Title: Antiferroquadrupolar correlations in the quantum spin ice candidate Pr2Zr2O7

Date: Jun 08, 2017 02:25 PM

URL: http://pirsa.org/17060053

Abstract: We present an experimental study of the quantum spin ice candidate pyrochlore compound Pr2Zr2O7 by means of magnetization measurements, specic heat and neutron scattering. We confirm that the spin excitation spectrum is essentially inelastic [1] and consists in a broad flat mode centered at about 0.4 meV with a magnetic structure factor which resembles the spin ice pattern. The new experimental results obtained under an applied magnetic field, interpreted in the light of mean field calculations, draw a new picture where quadrupolar interactions play a major role and overcome the magnetic exchange coupling. We determine a range of acceptable parameters able to account for the observations and propose that the actual ground state of this material is an antiferroquadrupolar liquid with spin-ice like excitations [2]. The influence of disorder is also discussed.

Antiferroquadrupolar correlations in the QSI $Pr_2Zr_2O_7$

S. Petit, N Martin, P. Bonville, S. Guitteny, I. Mirebeau — *LLB, CEA-CNRS Saclay, France*

E. Lhotel, J. Robert — *Institut Néel, Grenoble, France*

C. Decorse — *ICMMO, Orsay Univ., France*

M. Ciomaga-Hatnean, G. Balakrishnan — *Dpt of Physics, Warwick Univ, UK*

Our point:

1) Role of transverse interactions

PHYSICAL REVIEW B **94**, 165153 (2016)

$$\mathcal{H} = \frac{1}{2}\sum_{<i,j>} \mathcal{J}^{zz}\sigma_i^z\sigma_j^z + \sum_i (g_{\|}\mu_B \vec{z}_i \cdot \vec{h})\ \sigma_i^z$$
$$+\frac{1}{2}\sum_{<i,j>} -\mathcal{J}^{\pm}\left(\sigma_i^+\sigma_j^- + \sigma_i^-\sigma_j^+\right)$$
$$+\frac{1}{2}\sum_{<i,j>} \mathcal{J}^{\pm\pm}\left(\gamma_{ij}\sigma_i^+\sigma_j^+ + \gamma_{ij}^*\sigma_i^-\sigma_j^-\right)$$

Jeff's talk
S. Onoda et al,
S.B. Lee et al

2) Role of disorder : « Quantum SI route »

PHYSICAL REVIEW B **94**, 134428 (2016)

In preparation

$$\mathcal{H}_{\mathrm{m-el}} = \sum_i v_i\ \sigma_i^+ + v_i^*\ \sigma_i^-$$

Introduction

Highly Frustrated Magnetism 2008 (HFM 2008) IOP Publishing
Journal of Physics: Conference Series **145** (2009) 012031 doi:10.1088/1742-6596/145/1/012031

Spin Freezing in the Pyrochlore Antiferromagnet $Pr_2Zr_2O_7$

K. Matsuhira[1], C. Sekine[2], C. Paulsen[3], M. Wakeshima[4], Y. Hinatsu[4], T. Kitazawa[5], Y. Kiuchi[5], Z. Hiroi[5], S. Takagi[1]

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]

PHYSICAL REVIEW B **94**, 134428 (2016)

Magnetic properties and crystal field in $Pr_2Zr_2O_7$

P. Bonville
SPEC, CEA, CNRS, Université Paris-Saclay, CEA-Saclay, 91191 Gif-sur-Yvette, France

Introduction

PHYSICAL REVIEW B **94**, 165153 (2016)

Antiferroquadrupolar correlations in the quantum spin ice candidate $Pr_2Zr_2O_7$

S. Petit,[1,*] E. Lhotel,[2,†] S. Guitteny,[1] O. Florea,[2] J. Robert,[2] P. Bonville,[3] I. Mirebeau,[1] J. Ollivier,[4] H. Mutka,[4] E. Ressouche,[5] C. Decorse,[6] M. Ciomaga Hatnean,[7] and G. Balakrishnan[7]

Materials Research
Express

Structural and magnetic properties of single-crystals of the geometrically frustrated zirconium pyrochlore, $Pr_2Zr_2O_7$

M Ciomaga Hatnean[1], C Decorse[2], M R Lees[1], O A Petrenko[1], D S Keeble[1] and G Balakrishnan[1]
[1] Physics Department, University of Warwick, Coventry, CV4 7AL, UK
[2] SP2M-ICMMO, UMR 8182, Universite Paris-Sud 11, F-91405 Orsay, France
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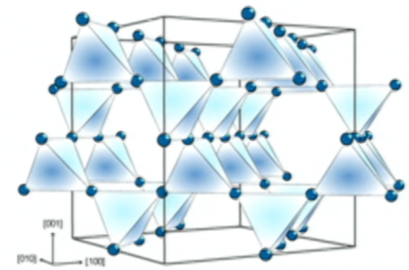
Materials Research Express 1 (2014) 026109
doi:10.1088/2053-1591/1/2/026109

PRL **118**, 107206 (2017) PHYSICAL REVIEW LETTERS week ending 10 MARCH 2017

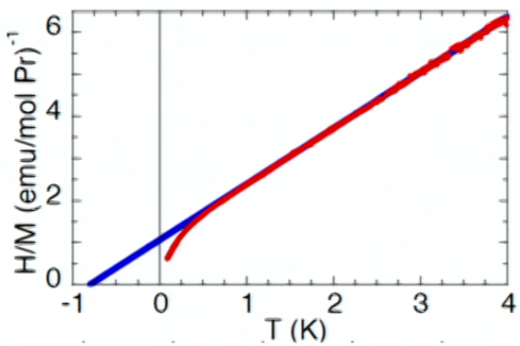
Disordered Route to the Coulomb Quantum Spin Liquid: Random Transverse Fields on Spin Ice in $Pr_2Zr_2O_7$

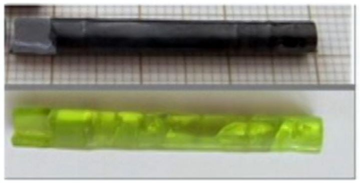
J.-J. Wen,[1,2,3] S. M. Koohpayeh,[1] K. A. Ross,[1,4] B. A. Trump,[5] T. M. McQueen,[1,5,6] K. Kimura,[7,8] S. Nakatsuji,[7,9] Y. Qiu,[4] D. M. Pajerowski,[4] J. R. D. Copley,[4] and C. L. Broholm[1,4,6]

Introduction



- A pyrochlore magnet
- Pr^{3+} is a NK ion
- Its CEF ground state is a doublet (spanning the spin ½ states) well protected from the 1st excited state (10 meV)
- No long range ordering down to 60 mK
- AF interactions, as infered from $\chi(T)$
- Crystal growth is an issue, role of disorder outlined long ago





Introduction

- C_p shows a peak at 2K, followed by an upturn at low T (interpreted in terms of monopoles dynamics)
- The neutron signal is mostly inelastic, with a lorentzian shape

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Quantum fluctuations in spin-ice-like $Pr_2Zr_2O_7$

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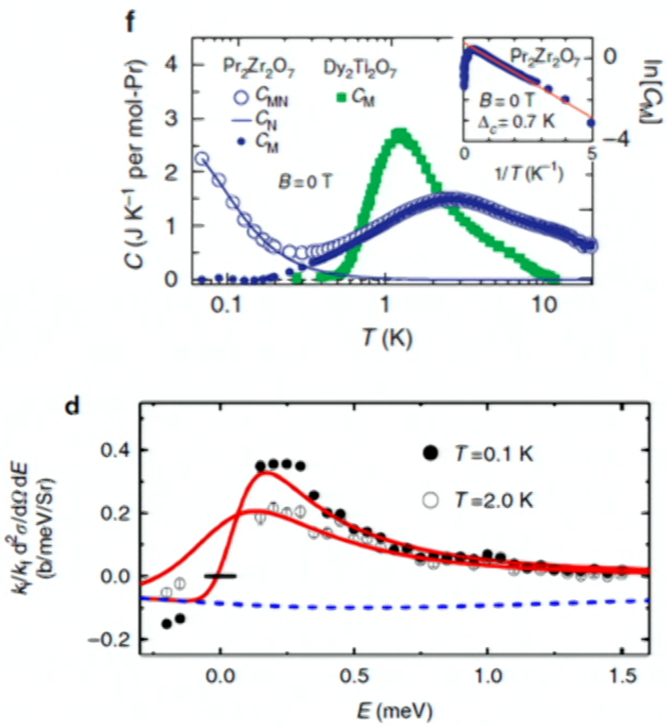

