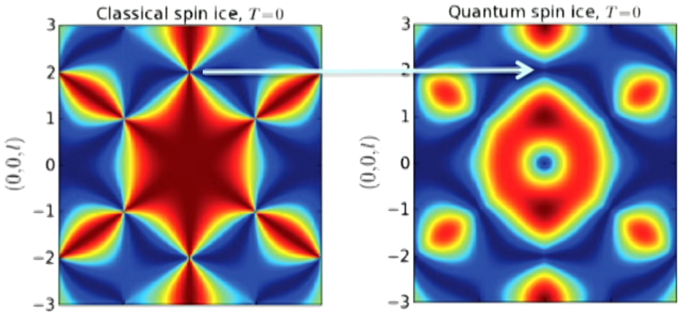
➢ QSI: emergent quantum electrodynamics (QED)

Energy landscape of QSI



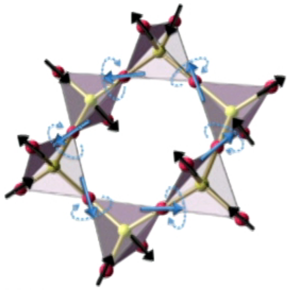


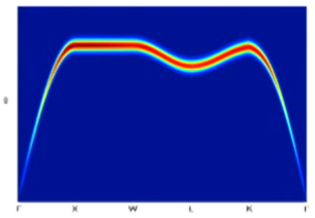
Melting of pinch-point



Emergence of artificial photons: collective spin fluctuations over hexagon-loop

-> could be observed as gapless excitations by inelastic neutron scattering





M.J.P. Gingras and P.A. McClarty, Rep. Prog. Phys. **77**, 056501 (2014)

Quantum spin ice

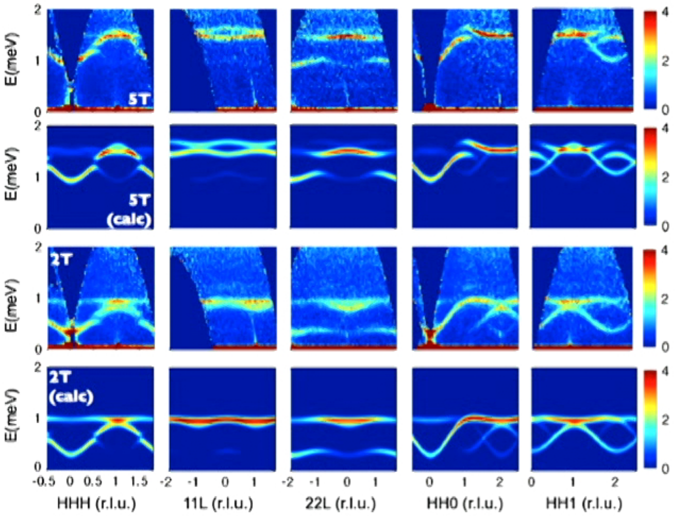
PHYSICAL REVIEW X **1**, 021002 (2011)

Quantum Excitations in Quantum Spin Ice $\boxed{Yb_2Ti_2O_7}$

Kate A. Ross,[1] Lucile Savary,[2] Bruce D. Gaulin,[1,3,4] and Leon Balents[5,*]

[1]*Department of Physics and Astronomy, McMaster University, Hamilton, Ontario, L8S 4M1, Canada*
[2]*Ecole Normale Supérieure de Lyon, 46, allée d'Italie, 69364 Lyon Cedex 07, France*
[3]*Canadian Institute for Advanced Research, 180 Dundas St. W., Toronto, Ontario, M5G 1Z8, Canada*
[4]*Brockhouse Institute for Materials Research, McMaster University, Hamilton, Ontario, L8S 4M1, Canada*
[5]*Kavli Institute for Theoretical Physics, University of California, Santa Barbara, California, 93106-4030, USA*

(Received 22 July 2011; published 3 October 2011)

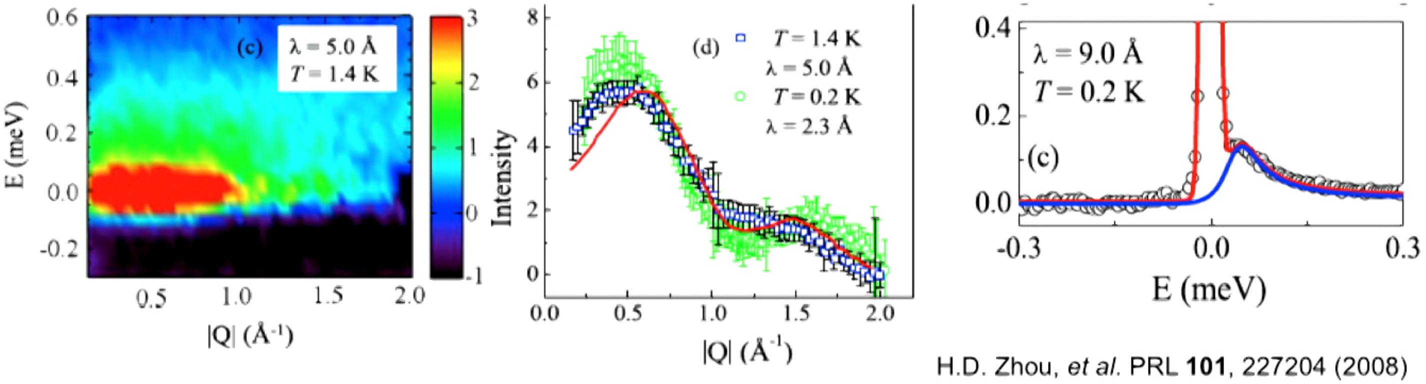


$$J_{zz} = 0.17 \pm 0.04, \qquad J_{\pm} = 0.05 \pm 0.01,$$
$$J_{\pm\pm} = 0.05 \pm 0.01, \qquad J_{z\pm} = -0.14 \pm 0.01,$$

➔Presence of significant transverse exchange terms!

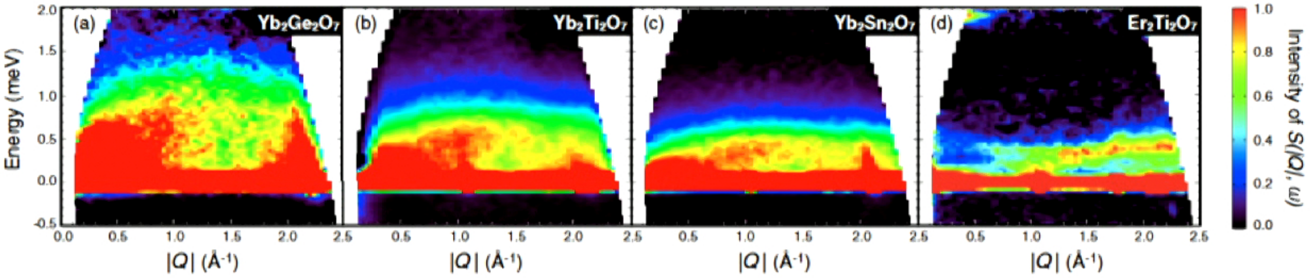
Spin dynamics in QSI candidates

$Pr_2Sn_2O_7$



H.D. Zhou, *et al.* PRL **101**, 227204 (2008)

