Quantum Fluctuations and Criticality in Pr based Spin Ice Systems

Satoru Nakatsuji

Institute for Solid State Physics, Univ. of Tokyo

Collaborators

Institute for Solid State Physics, University of Tokyo

Kenta Kimura
Jun Ishikawa
Yo Machida (Tokyo IT)
Yasuo Ohta

Toshiro Sakakibara
Takashi Tayama
Yoshiya Uwatoko

RIKEN
S. Onoda

Nagoya Univ.
E. Nishibori
H. Sawa

Univ. of Ryukyus
Y. Karaki

JHU/NIST, U.S.A.
Collin Broholm
Jiajia Wen
Chris Stock

Gottingen University
Y. Tokiwa
P. Gengenwart

National High Magnetic Field Lab. Tallahassee
Luis Balicas

University of California, Riverside
Douglas MacLaughlin

Acknowledgement to Leon Balents, Yong Baek Kim

Content

- Introduction
  - Spin Ice on Pyrochlore
  - Coulomb Phase and Monopoles
  - Quantum version of spin ice

- Quantum Fluctuation Effects in Pr based Spin Ices
  - Quantum monopolar fluctuations in $Pr_2Zr_2O_7$

  - Interplay between spin ice and conduction electron,
    Chiral Spin Liquid Behavior and Quantum Criticality in $Pr_2Ir_2O_7$

Spin Ice : Pyrochlore Magnet

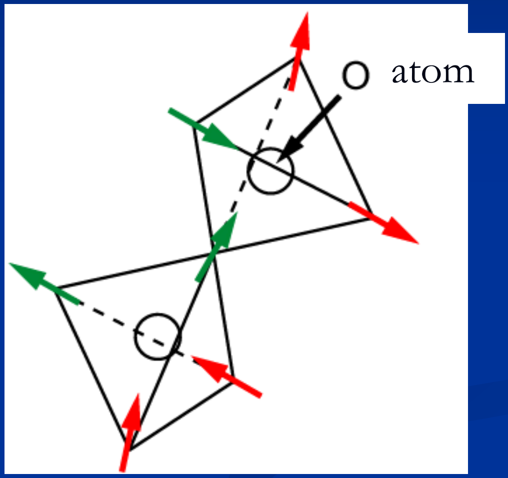
Nearest-neighbor FM coupled <111> Ising Spins

Classical Spins

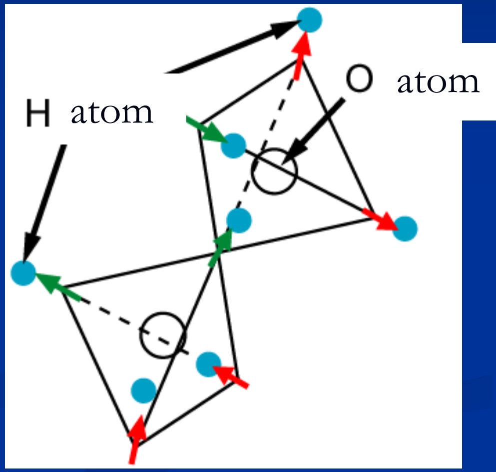
No Magnetic Order at $T > 0$

"Spin Ice"

M.J. Harris, S. Bramwell *et al*., (1997)



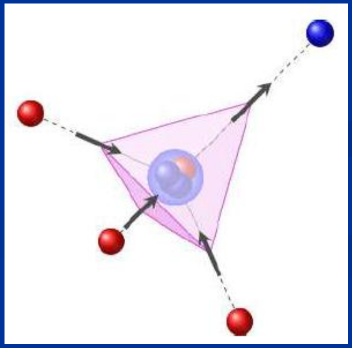
"2-in, 2-out"
${}_4C_2 = 6$ fold



"H_2O ice"

Disordered State w/ Residual Entropy
$S_0 \sim R/2\ln(3/2)$

A. Ramirez *et al*., (1997)



Low lying Defect
"3-in 1-out" state
Monopole
$\Delta = 2J_{ff}$

Castelnovo *et al*. (2008).

Coulomb Phase, Pinch point

Coulomb Phase: quantum dimer system, Heisenberg AF on pyrochlore,…

In Spin Ice, 2-in 2-out can be considered as a divergence free condition:

$$\vec{\nabla} \bullet \vec{M} = 0$$

It leads to power law spin-spin correlation~ $\vec{\nabla}_{r'} \vec{\nabla}_{r} \frac{1}{r - r'}$

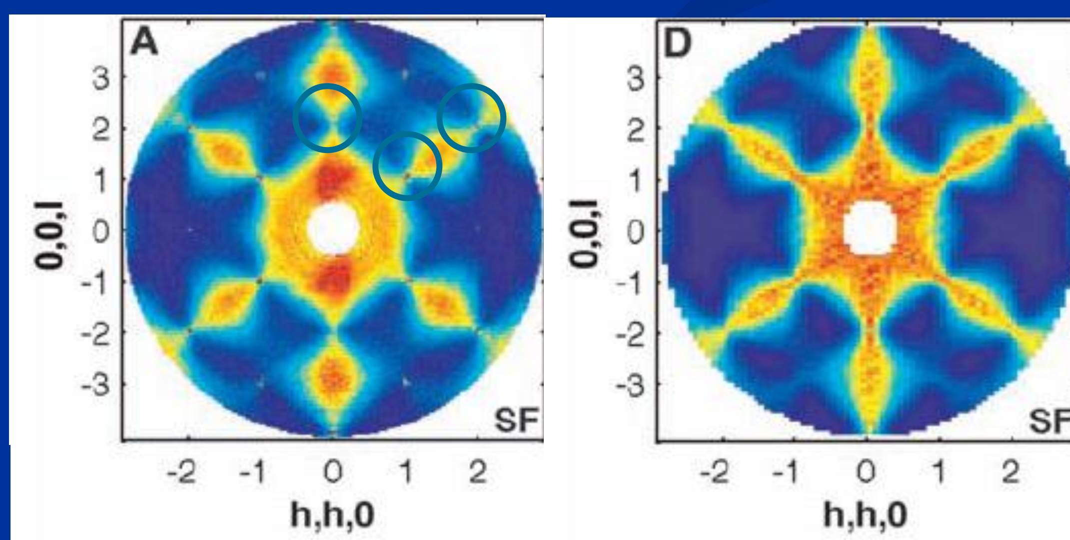
Result in 'pinch point' singularity in scattering experiment.