Introduction

ARTICLE
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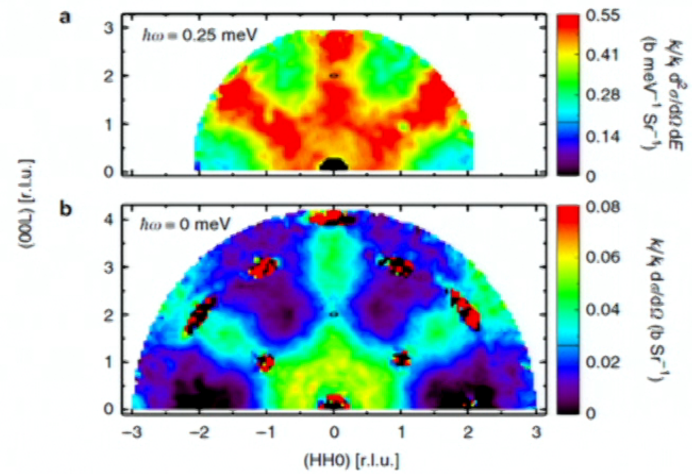
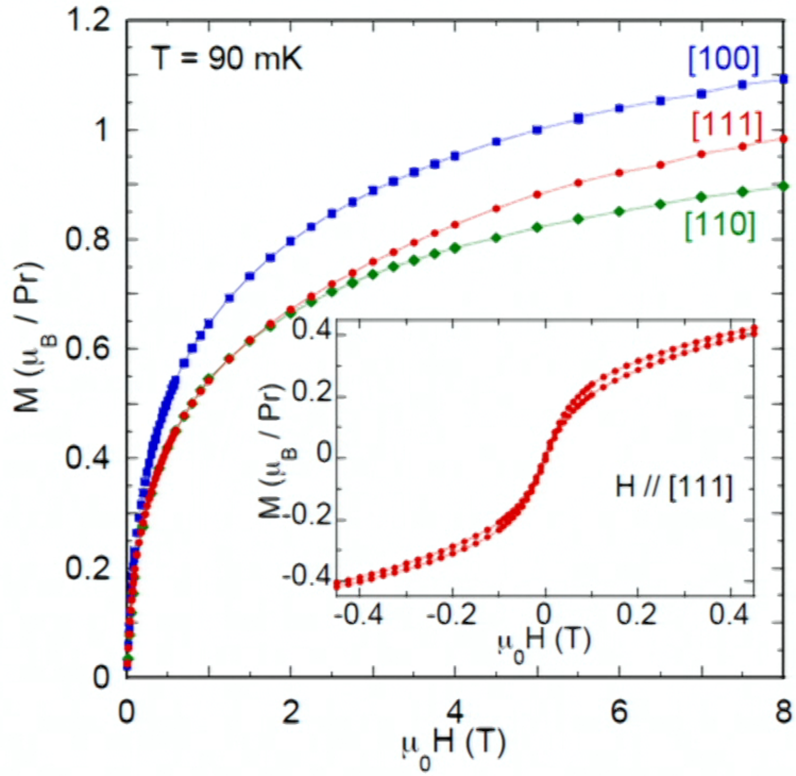
- C_p shows a peak at 2K, followed by an upturn at low T (interpreted in terms of monopoles dynamics)
- The neutron signal is mostly inelastic, with a quite **broad** lorentzian shape
- Described as a « dynamical spin ice » (the INS response resembles the spin ice pattern)



Magnetization

- M(H) struggles to grow with increasing field
- Different from the expected Ising behaviour
- Something fights against the growth of the magnetization

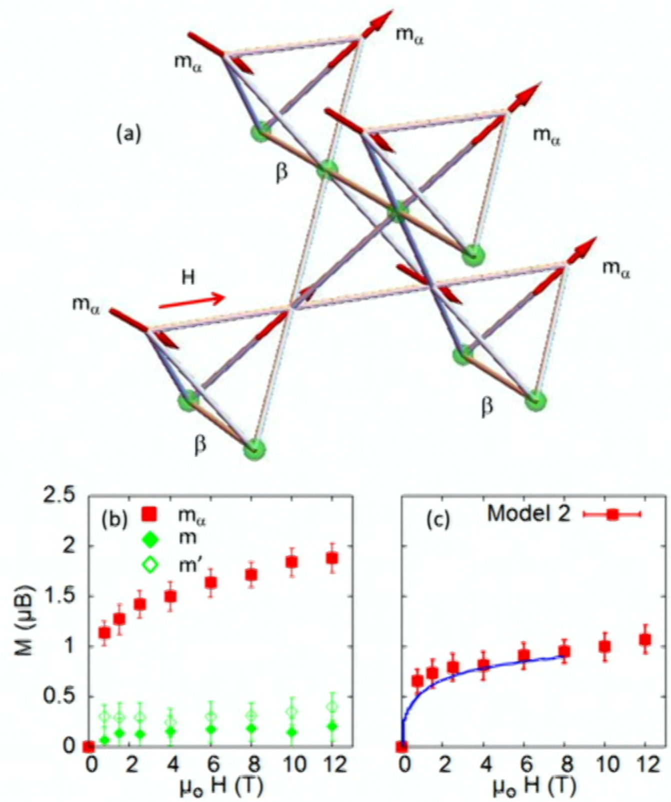


Neutron diffraction

H // (1 -1 0)

- Under applied field, no magnetic moment in the β chains inferred from neutron diffraction
- Smooth increase with H, consistent with M(H)

D23@ILL

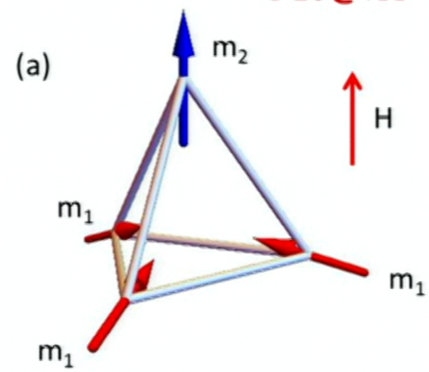


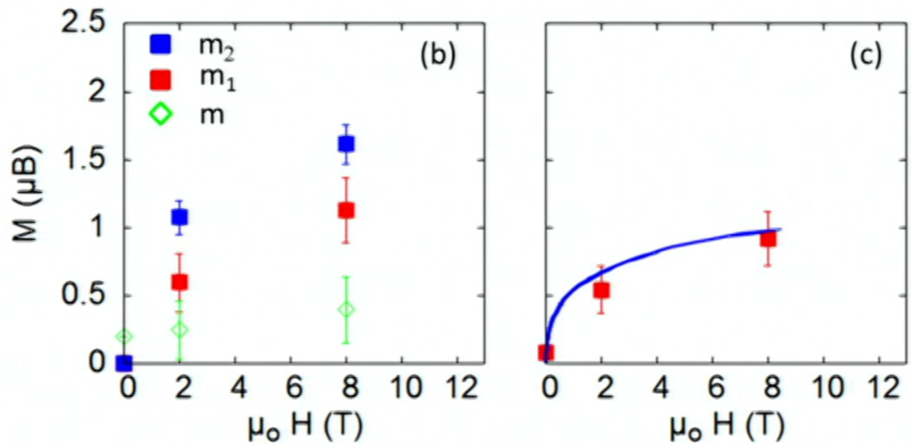
Neutron diffraction

D23@ILL

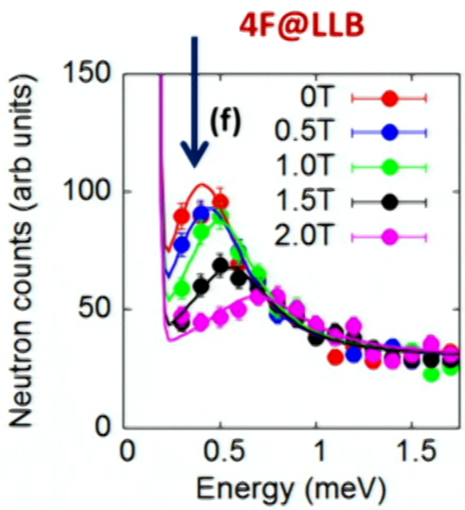
H // (1 1 1)

