Title: Antiferroquadrupolar correlations in the quantum spin ice candidate Pr$_2$Zr$_2$O$_7$

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 Pr$_2$Zr$_2$O$_7$ by means of magnetization measurements, specific 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$_2$Zr$_2$O$_7$

S. Petit, N Martin,  
P. Bonville, S. Guitteny,  
I. Mirebeau

E. Lhotel, J. Robert

C. Decorse

M. Ciomaga-Hatnean  
G. Balakrishnan

LLB, CEA-CNRS Saclay, France

Institut Néel, Grenoble, France

ICMNO, Orsay Univ., France

Dpt of Physics, Warwick Univ, UK
Our point:

1) Role of transverse interactions

\[ H = \frac{1}{2} \sum_{\langle i,j \rangle} J_{ij}^{zz} \sigma_i^z \sigma_j^z + \sum_i (g_{\parallel} \mu_B z_i \cdot \vec{h}) \sigma_i^z \]

\[ + \frac{1}{2} \sum_{\langle i,j \rangle} -J_{ij}^\pm (\sigma_i^+ \sigma_j^- + \sigma_i^- \sigma_j^+) \]

\[ + \frac{1}{2} \sum_{\langle i,j \rangle} J_{ij}^{\pm \pm} (\gamma_{ij} \sigma_i^+ \sigma_j^+ + \gamma_{ij}^* \sigma_i^- \sigma_j^-) \]

2) Role of disorder: « Quantum SI route »

\[ H_{m\text{-el}} = \sum_i v_i \sigma_i^+ + v_i^* \sigma_i^- \]

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

In preparation
Spin Freezing in the Pyrochlore Antiferromagnet Pr$_2$Zr$_2$O$_7$

K. Matsuhira$^1$, C. Sekine$^2$, C. Paulsen$^3$, M. Wakeshima$^4$, Y. Hinatsu$^1$, T. Kitazawa$^5$, Y. Kinchi$^7$, Z. Hiroi$^5$, S. Takagi$^1$

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

Quantum fluctuations in spin-ice-like Pr$_2$Zr$_2$O$_7$

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$_2$Zr$_2$O$_7$

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

Antiferroquadrupolar correlations in the quantum spin ice candidate Pr$_2$Zr$_2$O$_7$

S. Petit,1,* E. Lhotel,1,† 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. Balakrishnan7

Structural and magnetic properties of single-crystals of the geometrically frustrated zirconium pyrochlore, Pr$_2$Zr$_2$O$_7$

M Ciomaga Hatnean,1, C Decorse,6, M R Lees,1, O A Petenko,1, D S Keeble1 and G Balakrishnan1
1Physics Department, University of Warwick, Coventry, CV4 7AL, UK
2SPSMS, CEMI, UMR 8582, Université Paris-Sud, 11, F-91405 Orsay, France
E-mail: M.Ciomaga.Hatnean@warwick.ac.uk
Received 24 March 2014
Accepted for publication 30 April 2014
Published 29 May 2014
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$_2$Zr$_2$O$_7$

J.-J. Wen,1,2,3 S. M. Koochpayeh,1 K. A. Ross,4,5 B. A. Trump,5 T. M. McQueen,1,6,7 K. Kimura,6,7 S. Nakatsui,6,7 Y. Qiu,6 D. M. Pajerowski,4 J. R. D. Copley,4 and C. L. Broholm6,7
Introduction

- A pyrochlore magnet
- \( \text{Pr}^{3+} \) is a NK ion
- Its CEF ground state is a doublet (spanning the spin \( \frac{3}{2} \) states) well protected from the 1st excited state (10 meV)
- No long range ordering down to 60 mK
- AF interactions, as inferred from \( \chi(T) \)
- Crystal growth is an issue, role of disorder outlined long ago
○ $C_p$ 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
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 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
H // (1 -1 0)

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

H // (1 1 1)

Smooth increase with H, consistent with M(H)
Inelastic neutron scattering

(a) $\omega = 0.3$ meV, 0T

- Under applied field $H//\langle 1-1-0 \rangle$, the peak of the lorenztian profile shifts to higher energy
- Confirm the dynamical spin ice pattern
Inelastic neutron scattering

(b) $\omega = 0.3$ meV, 2.5T
(c) $\omega = 0.5$ meV, 2.5T

PRL 118, 107206 (2017)
\[ H = \frac{1}{2} \sum_{i \neq j} J^{zz} \sigma_i^z \sigma_j^z + \sum_i \left( g_i \mu_B z_i \cdot \vec{h} \right) \sigma_i^z + \frac{1}{2} \sum_{i \neq j} \mathcal{J}^\pm \left( \sigma_i^+ \sigma_j^- + \sigma_i^- \sigma_j^+ \right) + \frac{1}{2} \sum_{i \neq j} \mathcal{J}^{\pm \pm} \left( \gamma_{ij} \sigma_i^+ \sigma_j^+ + \gamma_{ij}^* \sigma_i^- \sigma_j^- \right) \]

- Mean field, K=0 phases
- Pretty « similar » to the work by Lee et al although by far less sophisticated
\[ \mathcal{H} = \frac{1}{2} \sum_{<i,j>} J^{zz} \sigma_i^z \sigma_j^z + \sum_i (g_{\| \mu_B} \bar{z}_i \cdot \vec{h}) \sigma_i^z \]

\[ + \frac{1}{2} \sum_{<i,j>} -J^\pm (\sigma_i^+ \sigma_j^- + \sigma_i^- \sigma_j^+) \]

\[ + \frac{1}{2} \sum_{<i,j>} J^{\pm \pm} (\gamma_{ij} \sigma_i^+ \sigma_j^+ + \gamma_{ij}^* \sigma_i^- \sigma_j^-) \]