Ice rule breaking states correspond to creation of magnetic monopole/ width.

Spin Ice $Ho_2Ti_2O_7$, Neutron Scattering Fennell, *et al* (2009)

Experiment

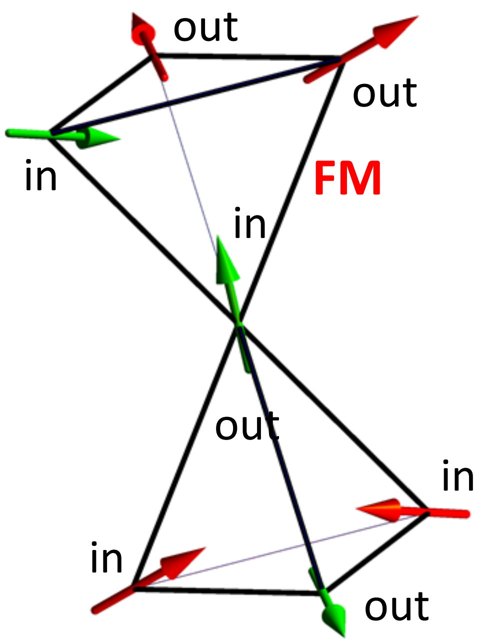
Pinch Points



Monte Carlo simulation

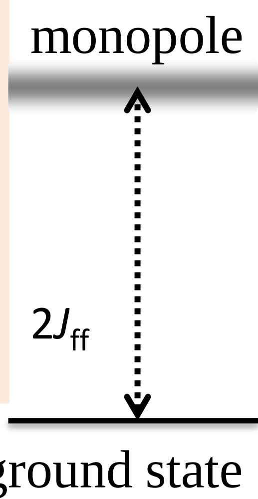
"*Classical*" dipolar spin ice system

[1] ex. Bramwell & Gingras, Science (2001).



Dipolar spin ice on pyrochlore lattice[1]

- Dipolar interaction, Classical → FM coupling 1 K
  ex. $Dy_2Ti_2O_7$ (Dy: large moment $10\mu_B$)
- No quantum fluctuations in macroscopically degenerate spin ice manifold
- Monopole dynamics is only diffusive.

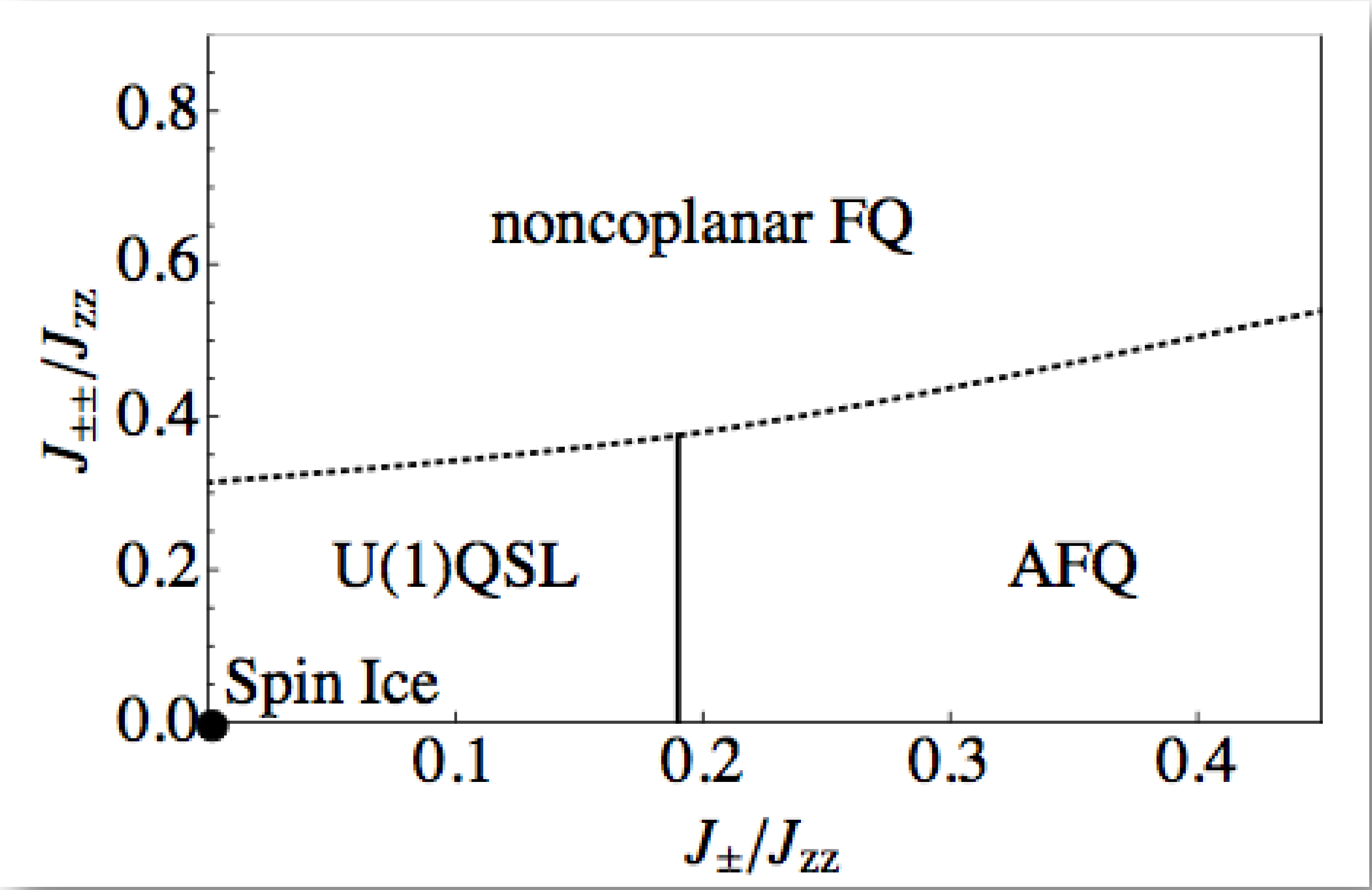


Effects of Quantum fluctuations?