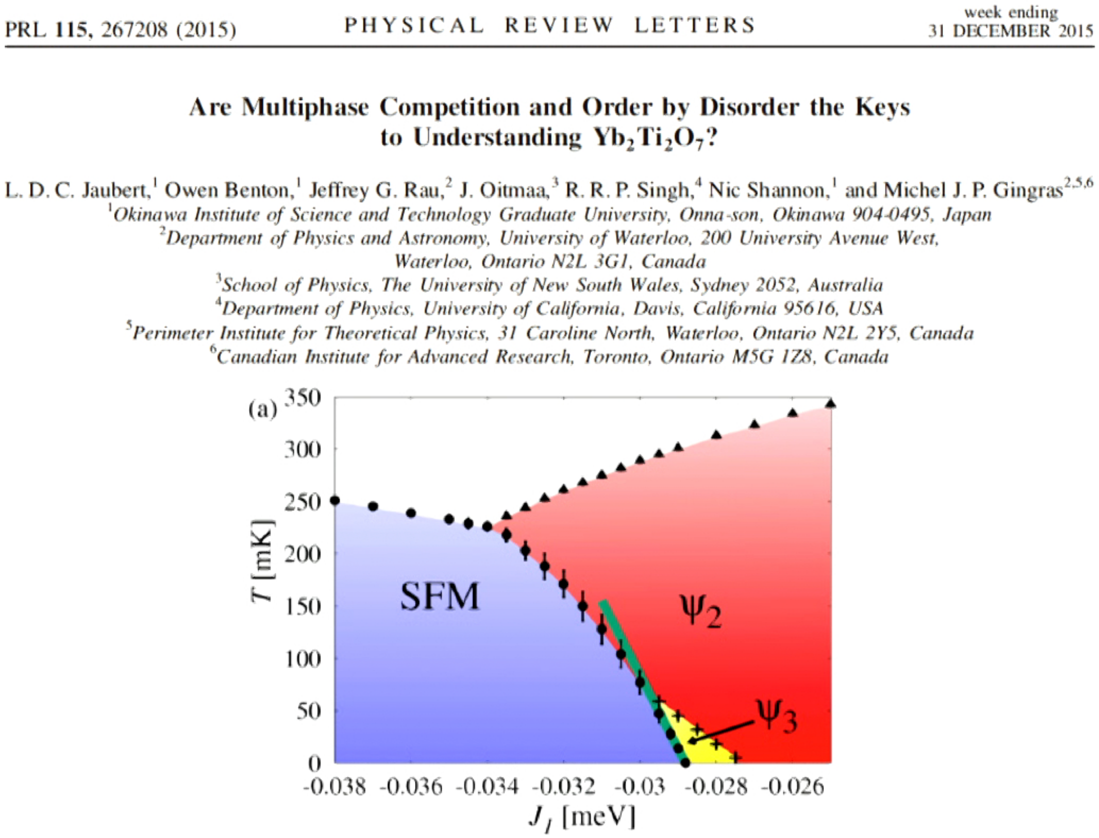
$Yb_2X_2O_7$



A.M. Hallas, *et al.* Phys. Rev. B **93**, 100403(R) (2016)

2. *Quasi-collinear* ferromagnetic structure and gapped magnetic excitations in $Yb_2Ti_2O_7$

$Yb_2Ti_2O_7$ – competing orders?



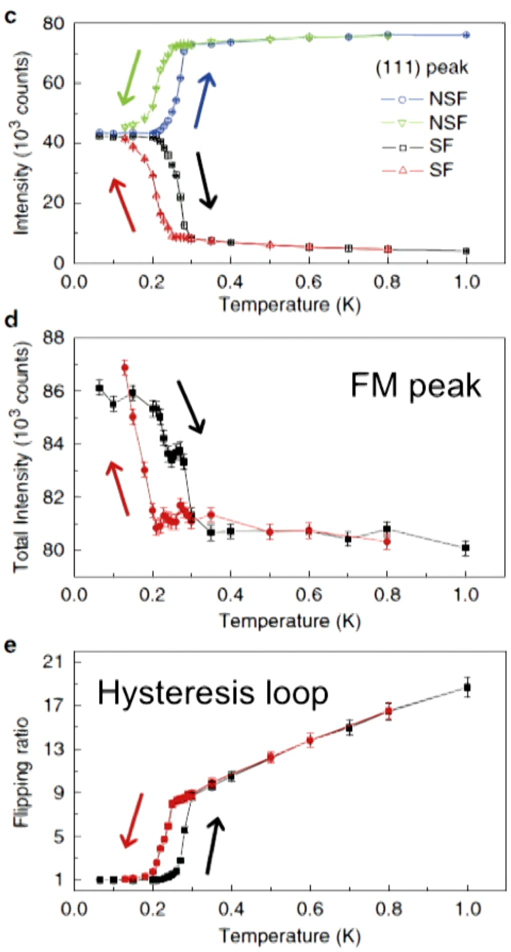
PRL 115, 267208 (2015) PHYSICAL REVIEW LETTERS week ending 31 DECEMBER 2015

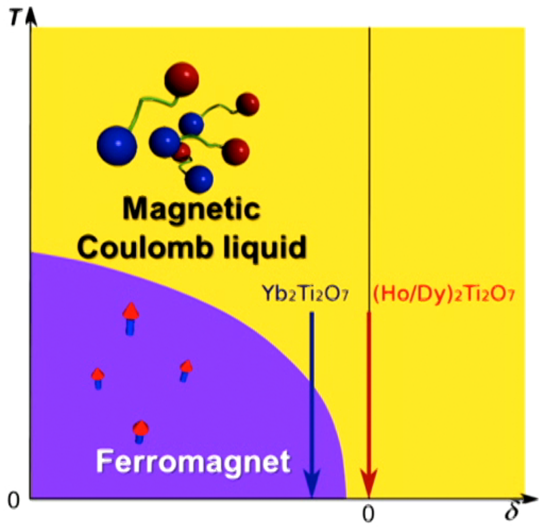
Are Multiphase Competition and Order by Disorder the Keys to Understanding $Yb_2Ti_2O_7$?

L. D. C. Jaubert,[1] Owen Benton,[1] Jeffrey G. Rau,[2] J. Oitmaa,[3] R. R. P. Singh,[4] Nic Shannon,[1] and Michel J. P. Gingras[2,5,6]
[1]*Okinawa Institute of Science and Technology Graduate University, Onna-son, Okinawa 904-0495, Japan*
[2]*Department of Physics and Astronomy, University of Waterloo, 200 University Avenue West, Waterloo, Ontario N2L 3G1, Canada*
[3]*School of Physics, The University of New South Wales, Sydney 2052, Australia*
[4]*Department of Physics, University of California, Davis, California 95616, USA*
[5]*Perimeter Institute for Theoretical Physics, 31 Caroline North, Waterloo, Ontario N2L 2Y5, Canada*
[6]*Canadian Institute for Advanced Research, Toronto, Ontario M5G 1Z8, Canada*

Strong sample dependence!

1st-order phase transition in $Yb_2Ti_2O_7$





Single crystals grown by Y. Yasui
(Nagoya University, Japan)

L.J. Chang, *et al.*, Nat. Commun. **3**, 992 (2012)

Stoichiometry of polycrystalline $Yb_2Ti_2O_7$

Neutron powder diffraction refinement

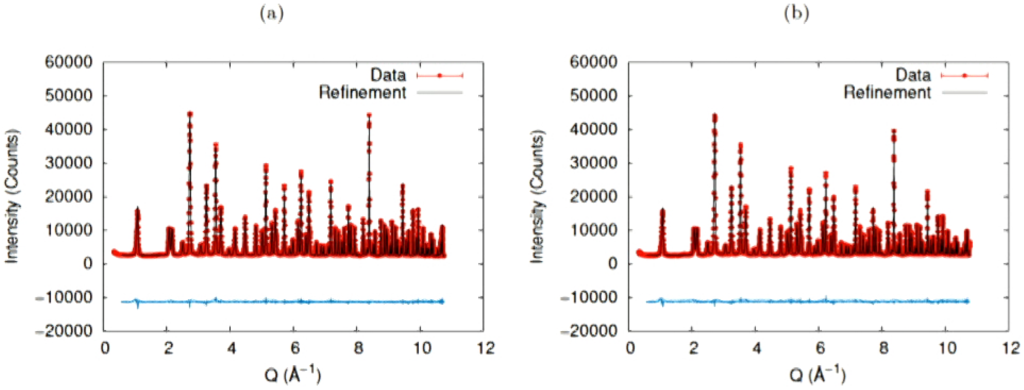


FIG. 1. Data (red dots) and best refinement (black lines) for (a) 2 K and (b) 150 K. The difference between calculated and measured intensities is given by the blue line below each pattern.