- Mean field, K=0 phases
- Pretty « similar » to the work by Lee et al although by far less sophisticated
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*
Based on the analysis of diffraction, INS and $C_p$, we propose:

$$0.7 \leq J^\pm \leq 0.8 \text{ K}$$
$$-0.5 \leq J^{zz} \leq 1 \text{ K}$$
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 \( B^3 \) of the type \( B^3 \). Here, \( B^3 \) represents the magnitude of a strain coupled to one quadrupole operator \( C^2 \). 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 correspond to point defects creating a random strain described by:

\[ g(e) = \frac{3}{4\pi^2} \frac{\gamma}{(e^2 + \gamma^2)^{5/2}}. \]

- The parameter \( \gamma \) (reflects the strength of disorder) \( \approx 10^{-4} \text{ to } 10^{-5} \)
Random strain and transverse « field »

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

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

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

In the pseudo spin ½ language this introduces a random transverse field \( \mathcal{v} \)

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

\[
\delta v \approx k \gamma
\]

\[
\gamma = 10^{-5} \text{ to } 10^{-4} \rightarrow \delta v \approx \text{few K}
\]

\[
P(v) (10^{-5})
\]

\[
-v (K)
\]
\begin{equation*}
\mathcal{H} = \frac{1}{2} \sum_{<i,j>} J^{zz} \sigma_i^z \sigma_j^z + \frac{1}{2} \sum_{<i,j>} -J^\pm \left( \sigma_i^+ \sigma_j^- + \sigma_i^- \sigma_j^+ \right)
\end{equation*}

\begin{equation*}
\mathcal{H}_{m-el} = \sum_i v_i \sigma_i^+ + v_i^* \sigma_i^-
\end{equation*}

For the relevant value of $J^\pm$ and $J^{zz}$, 
\( \delta v \) progressively kills the AIAO LRO 
but NN SI and/or Q correlations persist.

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

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

Real space mean field calculation 
(7x7x7 unit cells)
Input of MF

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

\[ |\delta v| = \frac{1}{\sqrt{2}} \left[ |\uparrow\rangle + |\downarrow\rangle \right] \]

Correlated by \( J^\pm \) and \( J^{zz} \)

Doublet splitting \( \Delta \) (K)

\[ \gamma \approx 1.25 \times 10^{-4} \quad \text{or} \quad \delta v \approx 2K \]

Distribution of Schottky peaks + Hyperfine interaction at very low T
\[
\mathcal{H} = \frac{1}{2} \sum_{<i,j>} J^{zz} \sigma_i^z \sigma_j^z + \frac{1}{2} \sum_{<i,j>} -J^\pm (\sigma_i^+ \sigma_j^- + \sigma_i^- \sigma_j^+) \\
\mathcal{H}_{m-el} = \sum_i v_i \sigma_i^+ + v_i^* \sigma_i^-
\]

For the relevant value of \( J^\pm \) and \( J^{zz} \),
\( \delta v \) 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

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

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

Correlated by \(J^z\) and \(J^{zz}\)

Doublet splitting \(\Delta\) (K)

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

Distribution of Schottky peaks + Hyperfine interaction at very low T
Input of MF

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

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

Correlated by \( J^\pm \) and \( J^{zz} \)

Doublet splitting \( \Delta \) (K)

\[ p(\Delta) \]

\[ |e\rangle \]

\[ e/\gamma \]

\[ 0.0 \quad 1.0 \quad 2.0 \quad 3.0 \]

\[ 0 \quad 10 \quad 20 \quad 30 \]

\[ -1 \quad 0 \quad 1 \quad 2 \quad 3 \]

\[ \gamma \approx 1.25 \times 10^{-4} \quad \text{or} \quad \delta v \approx 2K \]

Magnetic transition within the doublets

\[(\text{Pr}_2\text{Zr}_2\text{O}_7)\]

\[ Q = (1.8, 1.8, 0) \]

\[ \bullet \ 1.5 \text{ K} \quad \bullet \ 10 \text{ K} \]

\[ \bullet \ 3 \text{ K} \quad \bullet \ 5 \text{ K} \]

\[ \bullet \ 10 \text{ K} \]

[Energy transfer \( \omega \) (meV)]
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

$\delta v = 0.25\,\text{K}$

$\delta v = 0.5\,\text{K}$

$\delta v = 1.0\,\text{K}$

$\delta v = 1.5\,\text{K}$

$\delta v = 2.0\,\text{K}$

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

- **AF QUADRUPOLAR INTERACTIONS RELEVANT IN** $Pr_2Zr_2O_7 \rightarrow$ **FINITE AND POSITIVE** $J^\pm$
- $\rightarrow$ **INS (zero and applied field)**
- $\rightarrow$ **Fiel evolution of C_p**

**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**
- $\rightarrow$ **No long order**
- $\rightarrow$ **$C_p$ (peak at 2K and hyperfine upturn)**
- $\rightarrow$ **INS (broad blurred spin ice pattern)**

$\delta V \approx J^\pm \approx J^{zz}$
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!