Novel quantum spin liquids and/or Coherent motion of monopoles?

Quantum Spin Ice?

Theory: M. Hermele *et al*., (2004), R. Moessner *et al*., (2003), A. Banerjee *et al*., (2008), S. Onoda *et al*., (2010).



Content

- Introduction
  - Spin Ice on Pyrochlore
  - Coulomb Phase and Monopoles
  - Quantum version of spin ice

- Quantum Fluctuation Effects in Pr based Spin Ices
  - Quantum monopolar fluctuations in $Pr_2Zr_2O_7$

  - Interplay between spin ice and conduction electron, Chiral Spin Liquid Behavior and Quantum Criticality in $Pr_2Ir_2O_7$

$Pr_2Zr_2O_7$

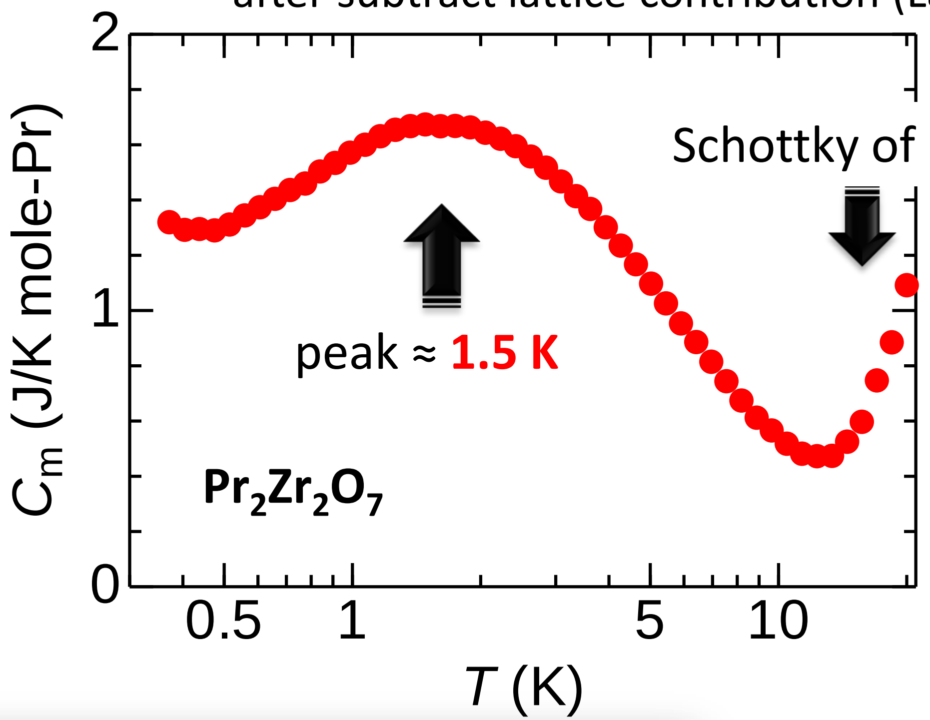
Quantum monopolar fluctuations

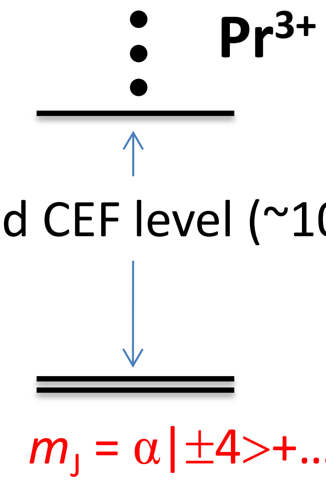
Specific heat

estimate of energy scale → ≈1 K

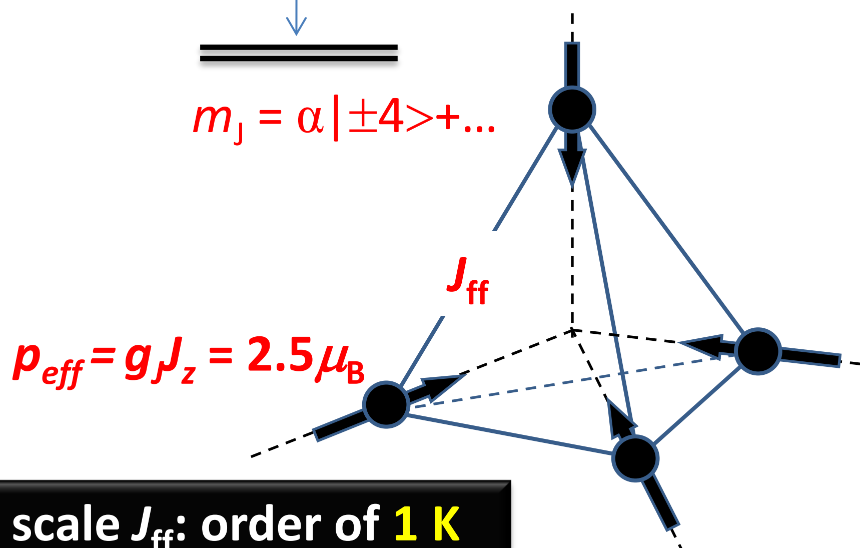
Magnetic specific heat C_m

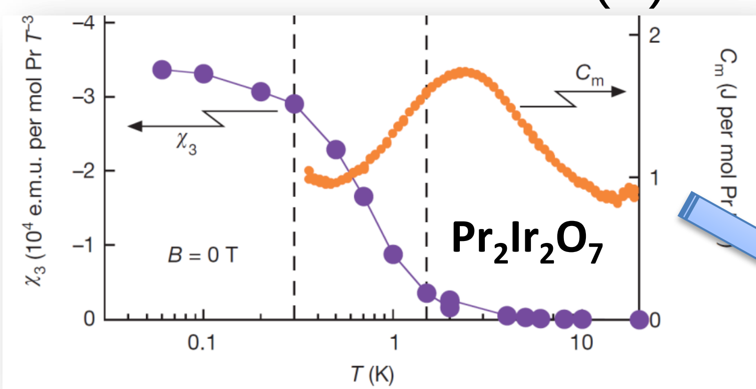
after subtract lattice contribution ($La_2Zr_2O_7$)





CEF confirmed by Inelastic NS.