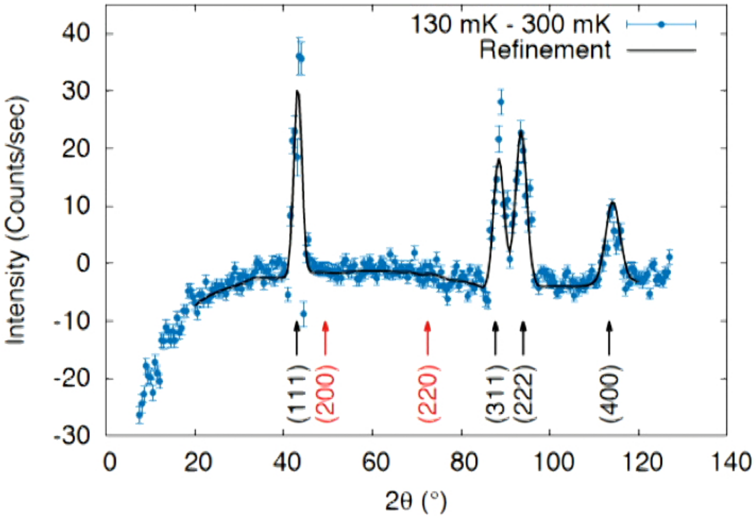
Temperature	Occupancy of Yb^{3+} at 16c site	Lattice parameter (Å)	x	R_{wp}
Stoichiometric model				
2 K	-	10.01275(2)	0.33140(5)	0.0671
150 K	-	10.02061(2)	0.33105(6)	0.0787
1.5 K (Yaouanc *et al.*)	-	10.0220(5)	0.332(1)	-
150 K (Ross *et al.*)	-	10.01111(3)	0.33122(3)	0.0414
Stuffed model				
2 K	0.0009(1)	10.01273(2)	0.33126(5)	0.0670
150 K	0.0010(1)	10.02059(2)	0.33095(6)	0.0785
150 K (Ross el al.)	0.002(1)	10.01111(3)	0.33121(3)	0.0415

TABLE I. Refined lattice parameters and oxygen 48f Wyckoff position of the space group for the stoichiometric and stuffed models. For comparison, results from Ross *et al.* [18] and Yaouanc *et al.* [2] are also shown. The refinement uncertainties are not mentioned in Ref. [2].

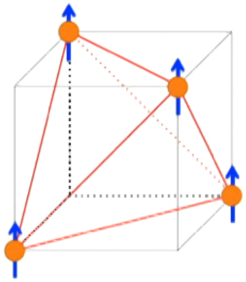
Viviane Pecanha-Antonio, *et al.* (in preparation)

Quasi-collinear ferromagnetic structure

Determination of LT magnetic structure

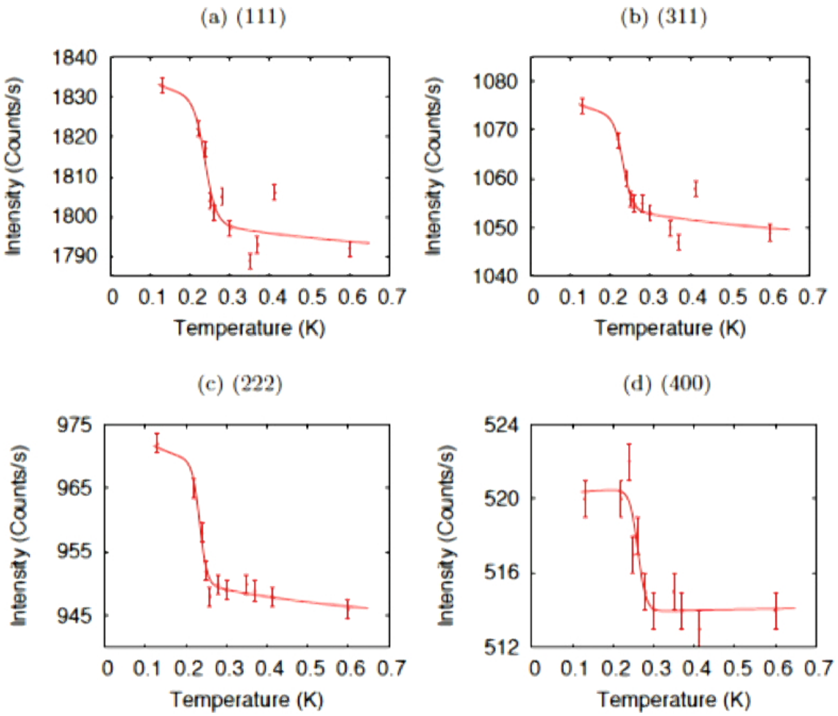


Site	X	Y	Z	$m^a(\mu_B)$	$m^b(\mu_B)$	$m^c(\mu_B)$
1	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	0.00(2)	0.00(2)	0.87(1)
2	$\frac{1}{4}$	$\frac{1}{4}$	$\frac{1}{2}$	-0.00(2)	-0.00(2)	0.87(1)
3	$\frac{1}{2}$	$\frac{1}{4}$	$\frac{1}{4}$	-0.00(2)	0.00(2)	0.87(1)
4	$\frac{1}{4}$	$\frac{1}{2}$	$\frac{1}{4}$	0.00(2)	-0.00(2)	0.87(1)

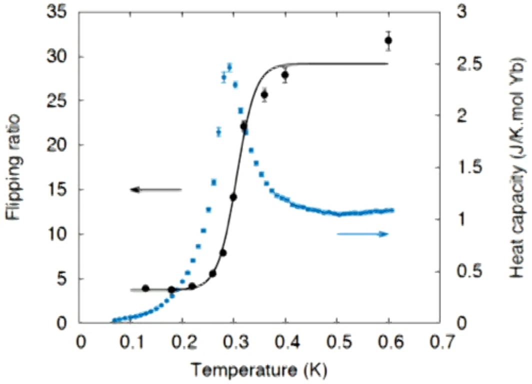


Viviane Pecanha-Antonio, *et al*. (in preparation)

Well-resolvable LT phase transition in $Yb_2Ti_2O_7$



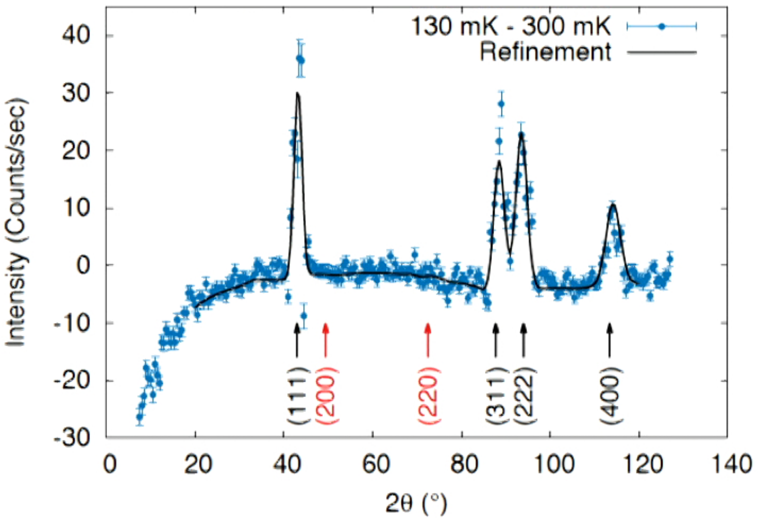
Neutron diffraction



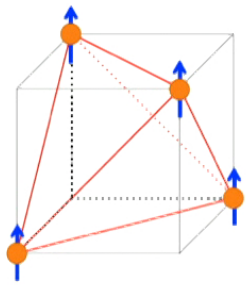
Heat capacity
Neutron depolarisation

Quasi-collinear ferromagnetic structure

Determination of LT magnetic structure



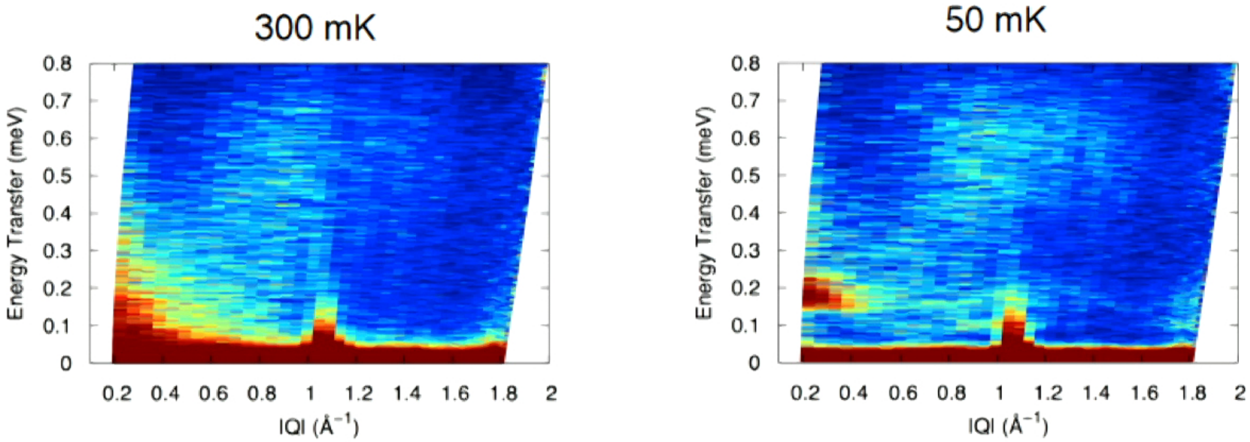
Site	X	Y	Z	$m^a(\mu_B)$	$m^b(\mu_B)$	$m^c(\mu_B)$
1	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	0.00(2)	0.00(2)	0.87(1)
2	$\frac{1}{4}$	$\frac{1}{4}$	$\frac{1}{2}$	-0.00(2)	-0.00(2)	0.87(1)
3	$\frac{1}{2}$	$\frac{1}{4}$	$\frac{1}{4}$	-0.00(2)	0.00(2)	0.87(1)
4	$\frac{1}{4}$	$\frac{1}{2}$	$\frac{1}{4}$	0.00(2)	-0.00(2)	0.87(1)