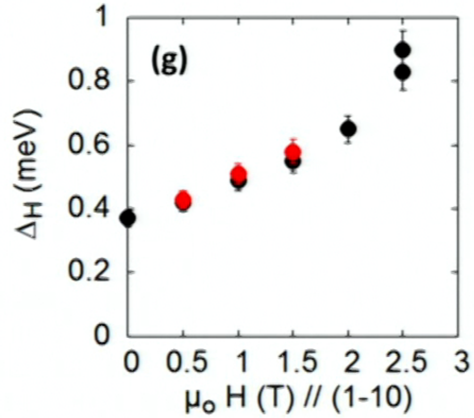
Smooth increase with H, consistent with M(H)

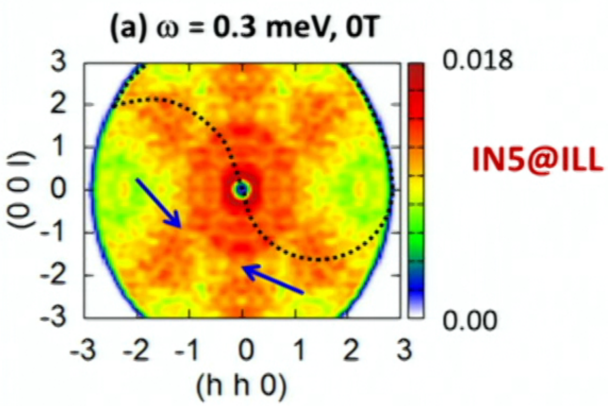




Inelastic neutron scattering

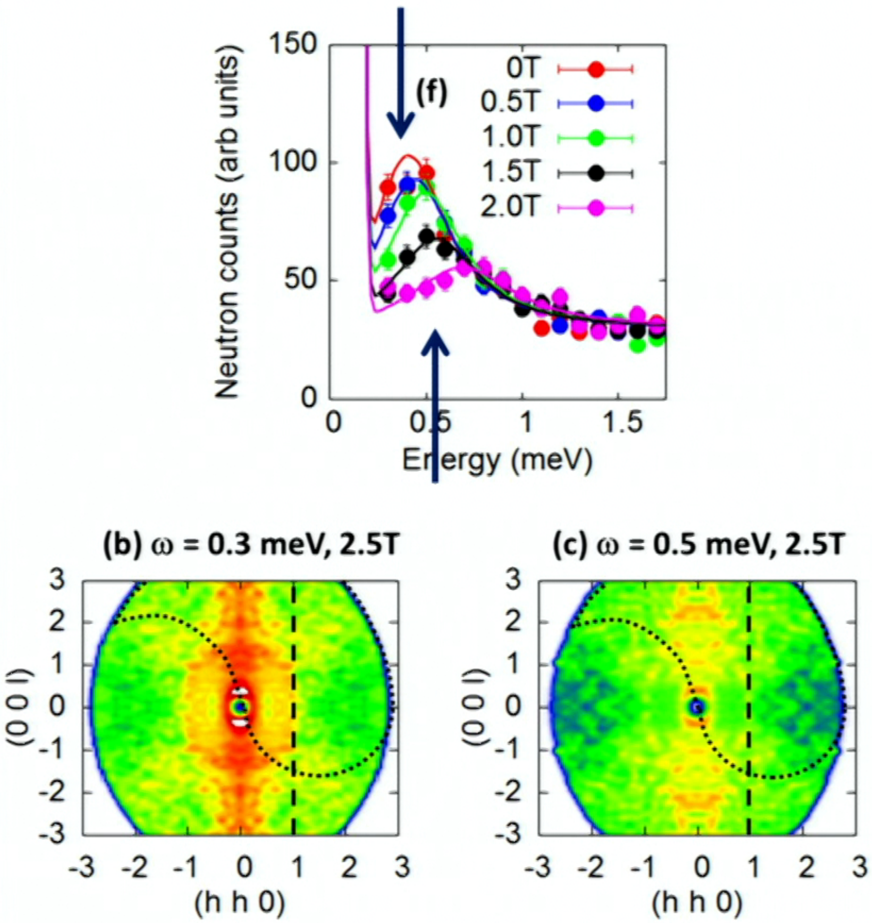




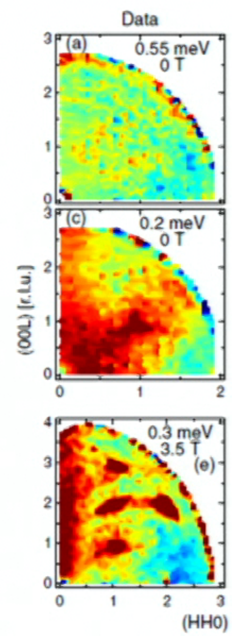


- Under applied field H//(1-1-0), the peak of the lorenztian profile shifts to higher energy
- Confirm the dynamical spin ice pattern

Inelastic neutron scattering



PRL 118, 107206 (2017)



Input of MF

$$\mathcal{H} = \frac{1}{2}\sum_{<i,j>} \mathcal{J}^{zz}\sigma_i^z\sigma_j^z + \sum_i (g_{\|}\mu_{\mathrm{B}}\vec{z}_i\cdot\vec{h})\,\sigma_i^z$$
$$+\frac{1}{2}\sum_{<i,j>} -\mathcal{J}^{\pm}\left(\sigma_i^+\sigma_j^- + \sigma_i^-\sigma_j^+\right)$$
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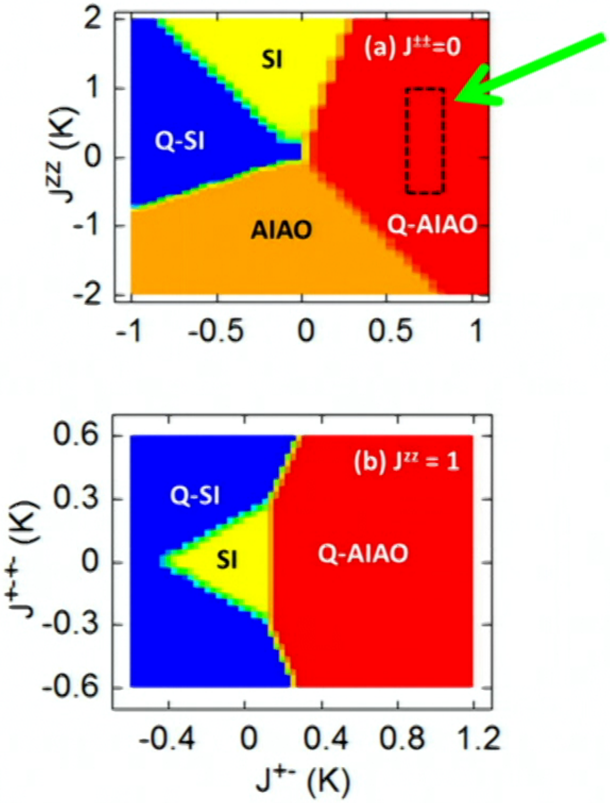
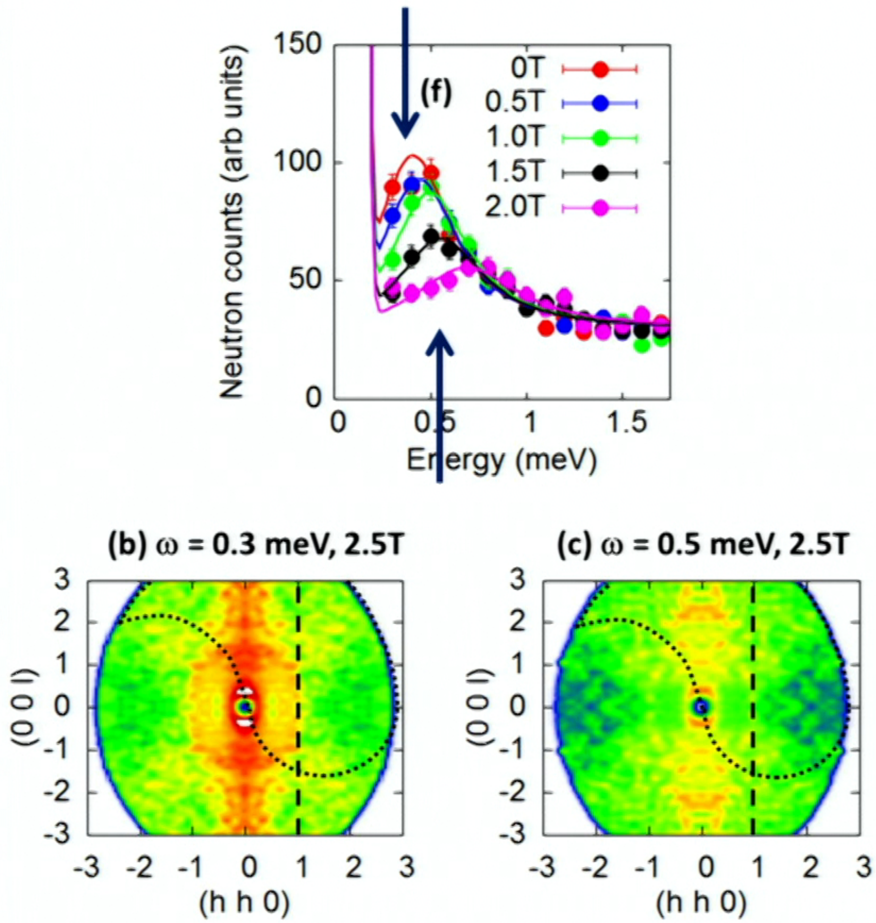
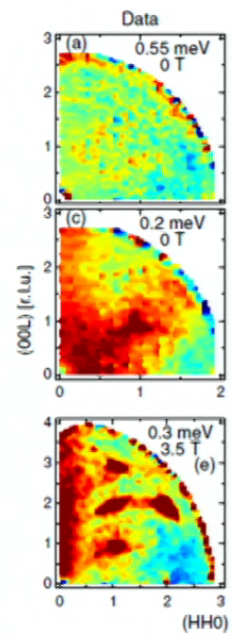