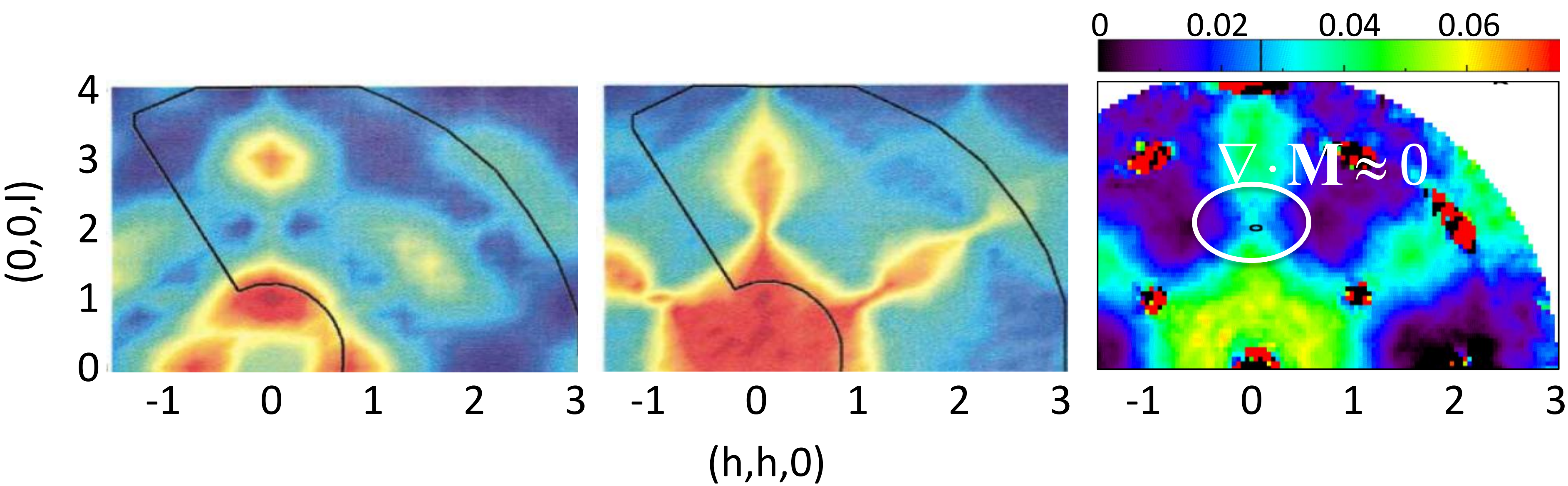
❑ **energy scale J_{ff}: order of 1 K**

◯ exchange coupling ✕ dipolar (0.1 K)

similar to $Pr_2Ir_2O_7$ case

→ common feature in Pr-based pyrochlore

Elastic neutron scattering for $Pr_2Zr_2O_7$



Dipolar spin ice | NN spin ice | $Pr_2Zr_2O_7$

Monte Carlo Simulation | $T = 0.1$ K

S.T. Bramwell et al. PRL (2001). | J. Wen, C. Broholm @ JHU

Measurements were performed at MACS, NCNR, NIST, USA
Background was subtracted using the data taken at $T = 15$ K

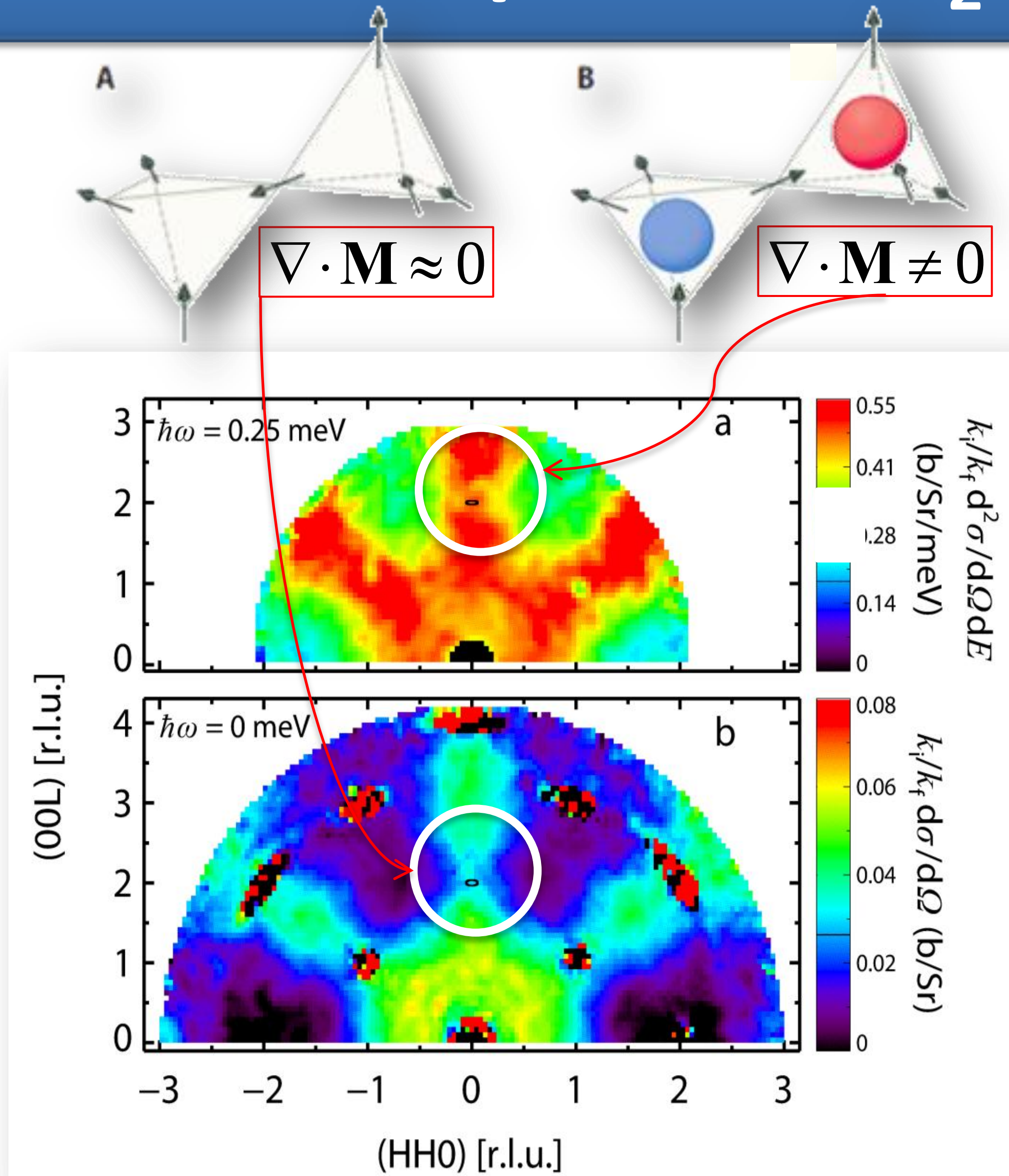
Pinch points at [200] , [111],… ⟹ Coulomb Phase with NN FM coupling