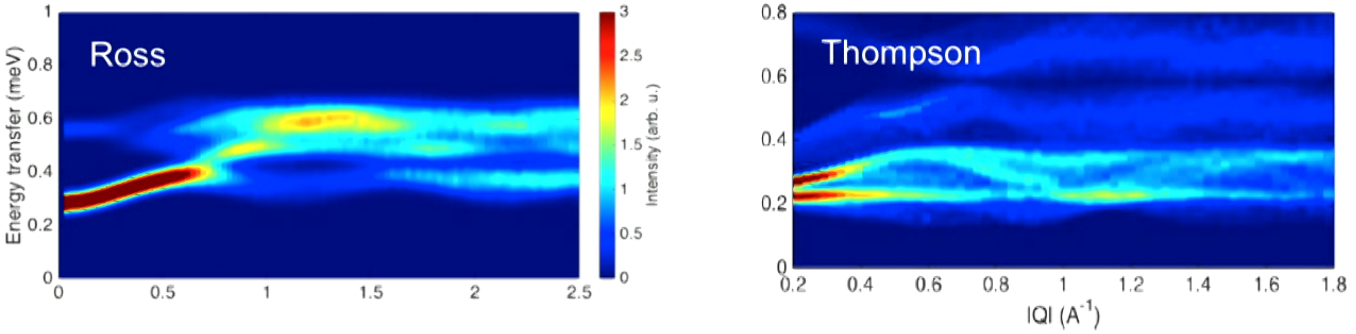
Viviane Pecanha-Antonio, *et al*. (in preparation)

Gapped magnetic excitations in $Yb_2Ti_2O_7$

Inelastic neutron scattering on polycrystalline sample



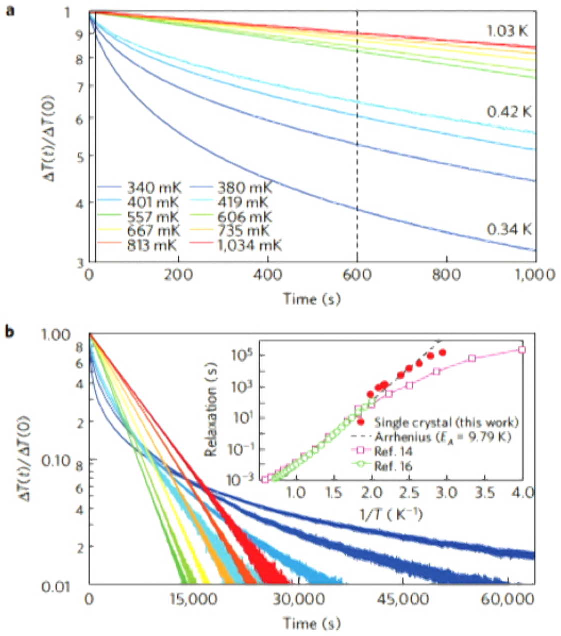
For comparison: LSWT simulations based on the known models



Viviane Pecanha-Antonio, *et al.* (in preparation)

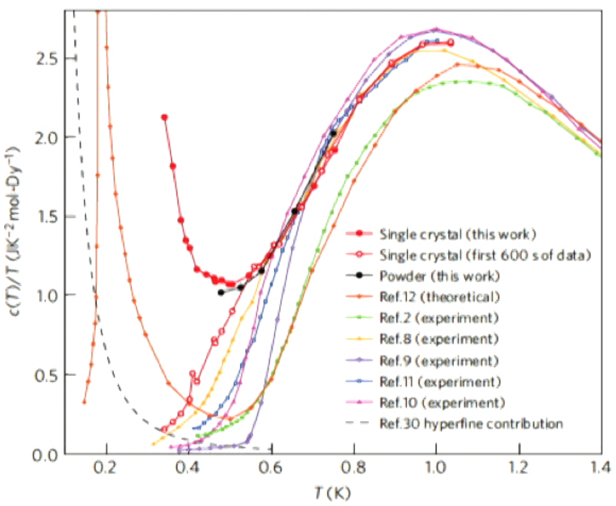
3. Approaching the ground state of classical dipolar spin ice $Dy_2Ti_2O_7$

Zero-point entropy in spin ice $Dy_2Ti_2O_7$



Estimated thermal relaxation time:

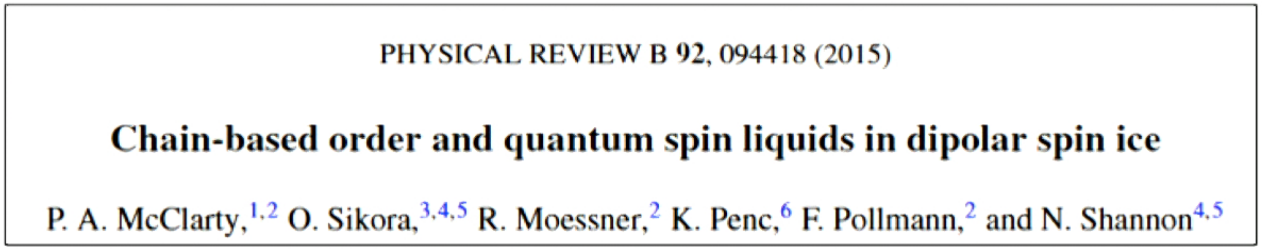
- 800mK: ~10^{-1} s
- 450mK: ~10^3-10^4 s
- 350mK: ~10^5 s



- ➢ **Extremely slow thermal relaxations ➔ out-of-equilibrium state ➔ Wien effect and monopole dynamics**
- ➢ **True ground state of spin ice?**

D. Pomaranski, *et al.*, Nat. Phys. **9**, 353 (2013)
V. Kaiser, *et al.*, Nat. Mat. **12**, 1033 (2013)

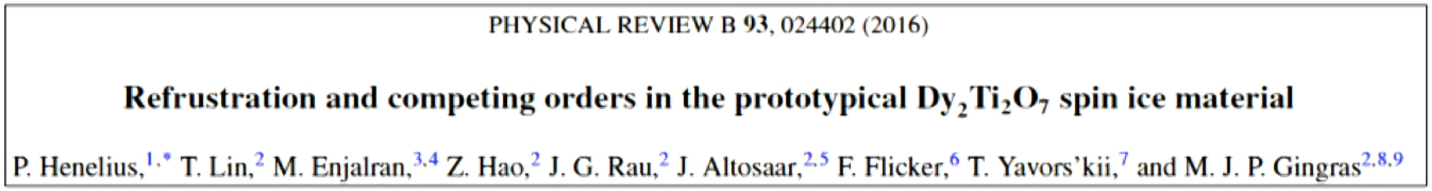
Magnetic ground state of dipolar spin ice



PHYSICAL REVIEW B **92**, 094418 (2015)