- Mean field, K=0 phases
- Pretty « similar » to the work by Lee et al although by far less sophisticated

Inelastic neutron scattering



PRL 118, 107206 (2017)



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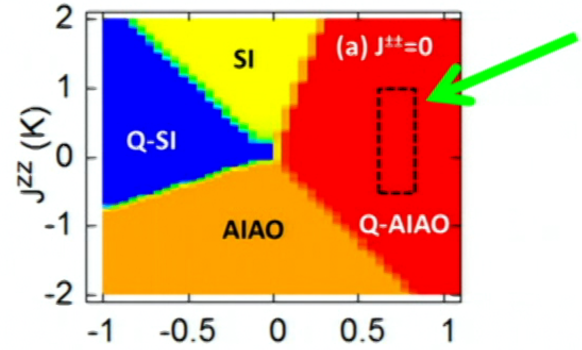
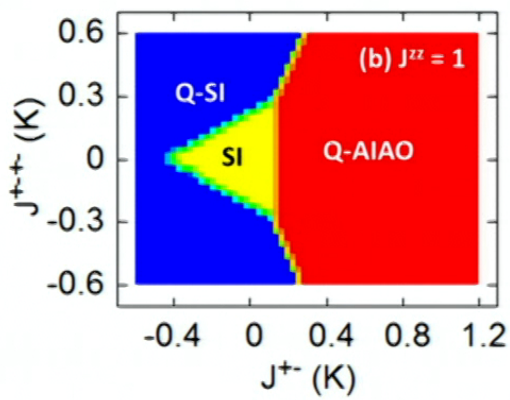
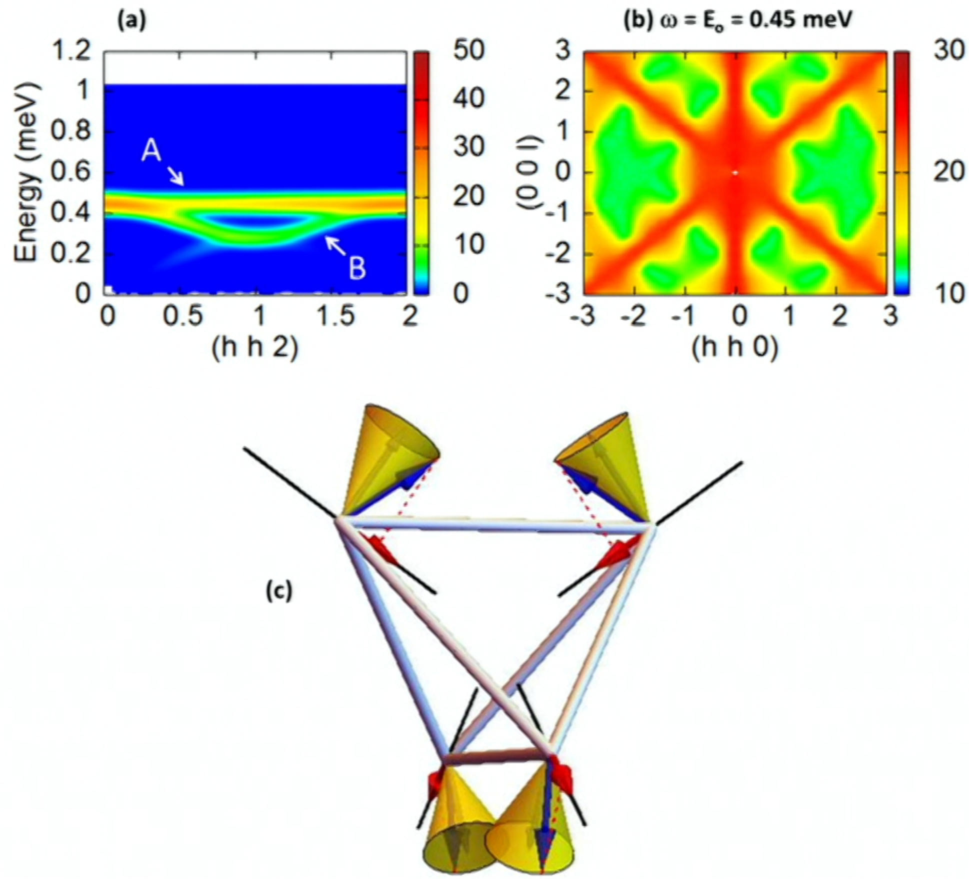


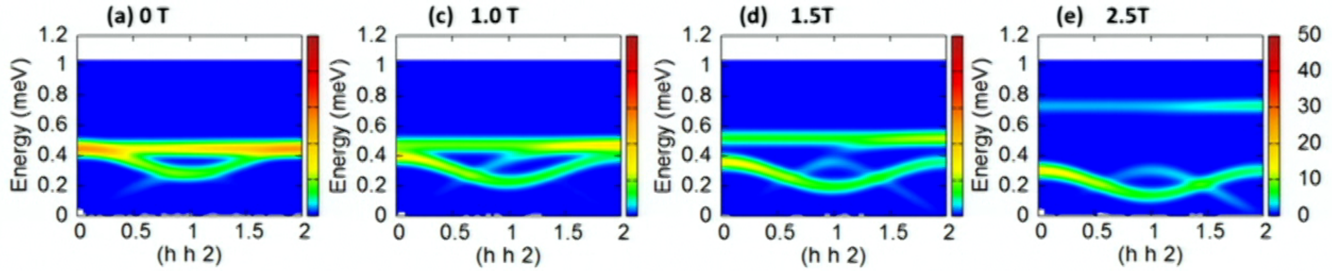


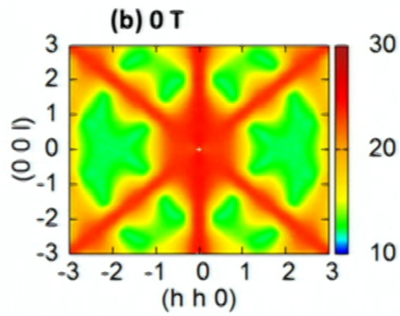
Spin and pseudo spin dynamics

- The RPA pseudo spin dynamics consists of
  - (A) a flat mode
  - (B) a dispersive feature
- (A) is characterized by the spin ice pattern
- The 2-in 2-out **magnetic** nature of excitation (A) arises from the peculiar projection of the corresponding « pseudo-spin » onto the z (magnetic) axis