Only 5 % of the total spectra weight is in the elastic channel: dynamic spin ice manifold

Quantum Monopoles in $Pr_2Zr_2O_7$

J. Wen,
C. Broholm
@ JHU

T = 0.1 K

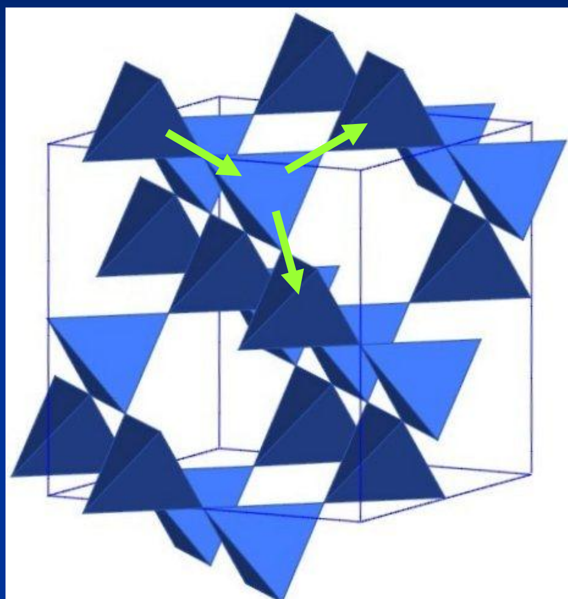


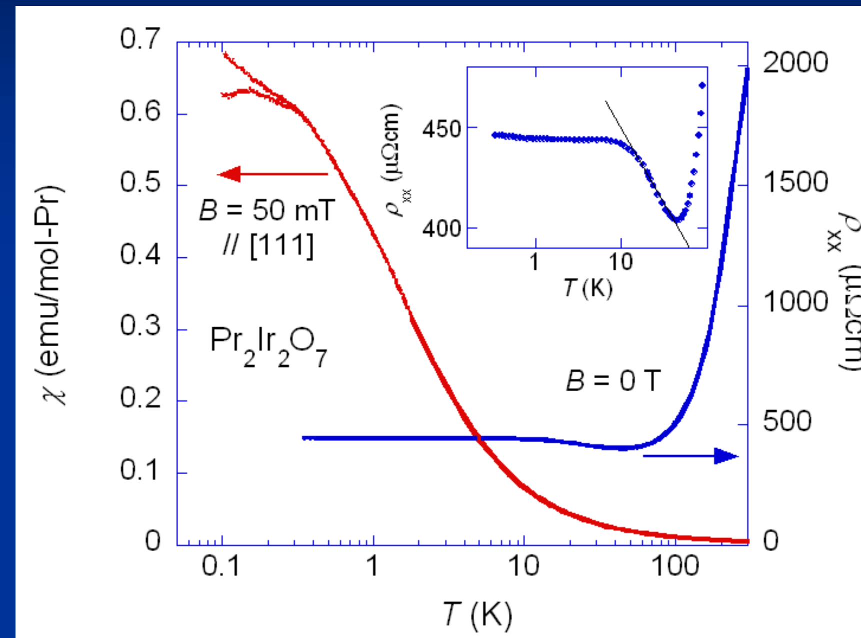
Pinch points are filled in for the inelastic map. ➔ Quantum dynamics of monopoles

$Pr_2Ir_2O_7$

Spin Ice + Exotic Liquids =?