Chain-based order and quantum spin liquids in dipolar spin ice

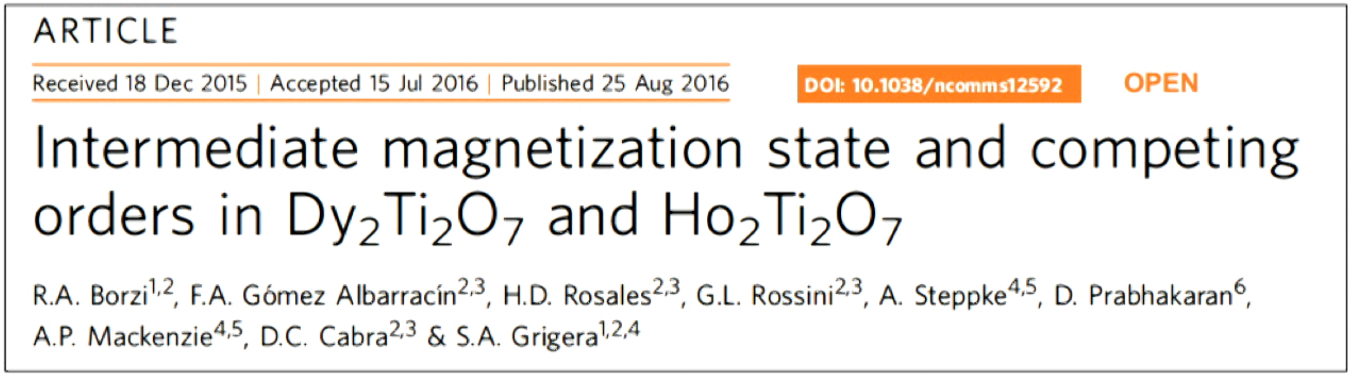
P. A. McClarty,[1,2] O. Sikora,[3,4,5] R. Moessner,[2] K. Penc,[6] F. Pollmann,[2] and N. Shannon[4,5]



PHYSICAL REVIEW B **93**, 024402 (2016)

Refrustration and competing orders in the prototypical $Dy_2Ti_2O_7$ spin ice material

P. Henelius,[1,*] T. Lin,[2] M. Enjalran,[3,4] Z. Hao,[2] J. G. Rau,[2] J. Altosaar,[2,5] F. Flicker,[6] T. Yavors'kii,[7] and M. J. P. Gingras[2,8,9]



ARTICLE

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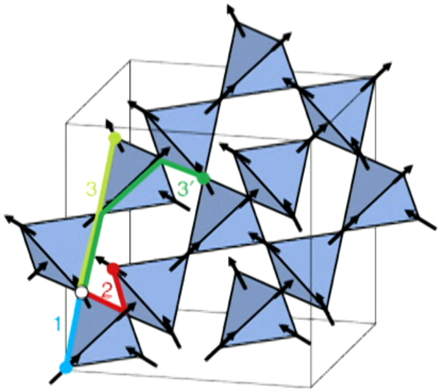
Intermediate magnetization state and competing orders in $Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$

R.A. Borzi[1,2], F.A. Gómez Albarracín[2,3], H.D. Rosales[2,3], G.L. Rossini[2,3], A. Steppke[4,5], D. Prabhakaran[6], A.P. Mackenzie[4,5], D.C. Cabra[2,3] & S.A. Grigera[1,2,4]

Competing orders and re-frustration



Model	D	J_1	J_2	J_{3a}	$(J_2/3)+J_{3a}$	J_{3b}
Upper	1.3224	3.30	-0.0949	0.07	0.038	-0.0167
Lower	1.3224	3.44	-0.1649	-0.02	-0.075	0.0433
Boundary	1.3224	3.38	-0.1349	0.02	-0.025	0.0167
g-DSM*	1.3224	3.41	-0.14	0.025	-0.0217	0.025
d-DSM	1.3224	3.41	0	-0.02	-0.02	0.07
s-DSM	1.41	1.56	0	0	0	0

1. important role of further-neighbor exchange interactions such as J_2, J_3
2. competing orders leading to re-frustration in the spin-ice regime
3. quantum effect may be enhanced due to dipole-octopole nature of ground state doublet of Dy^{3+}

P. Henelius *et al.*, Phys. Rev. B **93**, 024402 (2016).
P.A. McClarty *et al.*, Phys. Rev. B **92**, 094418 (2015).
J.G. Rau & M.J.P. Gingras, Phys. Rev. B **92**, 144417 (2015).
R.A. Borzi, *et al.*, Nat. Commun. **7**, 12592 (2016).

Estimation of possible quantum effects in spin ice

- Estimation by Jeff Rau et al. (PRB **92**, 144417 (2015)):

  $J_\pm \leq 56$ mK
  $g = 12J_\pm^3/J_{ZZ}^2 \sim 2$ mK (if taking $J_{ZZ} = 1$ K)

- Estimation by P. Henelius et al. (PRB **93**, 024042 (2016)):

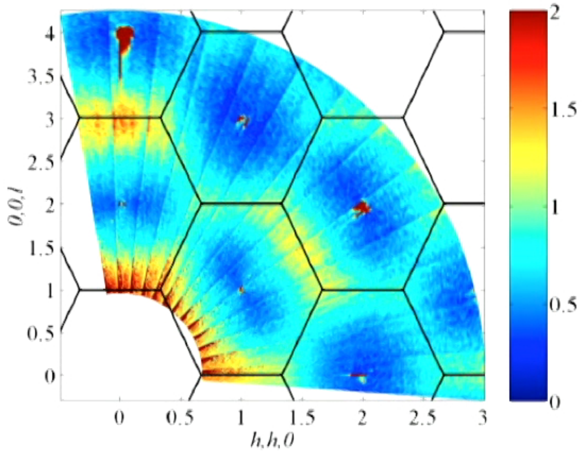
  $J_\pm \sim 3$ mK
  $g = 12J_\pm^3/J_{ZZ}^2 \sim 0.05$ mK (if taking $J_{ZZ} = 4$ K)

- Estimation by P.A. McClarty et al. (PRB **92**, 094418 (2016)):

  $g = 12J_\pm^3/J_{ZZ}^2 \sim 70$ mK could stabilize QSI for DTO

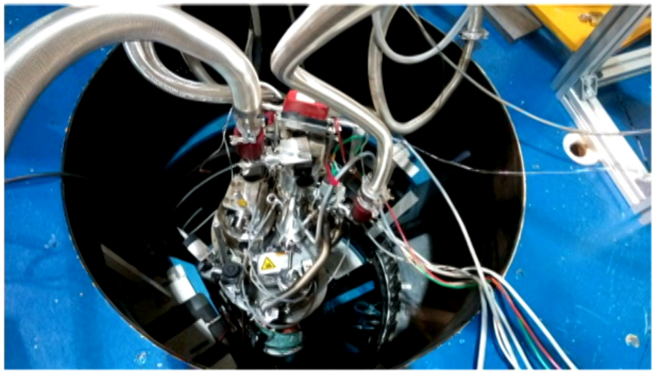
Experimental challenges with neutrons

1. strong neutron absorption of Dy^{3+} (σ_{abs}>4600 barn at 4.2 Å)
2. large incoherent cross-section of Dy^{3+} (~54 barn/Dy) -> high background
3. extremely slow thermal relaxation to equilibrium below ~600 mK



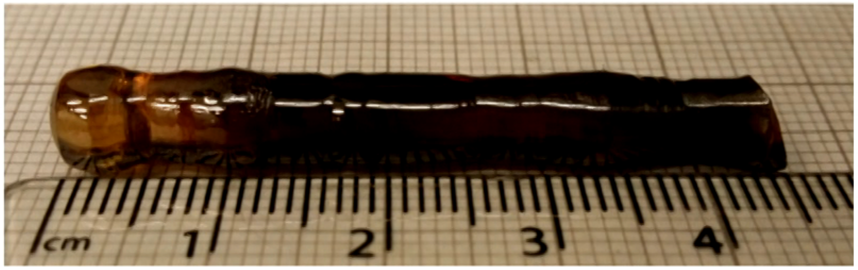
$^{162}Dy_2Ti_2O_7$

T. Fennell, *et al.* Phys. Rev. B **70**, 134408 (2004);
T. Yavors'kii, *et al.* Phys. Rev. Lett. **101**, 037204 (2008)



Cryogen-free dilution- and ^{3}He-inserts at MLZ
-> long time and stable operation