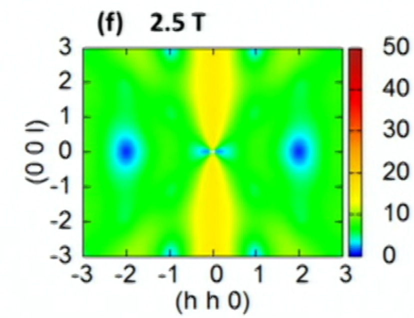


Spin and pseudo spin dynamics

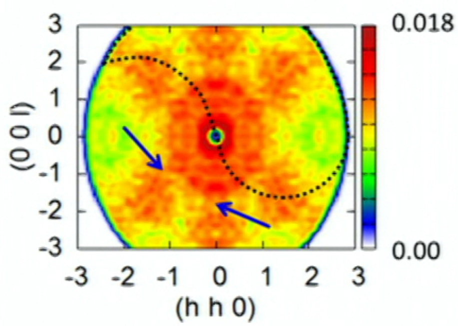


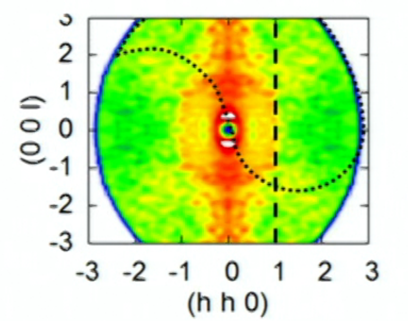


H // (1-10)

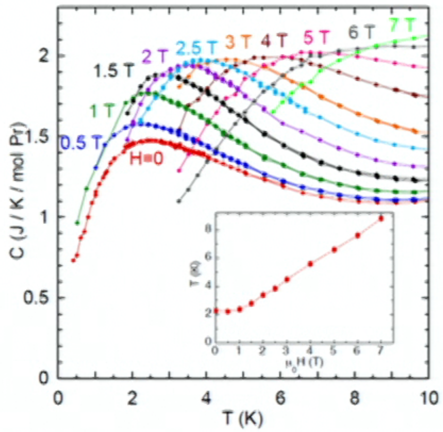


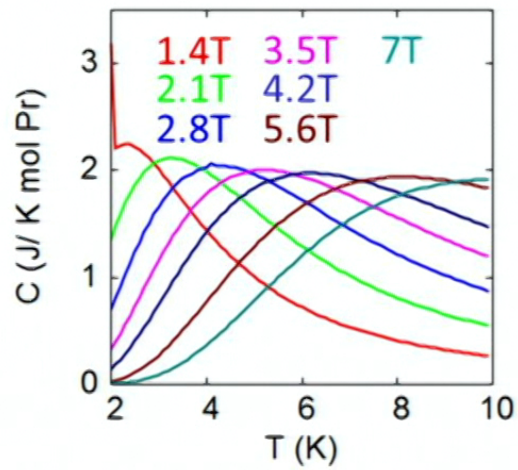
The low energy response is indeed a kind of « arm » along c*

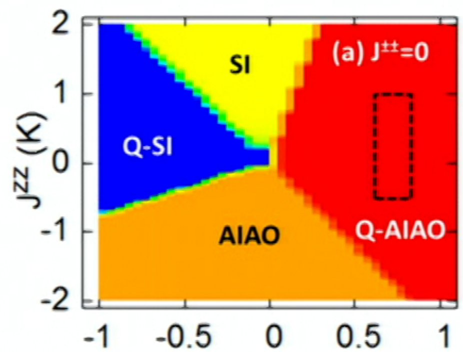




Specific heat







Based on the analysis of diffraction, INS and C_p, we propopse:

$$0.7 \leq \mathcal{J}^{\pm} \leq 0.8 \text{ K}$$
$$-0.5 \leq \mathcal{J}^{zz} \leq 1 \text{ K}$$

Discussion: input of MF

BUT

- *THE MEAN FIELD APPROACH IS CLEARLY NOT SATISFAYING SINCE IT PREDICTS LRO*
- *OTHER INGREDIENT ? → DISORDER, EXTREMELY IMPORTANT IN NK MAGNETS*

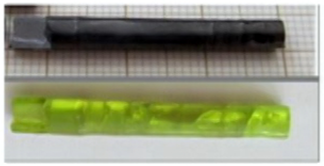
PHYSICAL REVIEW B **94**, 134428 (2016)

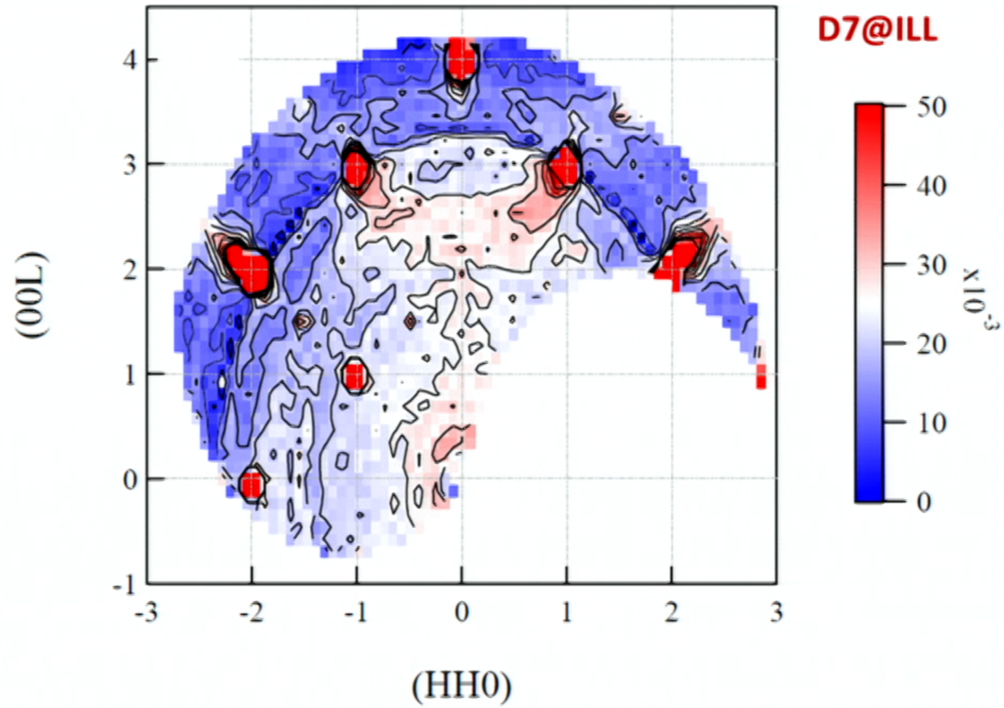
A. Local imperfections of the crystal field

In order to model local imperfections of the crystal field, we introduced a nontrigonal component of the crystal field (1) of the type $B_2^2 C_2^2$. Here, B_2^2 represents the magnitude of a strain coupled to one quadrupole operator (C_2^2). It would be more

See also
Duijn et al, *PRL* 94 177201 (2005)
Foronda, *PRL* 114, 017602 (2015)
Wen, *PRL* 118, 107206 (2017)
Savary, *PRL* 118, 087203 (2017)

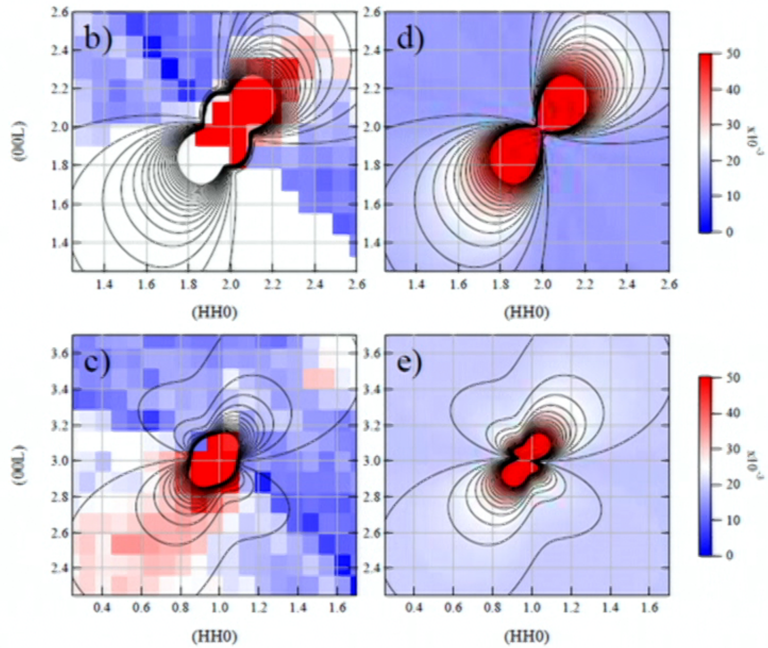
Diffuse scattering reveals disorder





- diffuse scattering measured by polarized neutron diffraction @D7 (ILL) → lattice origin
- Intrinsic defects in the material