$Pr_2Ir_2O_7$:
Geometrically Frustrated Kondo Lattice





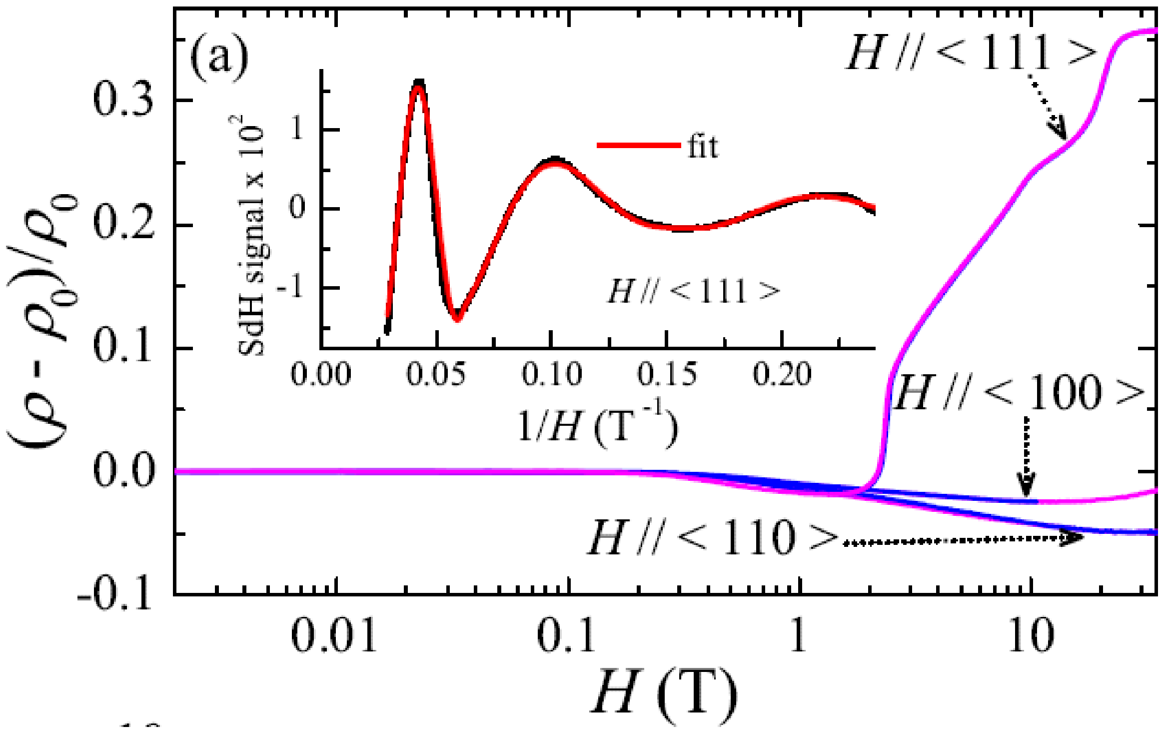
Pr^{3+}: $4f^2$ Localized **Ising** Moment
// **<111>** (CEF by Neutron Diffraction)

Ir^{4+} : $5d^5$ **conduction electron**,
Pauli Paramagnetism
Spin-Orbit Coupling Effect?

Strong Frustration:
No Long Range Order
T_f **~ 0.3 K<<** θ_W **~ –20 K**
No freezing at > 20 mK by μSR
(D. MacLaughlin et al.)

S. Nakatsuji, Yo Machida, Y. Maeno, T. Tayama, T. Sakakibara *et al.* PRL **96** (2006).

Magnetoresistance



T = 30 mK

- <100>, <110> :
  Smooth decrease

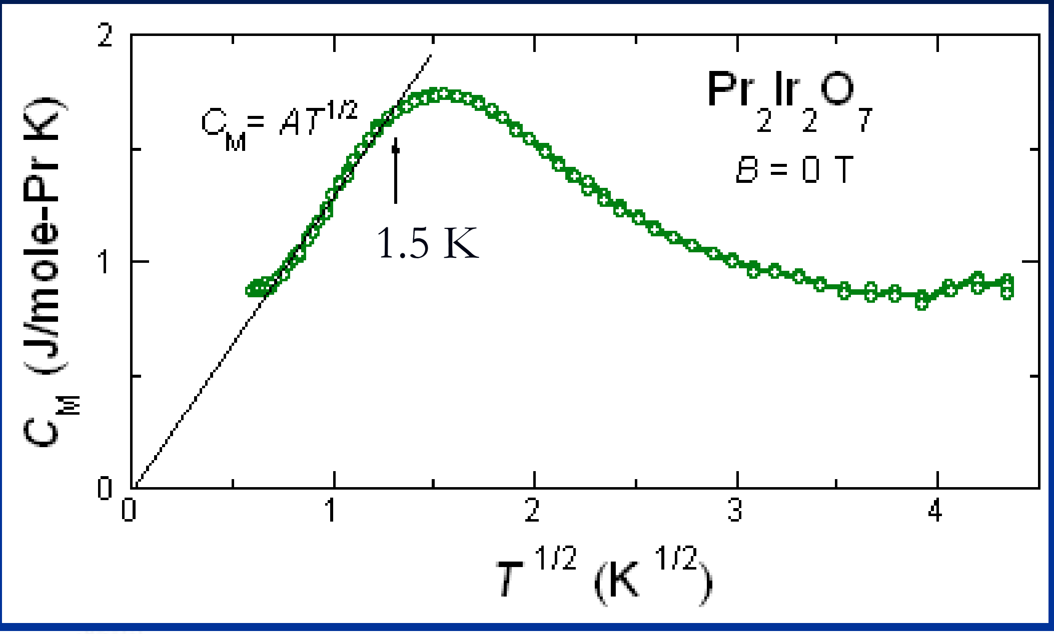
- <111> :
  Jump in resistance
  Metamagnetism

SdH Oscillation seen for [111] fit to Lifshitz-Kosevich formula at $B > B_c$: Loss of scattering due to disordred Spin Ice State