New generation of isotope-enriched $^{162}Dy_2Ti_2O_7$ single crystal

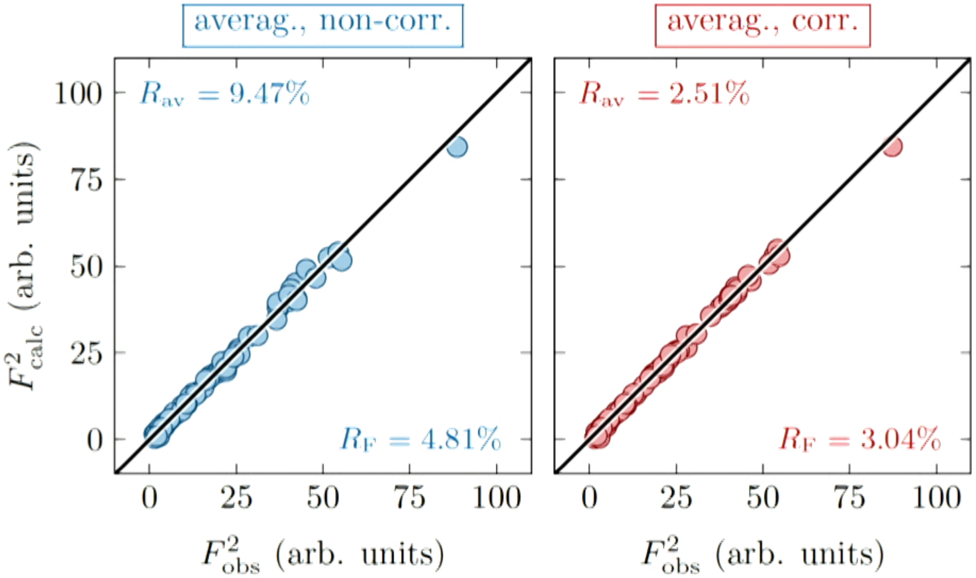


1. much reduced neutron absorption: σ_{abs}<1000 barn at 4.2 Å
2. much reduced incoherent scattering background: 0 barn/^{162}Dy
3. grown under optimal conditions to reduce possible stuffing and oxygen defects

M. Ciomaga Hatnean
G. Balakrishnan
(University of Warwick)

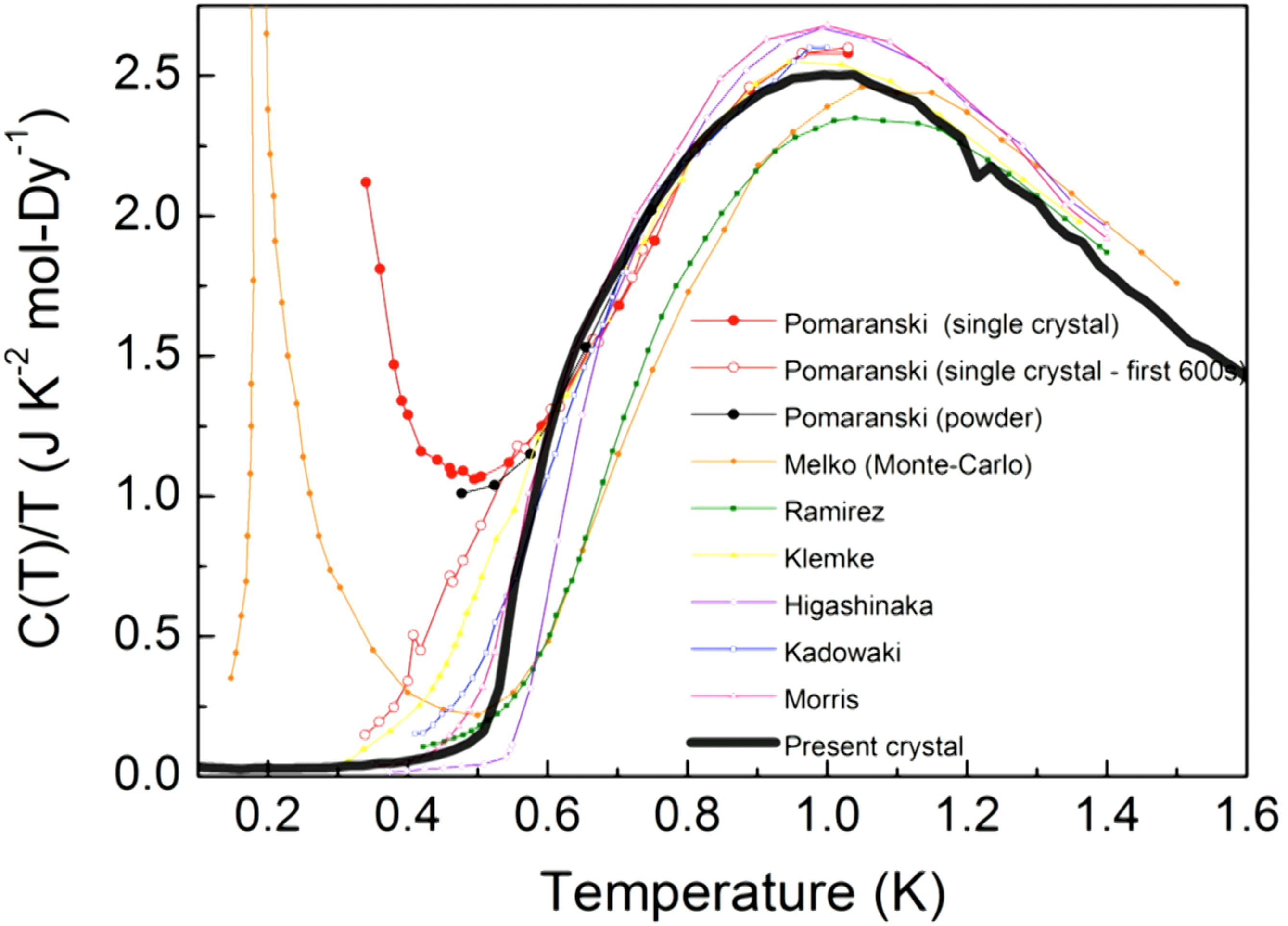
Stoichiometry of crystal

- hot-neutron diffractometer POLI@MLZ: wavelength λ=0.7 Å
- 296 reflections measured on a ~200 mg single crystal
- absorption correction via the CCSL package



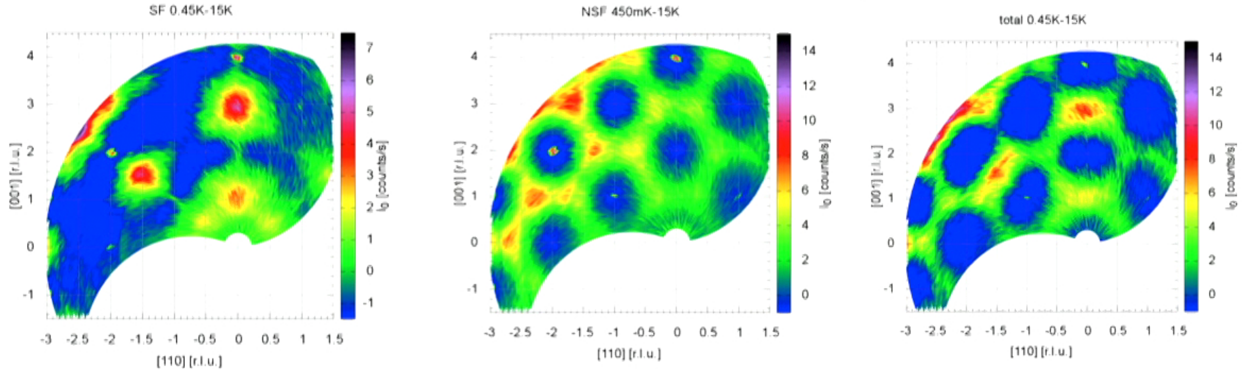
	162-Dy	natural-Dy	Ti	O(1)	O(2)
Occupancy	0.91(2)	0.09(2)	1.00(2)	0.99(2)	1.00(2)