Diffuse scattering reveals disorder



- Well represented by so called Huang scattering with lobes and « pinch points » !
- This would correspont to point defects creating a random strain described by :

$$g(\mathbf{e}) = \frac{3}{4\pi^2} \frac{\gamma}{(e^2+\gamma^2)^{5/2}},$$

- The parameter γ (reflects the strengh of disorder) ≈ 10 $^{-4 \text{ to } -5}$

Random strain and transverse « field »

By virtue of the magneto-elastic coupling, the random strain pertubs the electronic density:

In the pseudo spin ½ language this introduces a random transverse field v

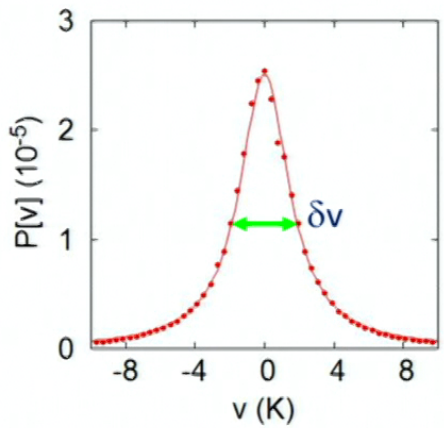
$$\mathcal{H}_{m-el} = e_1(\Gamma_3^1)[B_{21}^{xx}\mathbf{Q}_{xz} + B_{22}^{xx}\mathbf{Q}_{x^2-y^2}]$$
$$+ e_2(\Gamma_3^1)[-B_{21}^{xx}\mathbf{Q}_{yz} + B_{22}^{xx}\mathbf{Q}_{xy}]$$
$$+ e_1(\Gamma_3^2)[B_{21}^{zz}\mathbf{Q}_{xz} + B_{22}^{zx}\mathbf{Q}_{x^2-y^2}]$$
$$+ e_2(\Gamma_3^2)[-B_{21}^{zx}\mathbf{Q}_{yz} + B_{22}^{zx}\mathbf{Q}_{xy}],$$

$$g(\mathbf{e}) = \frac{3}{4\pi^2}\,\frac{\gamma}{(e^2+\gamma^2)^{5/2}},$$

$$\mathcal{H}_{\mathrm{m-el}} = \sum_i v_i\ \sigma_i^+ + v_i^*\ \sigma_i^-$$

$$\delta v \approx k\gamma$$

$\gamma \approx 10^{-5}$ to 10^{-4} → $\delta v \approx$ few K



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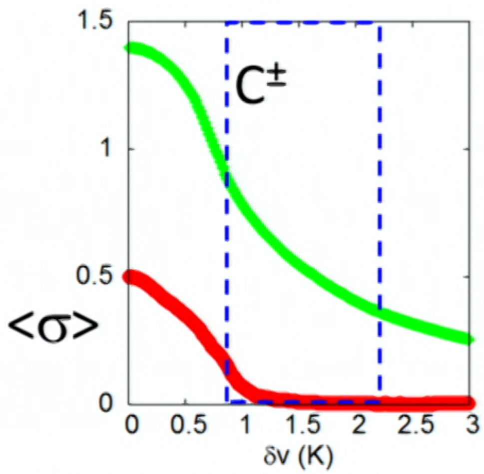
$$\mathcal{H}_{\mathrm{m-el}} = \sum_i v_i\ \sigma_i^+ + v_i^*\ \sigma_i^-$$

For the relevant value of $J^{\pm}$ and J^{zz},
δv progressively kills the AIAO LRO
yet NN SI and/or Q correlations persist



$$C^{zz} = \frac{1}{N}\sum_{i,\langle j\rangle_i} \langle\sigma_i^z\rangle\langle\sigma_j^z\rangle$$

$$C^{\pm} = \frac{1}{N}\sum_{i,\langle j\rangle_i} \langle\sigma_i^+\rangle\langle\sigma_j^-\rangle$$

Real space mean field calculation
(7x7x7 unit cells)

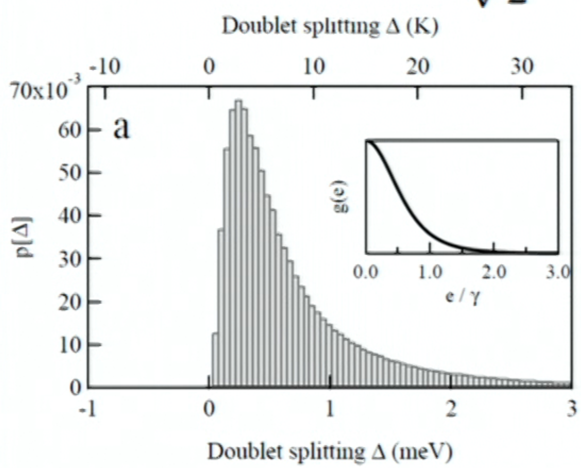
Input of MF

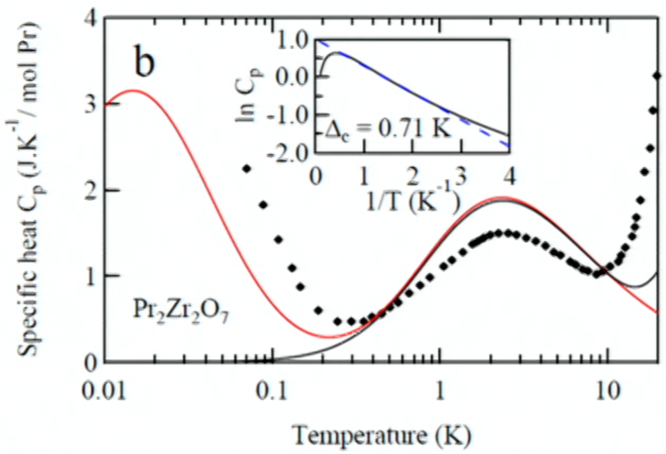
$|\uparrow\downarrow\rangle$ $|\delta v|$

$$|a\rangle \simeq \frac{1}{\sqrt{2}}\left[|\uparrow\rangle - |\downarrow\rangle\right]$$

$$|s\rangle \simeq \frac{1}{\sqrt{2}}\left[|\uparrow\rangle + |\downarrow\rangle\right]$$

Correlated by $J^{\pm}$ and J^{zz}





$\gamma \approx 1.25 \times 10^{-4}$ or $\delta v \approx 2K$

Distribution of Schottky peaks +
Hyperfine interaction at very low T

Input of MF

$$\mathcal{H} = \frac{1}{2}\sum_{<i,j>} \mathcal{J}^{zz}\sigma_i^z\sigma_j^z + \frac{1}{2}\sum_{<i,j>} -\mathcal{J}^{\pm}\left(\sigma_i^+\sigma_j^- + \sigma_i^-\sigma_j^+\right)$$

$$\mathcal{H}_{\mathrm{m-el}} = \sum_i v_i\ \sigma_i^+ + v_i^*\ \sigma_i^-$$

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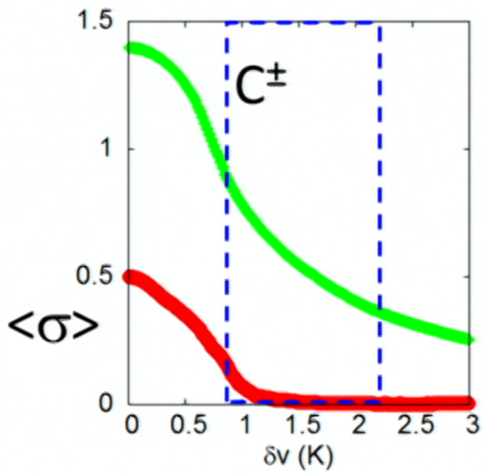
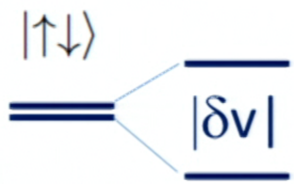


$$C^{zz} = \frac{1}{N}\sum_{i,\langle j\rangle_i} \langle\sigma_i^z\rangle\langle\sigma_j^z\rangle$$

$$C^{\pm} = \frac{1}{N}\sum_{i,\langle j\rangle_i} \langle\sigma_i^+\rangle\langle\sigma_j^-\rangle$$

Real space mean field calculation
(7x7x7 unit cells)

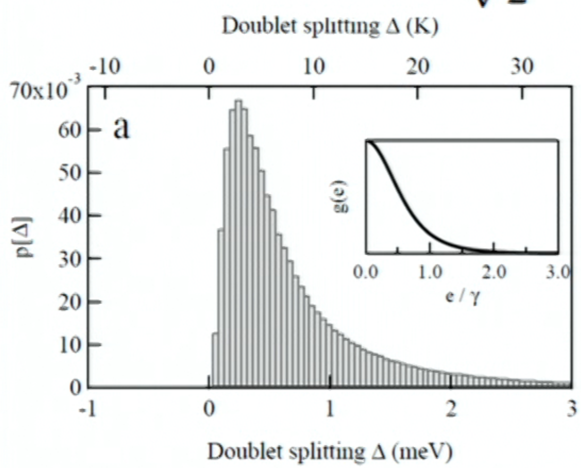
Input of MF

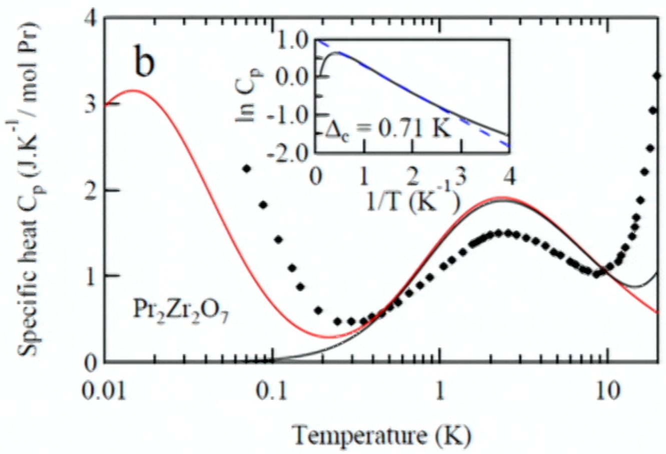


$$|a\rangle \simeq \frac{1}{\sqrt{2}}\left[|\uparrow\rangle - |\downarrow\rangle\right]$$

$$|s\rangle \simeq \frac{1}{\sqrt{2}}\left[|\uparrow\rangle + |\downarrow\rangle\right]$$

Correlated by $J^{\pm}$ and J^{zz}

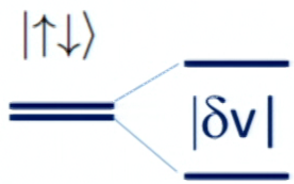




$\gamma \approx 1.25 \times 10^{-4}$ or $\delta v \approx 2K$

Distribution of Schottky peaks +
Hyperfine interaction at very low T

Input of MF



$$|a\rangle \simeq \frac{1}{\sqrt{2}} \left[|\uparrow\rangle - |\downarrow\rangle \right]$$

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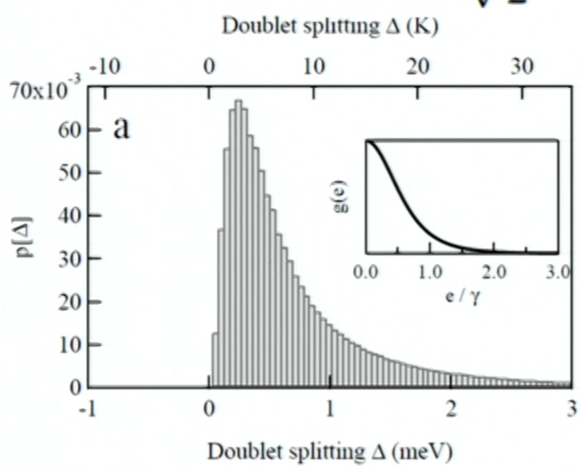
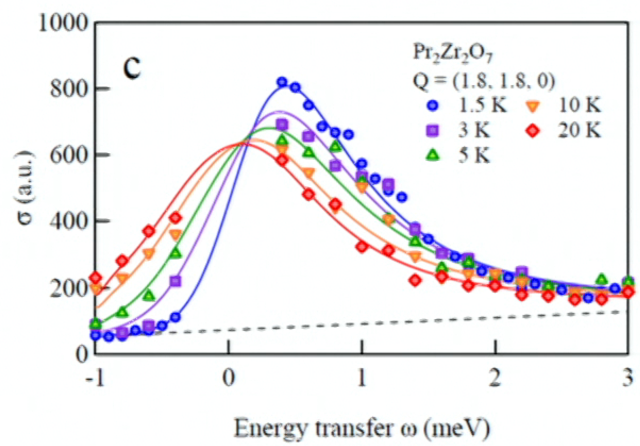
Correlated by $J^{\pm}$ and J^{zz}

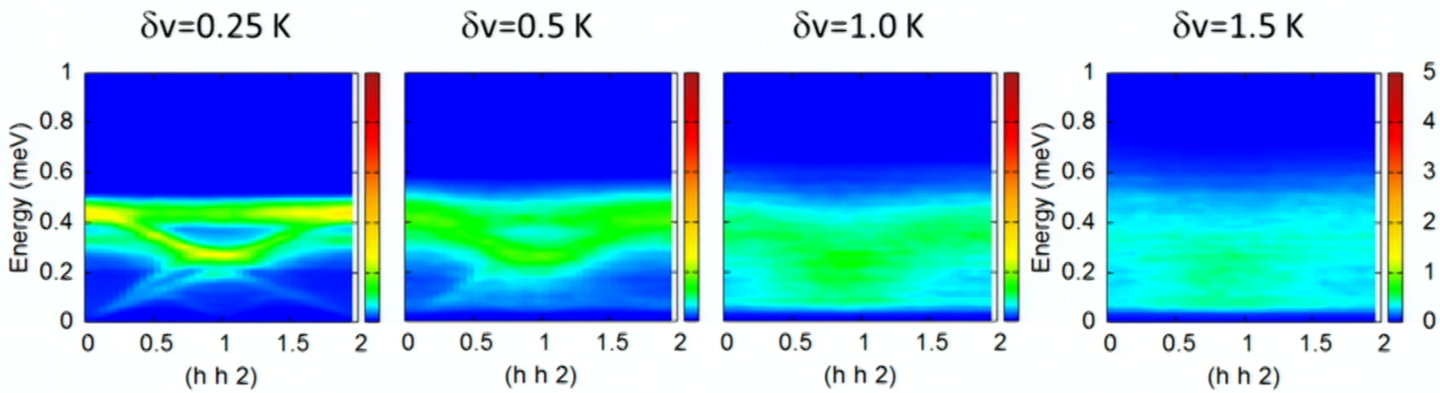


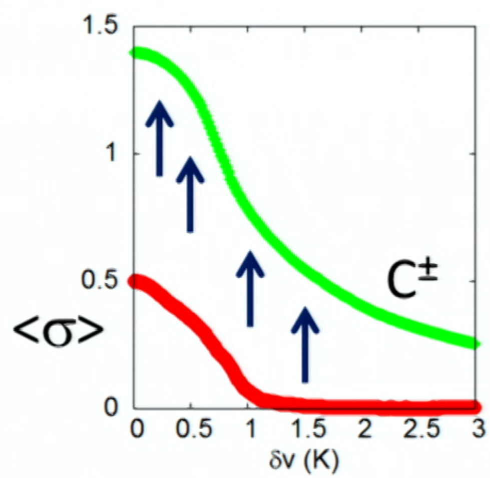


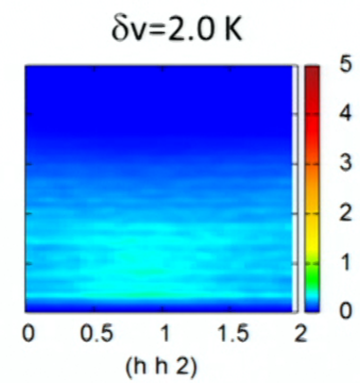
$\gamma \approx 1.25 \times 10^{-4}$ or $\delta v \approx 2K$