L **~ 500 – 800 Å : coherence due to uniform [1-in,3-out] state**

L. Balicas, S. Nakatsuji, Y. Machida, and S. Onoda, Phys. Rev. Lett. **106**, 217204 (2011).

FM correlation below $T \sim 2J_{ff} \sim 1.5$ K



Monopole Creation

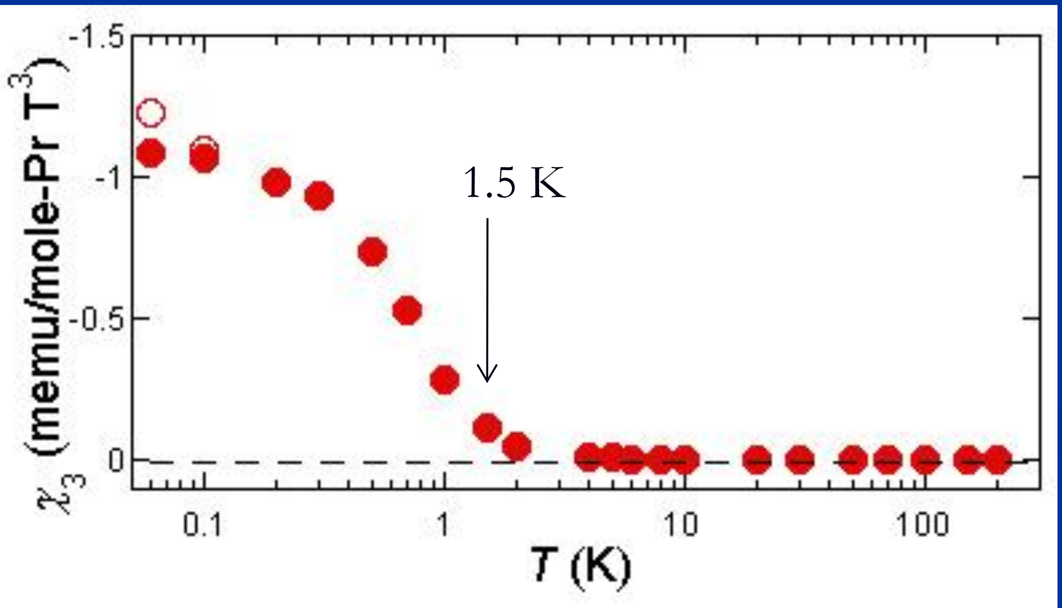
$\Delta = 2J_{ff}$

Broad peak at ~ 1.5 K

$T < 1.5$ K $\quad C_M = AT^{1/2}$

$S_M = AT^{1/2}$

Highly degenerate state



$\chi_3(q=0)$: a steep negative increase, and saturate to a large negative value

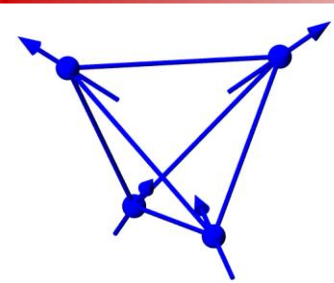
⇨ **FM correlation between Ising moments**

⇔ ~~**Quadrupoler Order**~~

$Pr_2Ir_2O_7$: Geometrically Frustrated Kondo Lattice

T_f~0.3 K | $2J_{ff}$~1.4K | $|\theta_W|$ ~ 20 K | T (K)