Low-temperature heat capacity

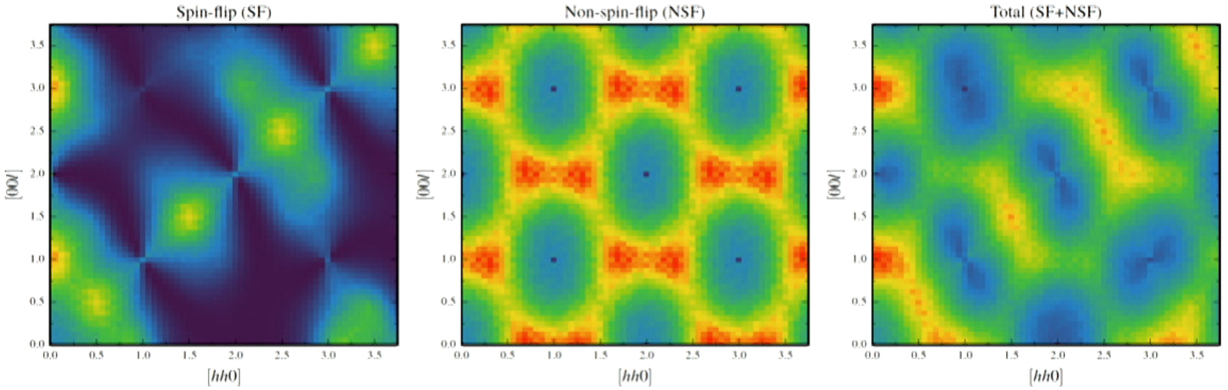


Magnetic diffuse scattering on $^{162}Dy_2Ti_2O_7$

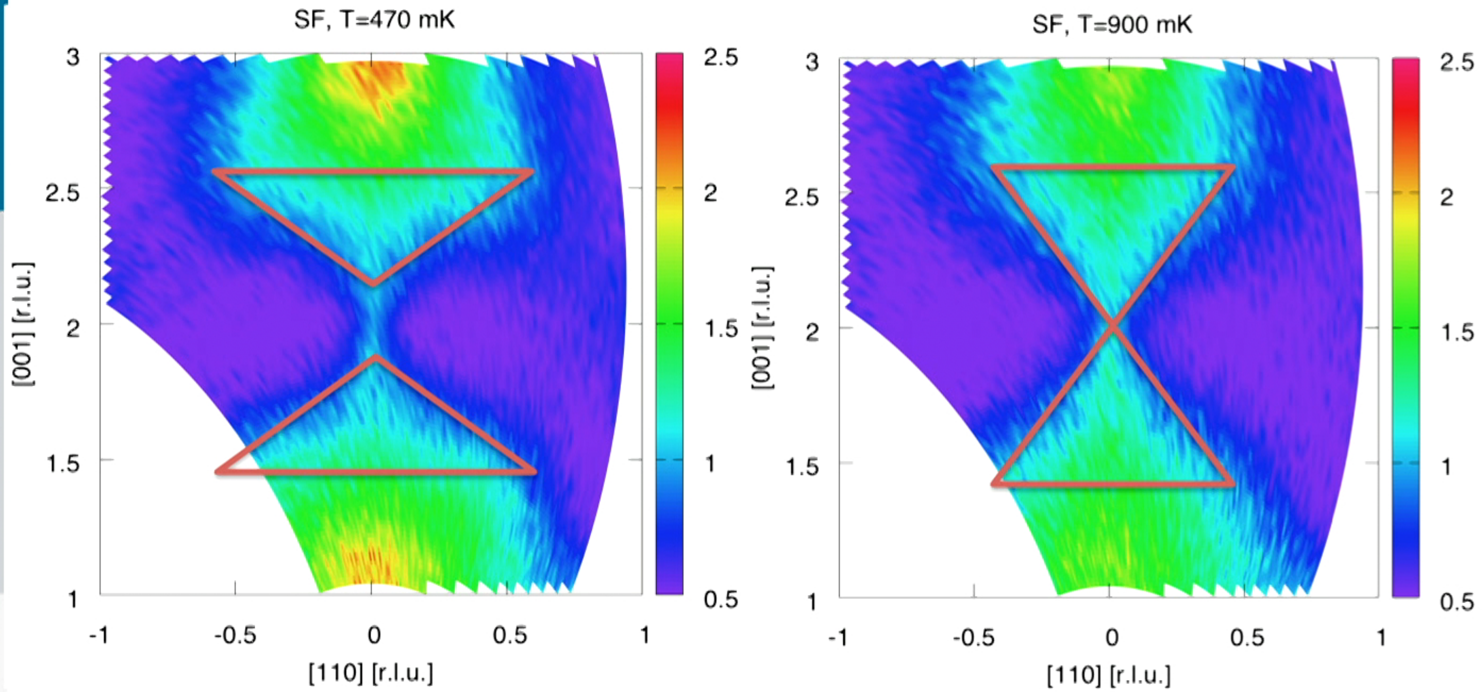
Polarized neutron scattering



Monte-Carlo simulations based on g-DSM* (J.G. Rau and M. Gingras)

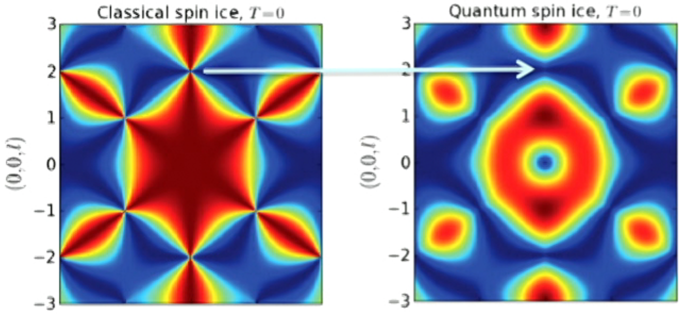


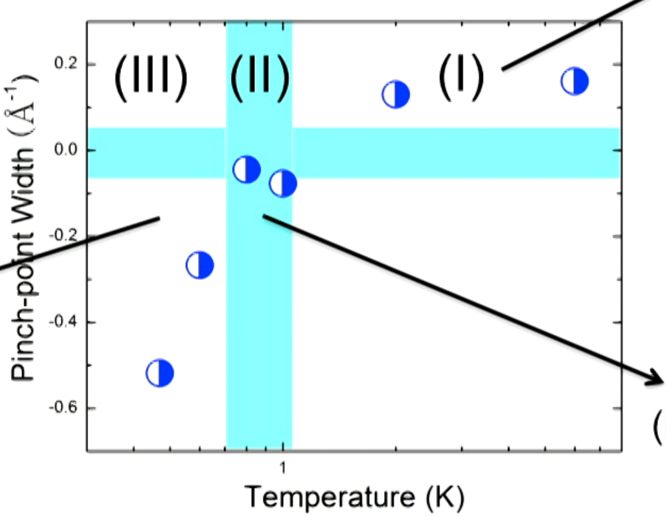
Fate of pinch point



Fate of pinch point

Melting of pinch-point
(Owen Benton)





Defected monopole topological phase

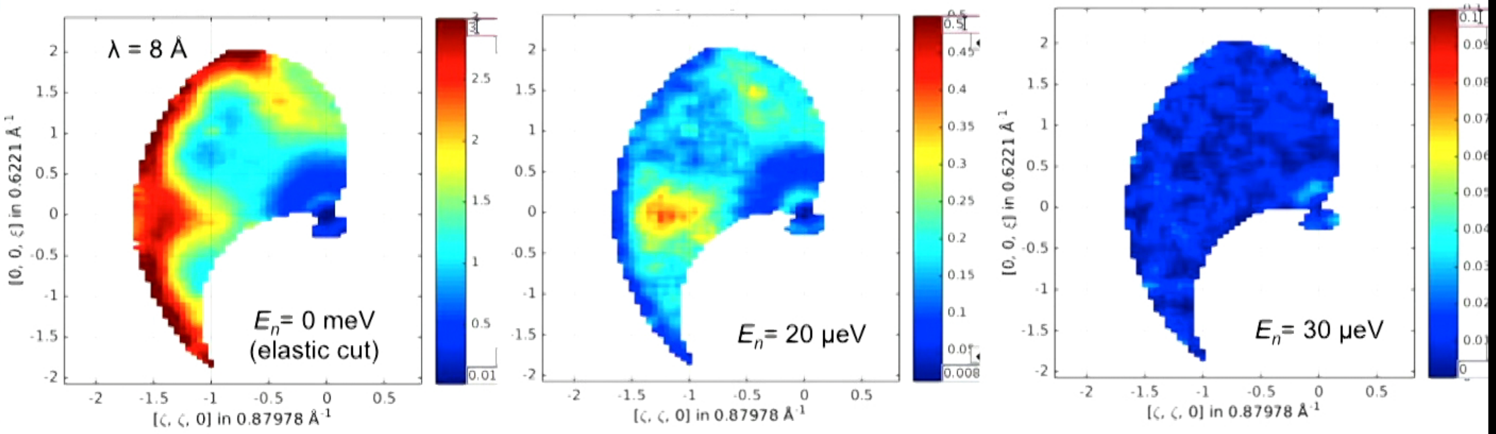
Possible new monopole topological phase?