Magnetic transition within the doublets

Input of MF

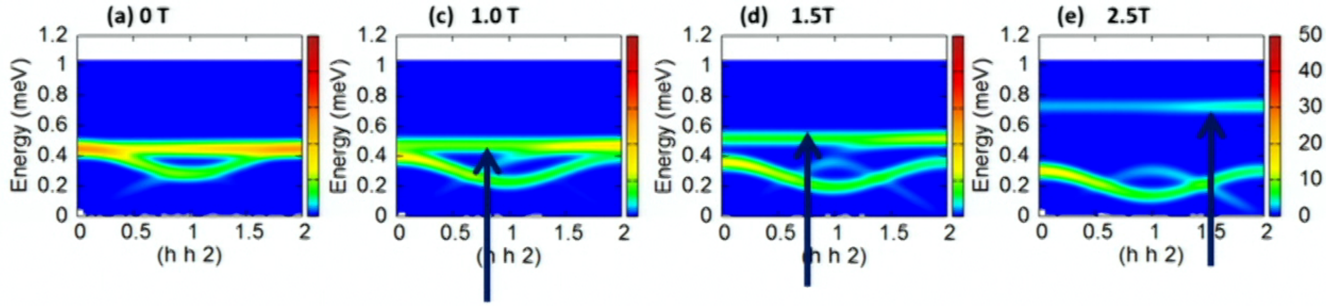




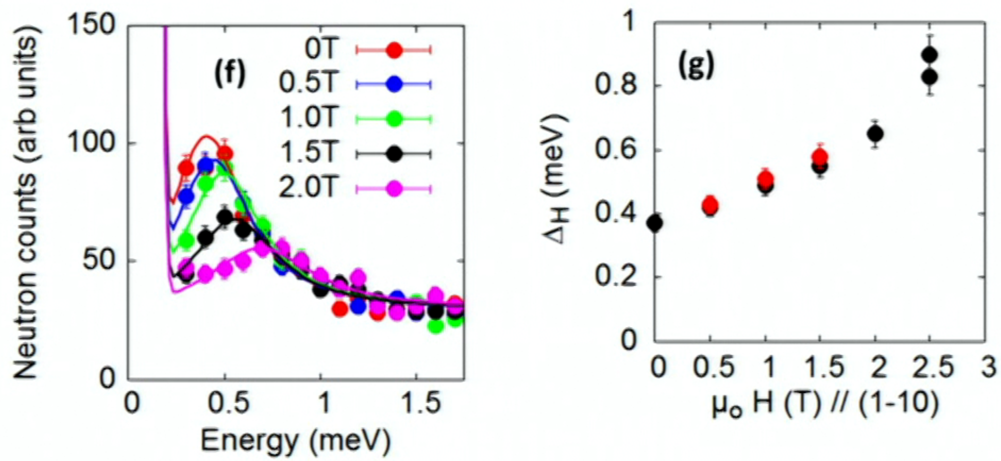


Classical spin dynamics calculation
(real space 7x7x7 unit cells)

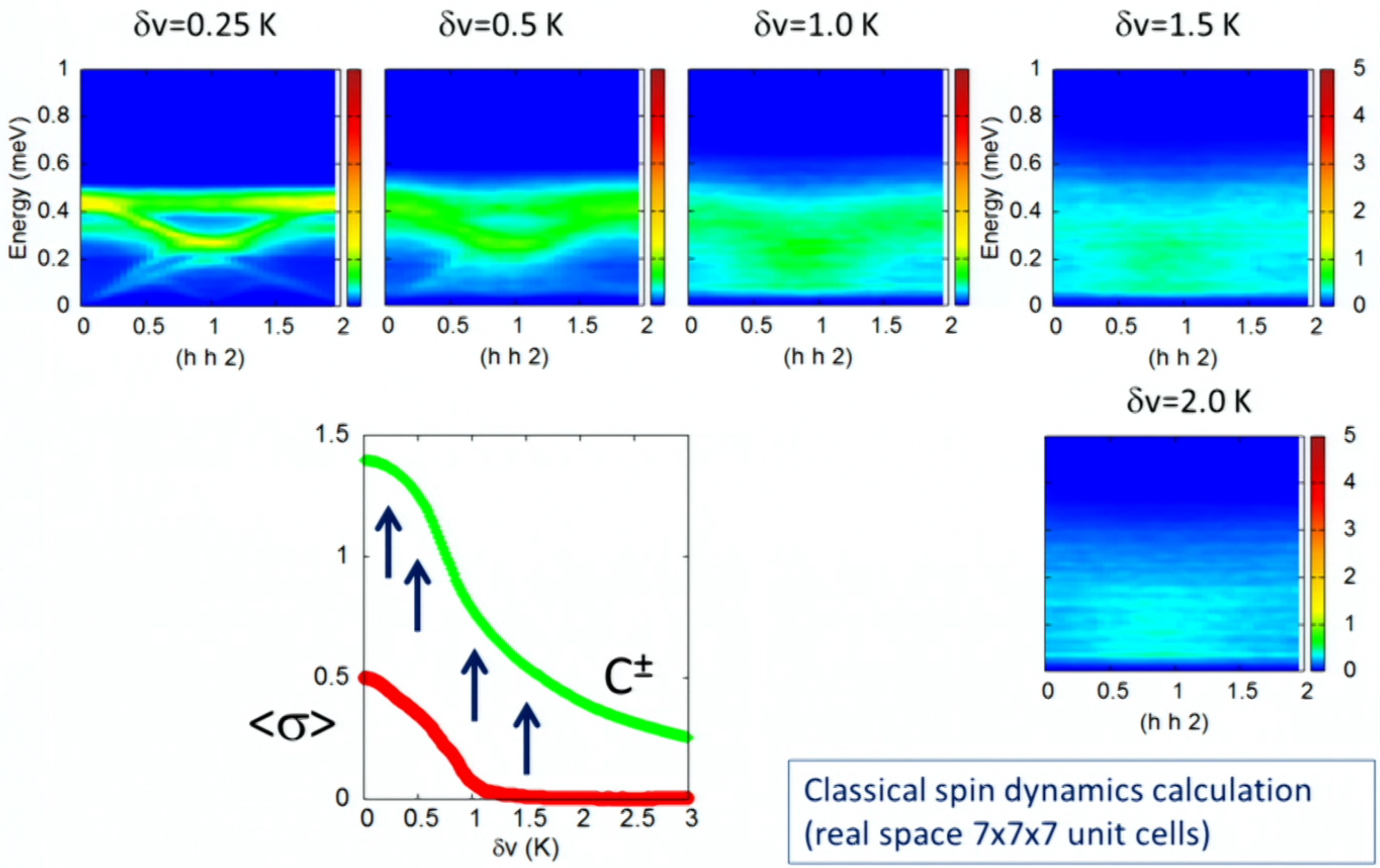
Spin and pseudo spin dynamics



Under applied field, a featureless flat mode detaches from the dynamical spin ice mode and shifts to higher energy



Input of MF



Classical spin dynamics calculation
(real space 7x7x7 unit cells)

Summary

Transverse interactions

- *AF QUADRUPOLAR INTERACTIONS RELEVANT IN $Pr_2Zr_2O_7$ → FINITE AND POSITIVE $J^{\pm}$*

- *→ INS (zero and applied field)*
- *→ Fiel evolution of Cp*

$\delta v \approx J^{\pm} \approx J^{zz}$

Role of disorder : stabilizes a disordered yet correlated state

- *DIRECT OBSERVATION OF DIFFUSE SCATTERING THAT DEMONSTRATE THE EXISTENCE OF INTRINSIC DEFECTS WHICH MODIFY THE 4f ELECTRONIC DENSITY*
- *→ No long order*
- *→ C_p (peak at 2K and hyperfine upturn)*
- *→ INS (broad blurred spin ice pattern)*

Special thanks to

ILL local contacts (IN5, D7): J. Ollivier, H. Mutka, A Wildes

CEA-CRG local contacts (D23) : E. Ressouche

Technical staff (cryogenics): X. Thonon, Ph. Boutrouille

Thank you all for your attention !