Partial Spin Freezing

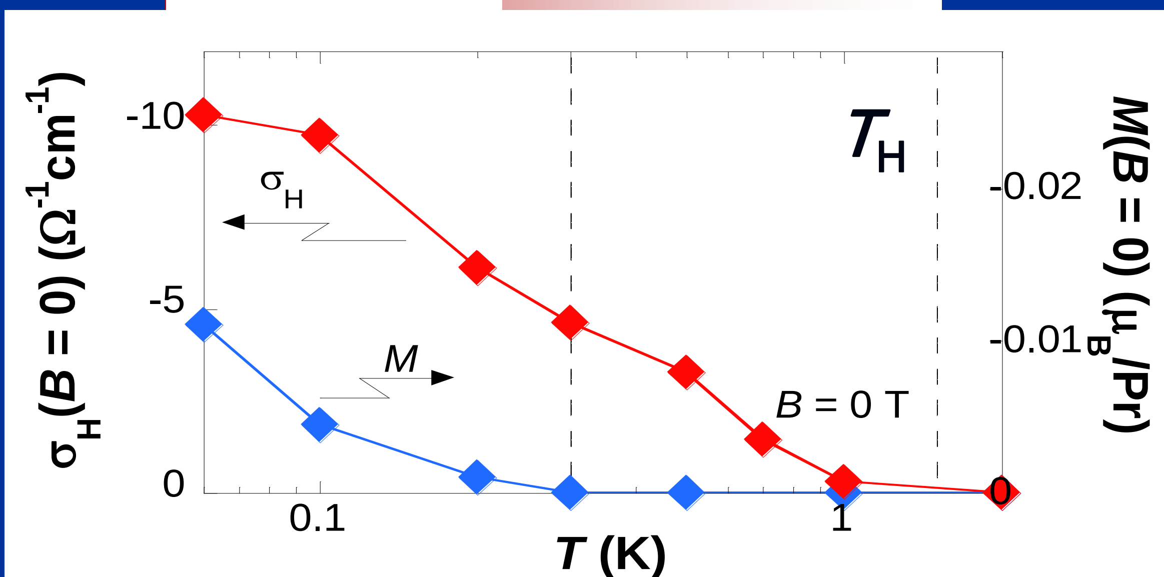


"2-in 2-out" configurations

Paramagnetic Spin Liquid Phase

Kondo like Behavior

Dipolar Spin Ice (eg. $Dy_2Ti_2O_7$)
Spin freezing at $T < J_{ff}$

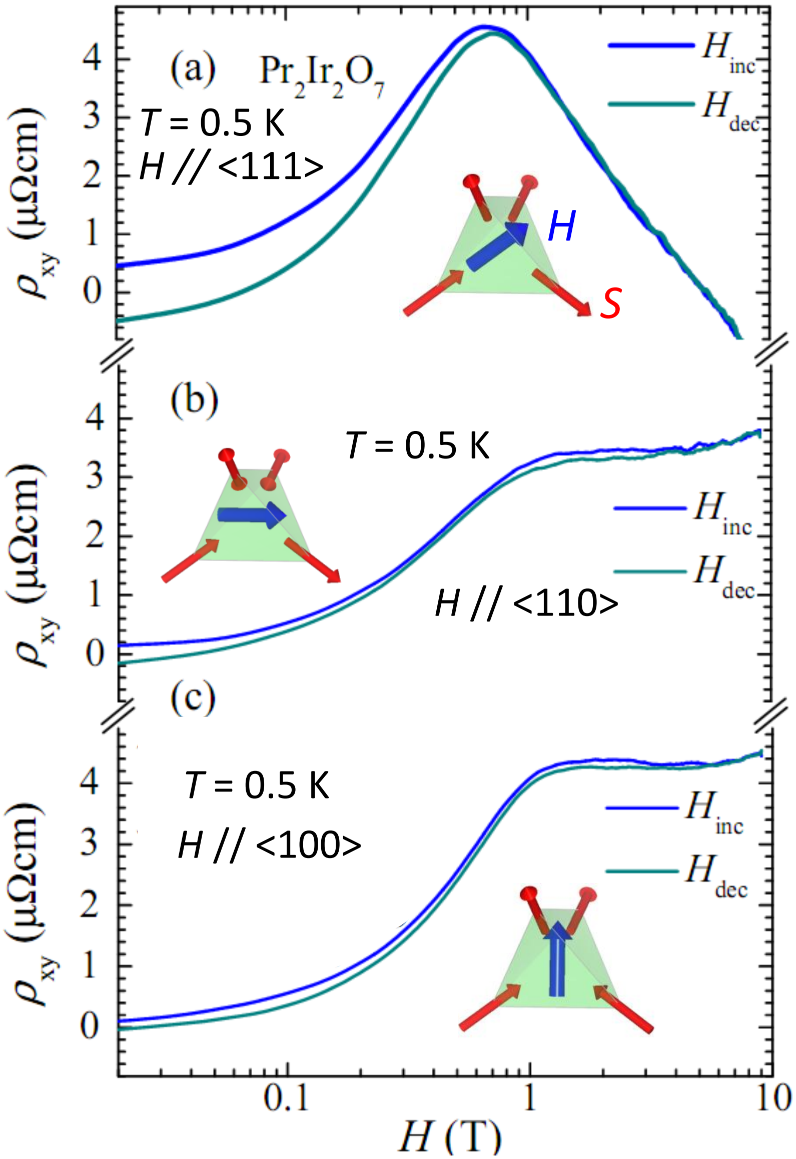


Onset of Spontaneous HE at $T_H \sim J_{ff}$

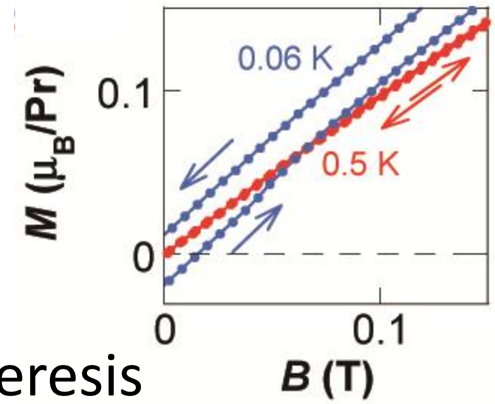
TRS broken Spin Liquid Phase

Yo Machida, S. Nakatsuji, S. Onoda, T. Tayama, and S. Sakakibara, Nature **463**, 210 (2010).

AHE without Magnetic Order



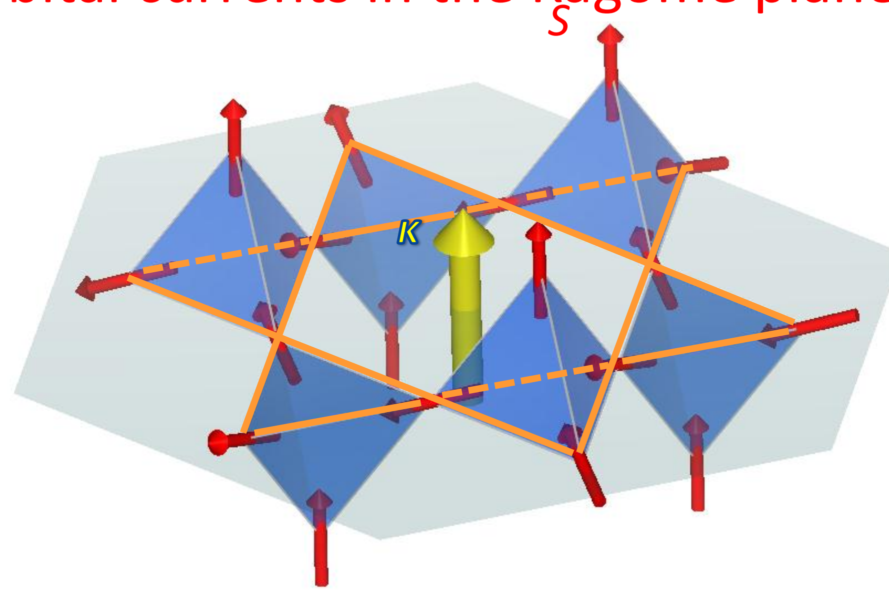
T = 0.5 K > T_f(~0.3 K)



<111> : Large hysteresis

<100>, <110> : Small hysteresis

Orbital currents in the Kagome plane



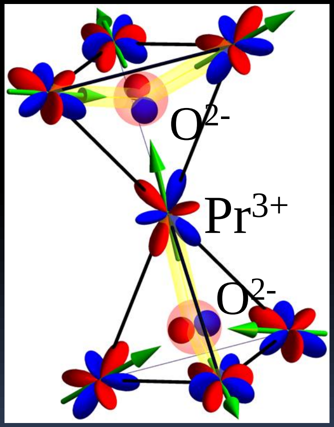
L. Balicas, S. Nakatsuji, Y. Machida, and S. Onoda, Phys. Rev. Lett. **106**, 217204 (2011).

Summary:
Quantum effects in Pr based Spin Ice

- Spin Liquid with Spin Ice Correlation

  at $T < 1.5$ K ~ $2J$: Monopole creation scale

- **$Pr_2Zr_2O_7$**
  - Quantum Monopolar Fluctuations



- **$Pr_2Ir_2O_7$**
  - **Chiral Spin Liquid Behavior and Quantum Criticality**
  - **Role of Spin Orbit Coupling in the Ir Pyrochlore Network?**