Ideal monopole topological phase (i.e. all monopoles are Dirac string bound)

Jülich Centre for Neutron Science

INS on $^{162}Dy_2Ti_2O_7$: quantum dynamics?

Heinz Maier-Leibnitz Zentrum

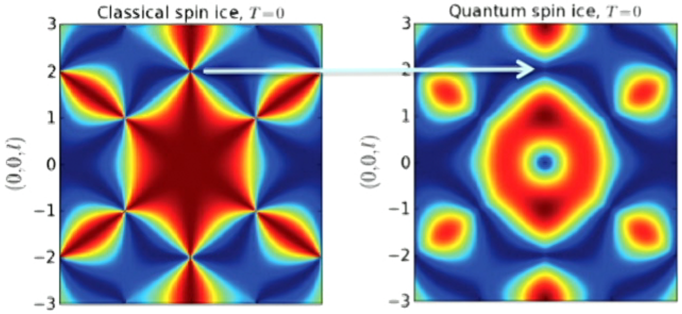


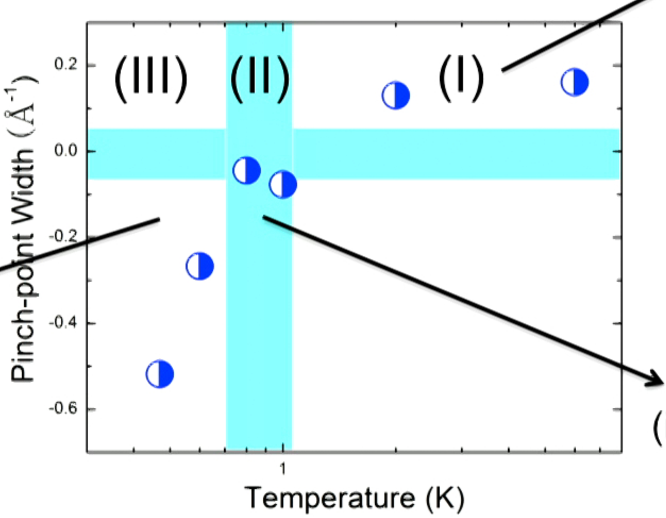
- magnetic scattering almost entirely elastic
- however, indication exists for an extended quasi-elastic component with energy transfer up to ~20 μeV
  - ➢ upper limit for any possible quantum effects
  - ➢ possibly due to competing orders?

IN5@ILL

Fate of pinch point

Melting of pinch-point (Owen Benton)





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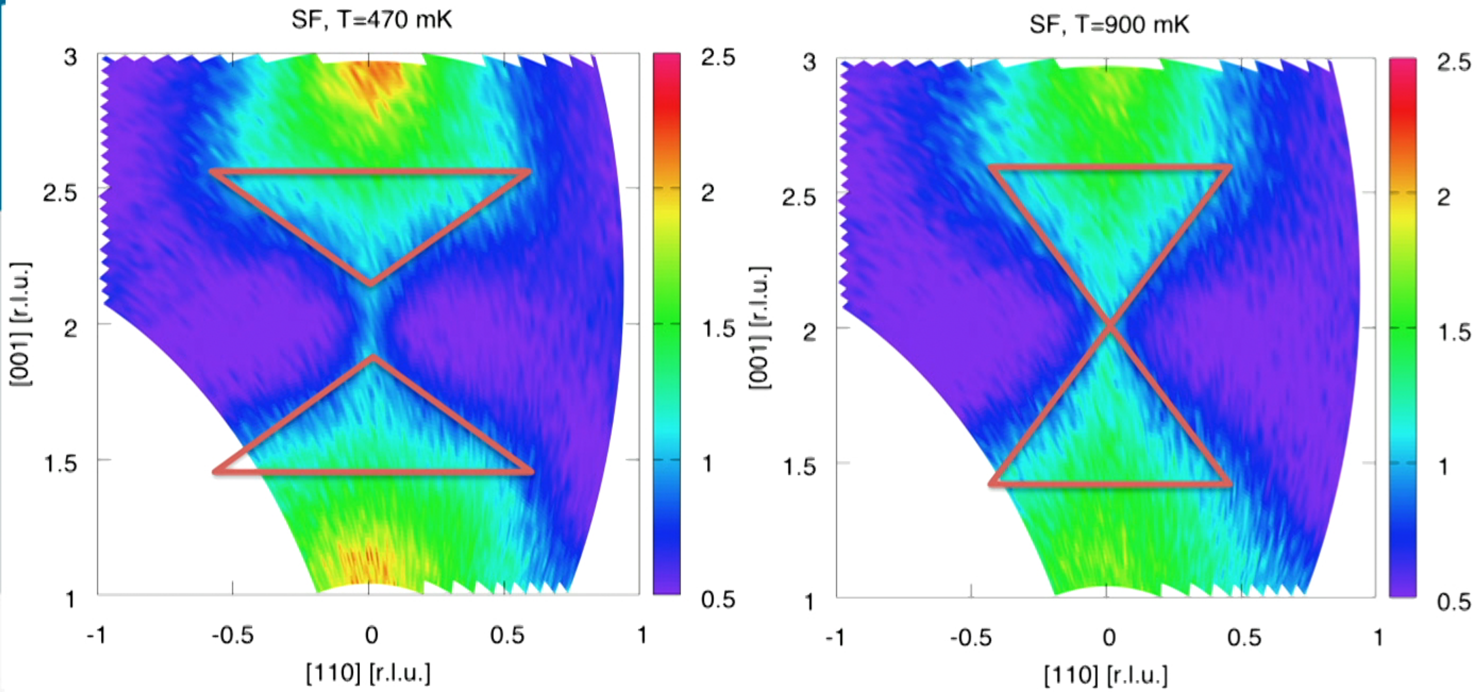
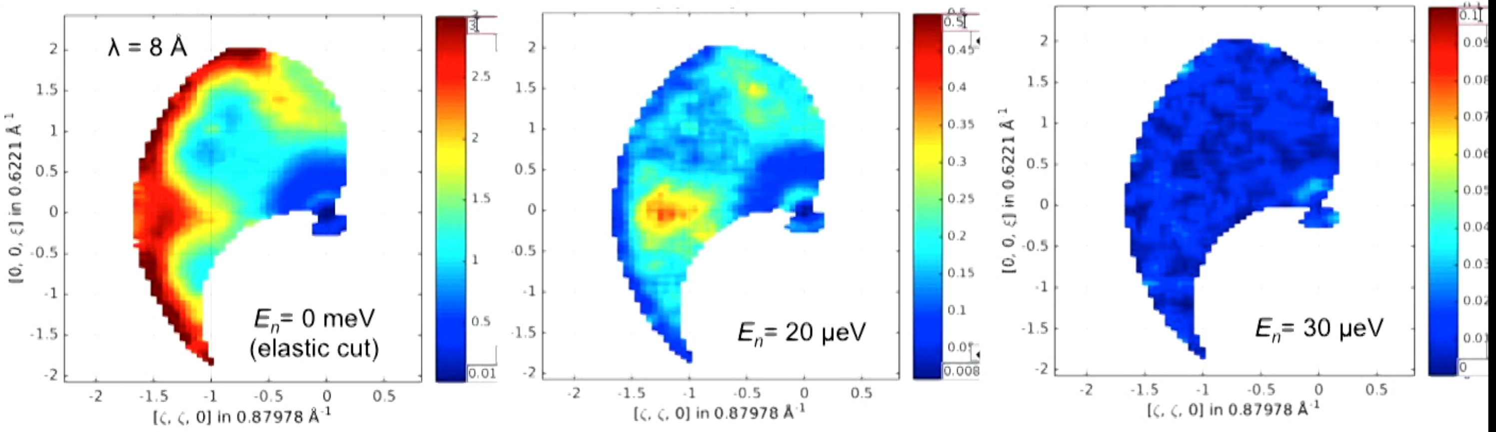


INS on $^{162}Dy_2Ti_2O_7$: quantum dynamics?



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Summary

1. Magnetic ground state of $Yb_2Ti_2O_7$
   - Quasi-collinear ferromagnetic structure
   - Gapped magnetic excitations

2. Magnetic ground state of dipolar spin ice $Dy_2Ti_2O_7$
   - Polarized diffuse scattering data ⇔ various DSM models
   - Evidence of possible melting of pinch point
   - Competing orders and re-frustration

Thank you for your attention!

MLZ is a cooperation between: