INSTITUTE FOR QUANTUM MATTER

A collaboration between JOHNS HOPKINS UNIVERSITY and PRINCETON UNIVERSITY

Raman spectroscopic signatures of magnetic fluctuations in Kagome planes of $Pr_2Zr_2O_7$ in H || [111]

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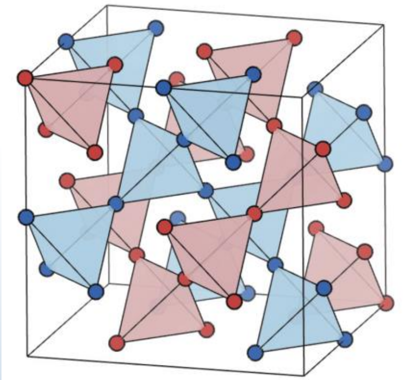
QuantEmX Award

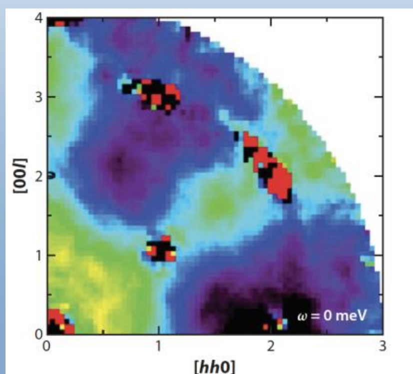
$Pr_2Zr_2O_7$: Frustrated 3D magnet



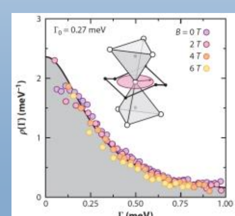
Pyrochlore lattice is a frustrated 3 D lattice

Pr^{3+} Ising spins, J=4, ground state non-Kramers doublet

With only J_z would form a spin ice state
An appearance of J_x would lead to spin ice fluctuations



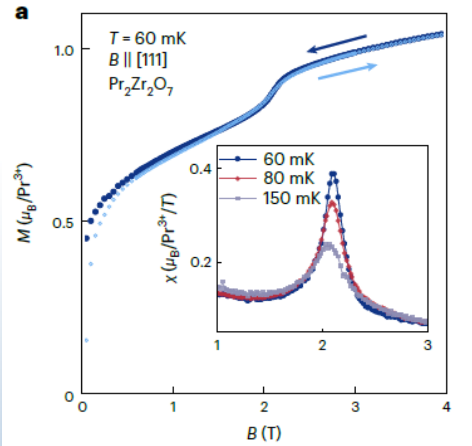
Neutron scattering: Spin-ice-like pattern
90% inelastic signal



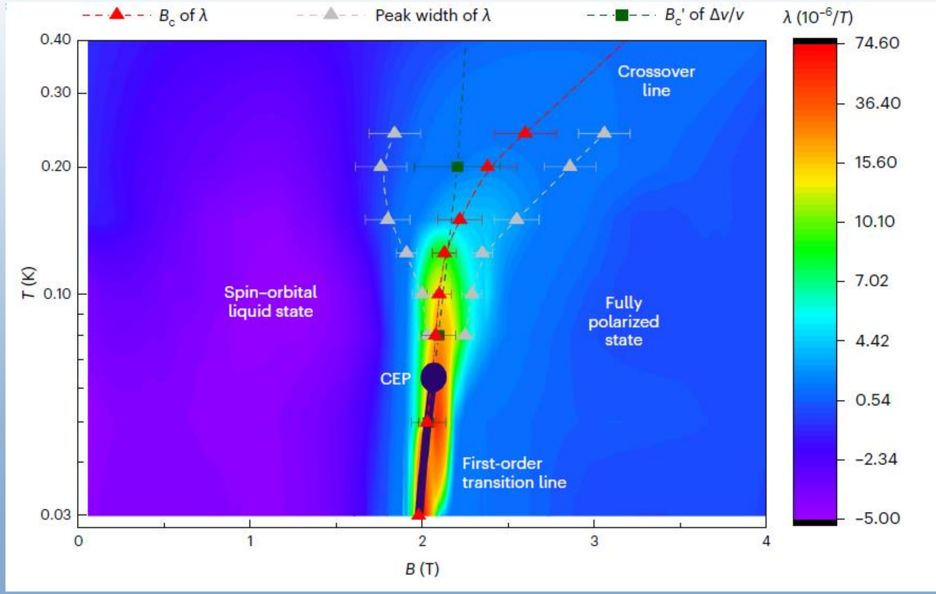
Quenched transverse-field disorder
U(1) quantum spin liquid?

K. Kimura et al., Nat. Commun 4.1 (2013): 1-6
J-J. Wen et al., PRL 118.10 (2017): 107206
N. Martin et al. PRX 7.4 (2017): 041028
J. Rau and M. Gingras, ARCMP (2019)

Unsolved questions



At T<0.4 K under H||[111] @ 2 T:
a transition between
spin-orbital liquid and fully polarized state
Evidenced by magnetostriction



S. Petit et al
Phys Rev B 94 165153

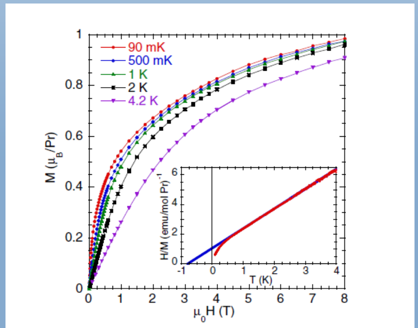


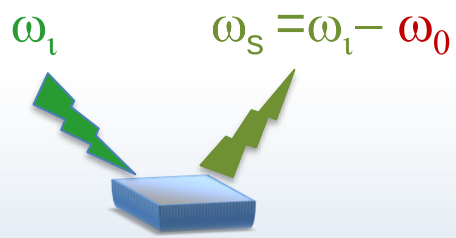
FIG. 3. $H \parallel [111]$: M vs H for several temperatures. Inset: H/M vs T in $\mu_0 H = 9$ mT. The line is a fit to the Curie-Weiss law between 1 and 4.2 K: $H/M = 1.055 + 1.328\,T$.

N. Tang et al Nature Physics 19 92 (2022)

Why magnetization saturates at lower values?

Will we observe spin polarized state at higher temperatures?

Raman scattering spectroscopy to study $Pr_2Zr_2O_7$

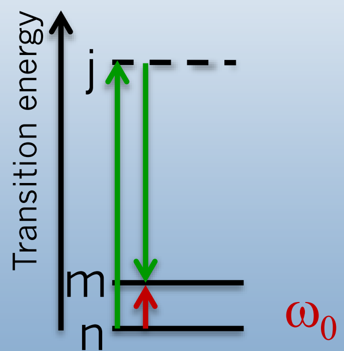


Studies of magnetic systems by Raman scattering: coupling with ***E*** vector of e-m wave

Collective excitations:

Two-magnon scattering
In non-colinear AFM

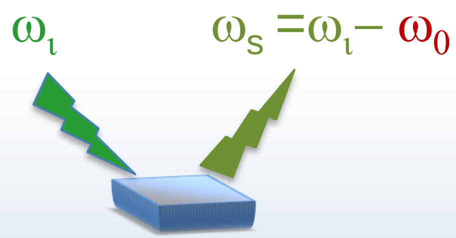
One-magnon
In systems with strong SOC
@ Γ-point

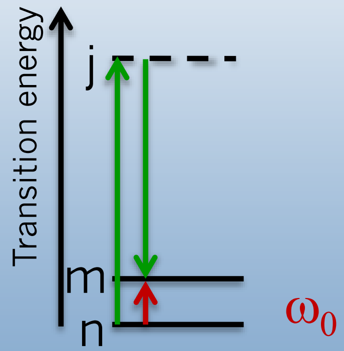


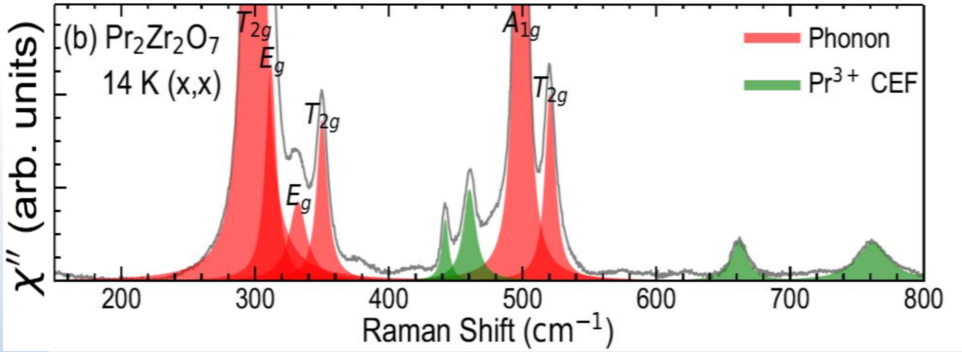
Local excitations:
Crystal field excitations (CF)

$$\left(\tilde{\alpha}_{\alpha\beta}\right)_{mn} = \frac{1}{\hbar}\sum_{j\neq m,n}\left[\frac{\langle\tilde{\Psi}_m|\hat{\mu}_\alpha|\tilde{\Psi}_j\rangle\langle\tilde{\Psi}_j|\hat{\mu}_\beta|\tilde{\Psi}_n\rangle}{\omega_{jn}-\omega_0-i\Gamma_j}+\frac{\langle\tilde{\Psi}_m|\hat{\mu}_\beta|\tilde{\Psi}_j\rangle\langle\tilde{\Psi}_j|\hat{\mu}_\alpha|\tilde{\Psi}_n\rangle}{\omega_{jm}+\omega_0+i\Gamma_j}\right]$$

Raman scattering spectroscopy to study $Pr_2Zr_2O_7$







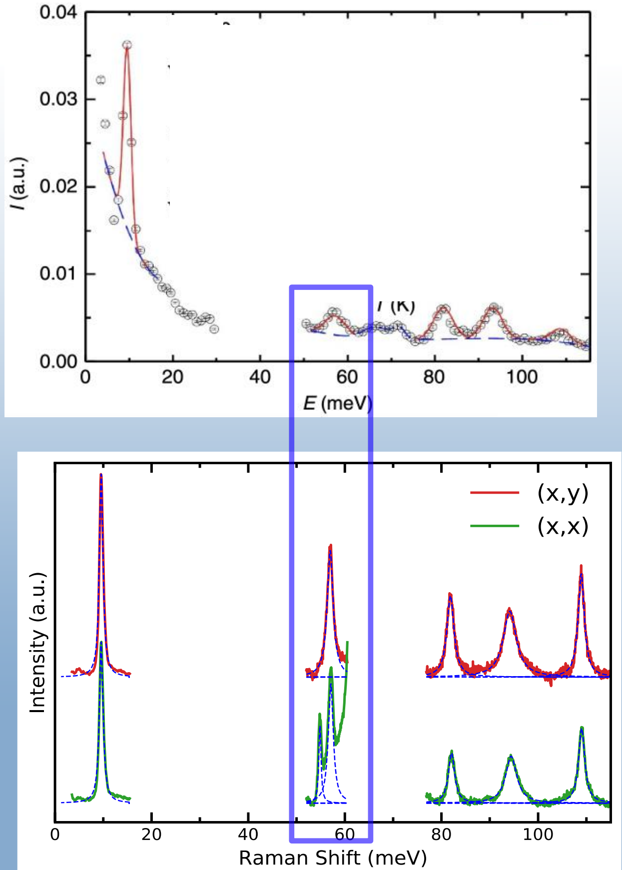
Y. Xu et al PRB 105, 075137 (2022)

For $Pr_2Zr_2O_7$ we observe:

- Γ-point phonons: potentially can detect disorder
- **Crystal field excitations (CF)**

Pr^{3+} crystal field excitations in $Pr_2Zr_2O_7$

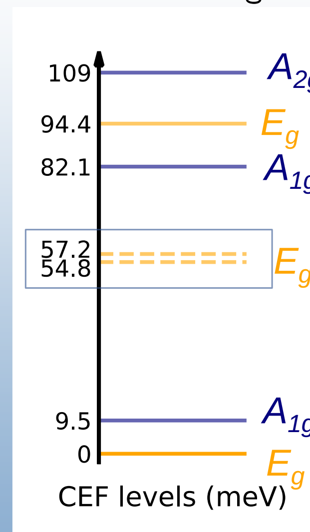
Neutron Scattering @ T = 7.8 K



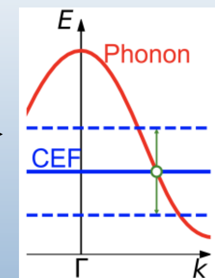
Raman Scattering @ T = 6 K

In D_{3d} symmetry, Pr $^{3+}$ $^{3}H_{4}$ level is split into 3 doublets and 3 singlets:

$\Gamma = 3E_g + 2A_{1g} + A_{2g}$



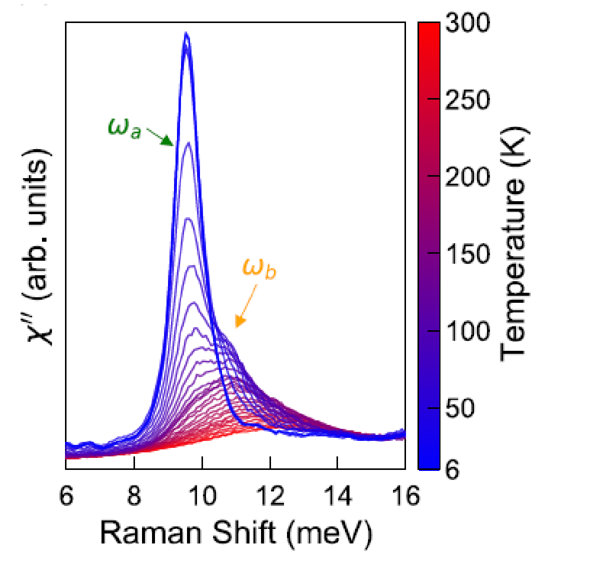
Vibronic coupling



- Raman CEF spectra are in agreement with NS
- Provide much higher energy resolution: 0.2 meV. Allow to resolve vibronic effects and levels splitting

Neutron scattering: K. Kimura et al., Nat. Commun (2013)
Raman scattering: Y. Xu et al PRB 104, 075125 (2021)

$E_g^0 \rightarrow A_{1g}$ CEF transition in $Pr_2Zr_2O_7$

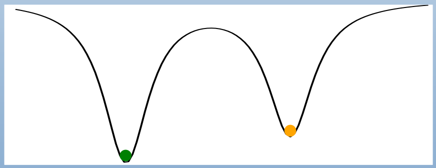


- Ground (E_g) to 1st exited state (A_{1g}) transition characterizes ground state splitting.

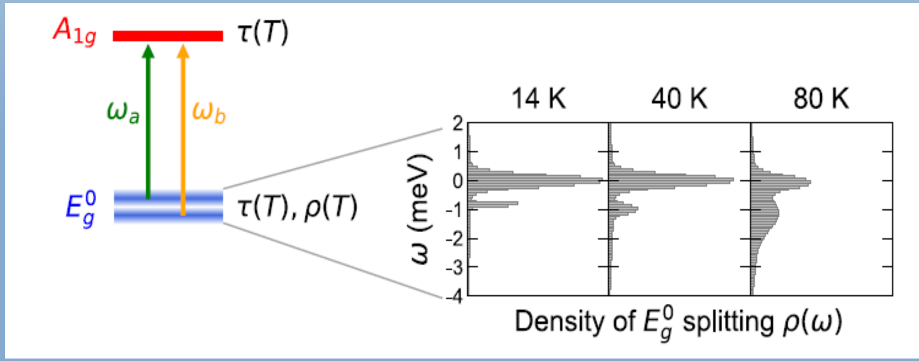
$$\chi''(\omega_i, T) = B_{nk} \int_{-\infty}^{\infty} \rho(\omega', T) L(\omega_i - \omega', T) d\omega'$$

E_g DOS   Life time –determined width

- Splitting due to a double potential for Pr^{3+}, static or slow dynamic

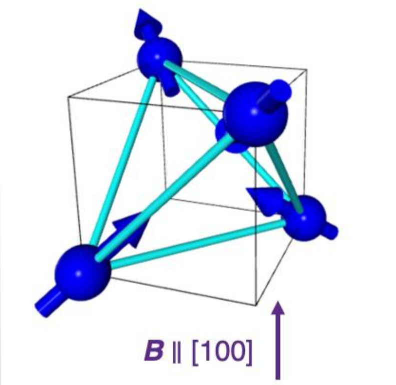


"Freezes out" below 30 K

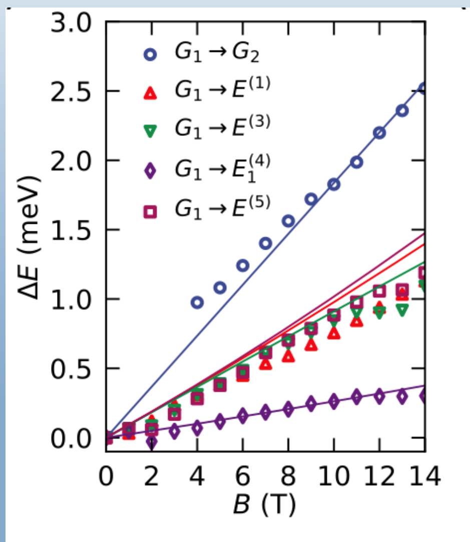


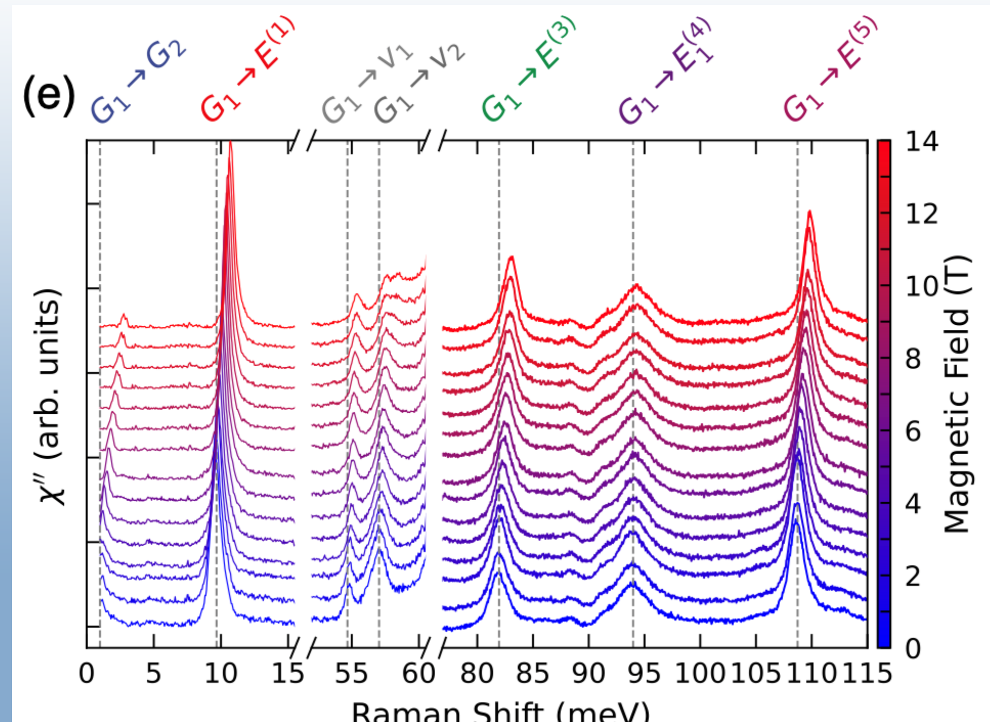
Y. Xu et al PRB 104, 075125 (2021)

H||[100]: Zeeman splitting



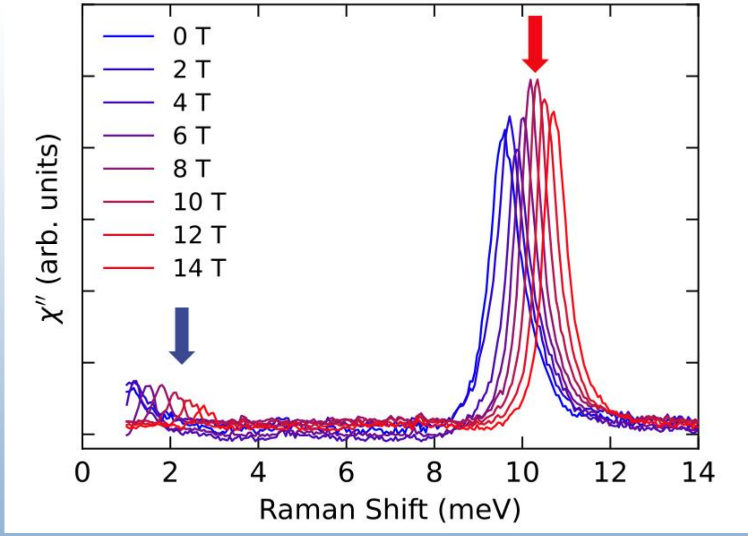
To simplify: assuming 2 in- 2-out state for calculations of Zeeman splitting
$\Delta\omega$ (H)= $-\mu_B g_J$ (**B J**), g_J=4/5

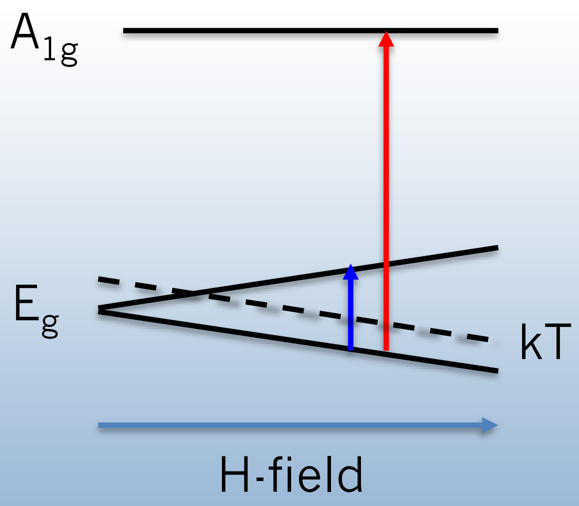


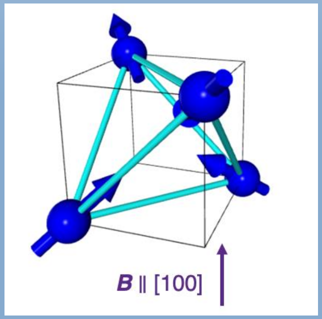


Experimental Zeeman splitting is in a good agreement with calculations

$E_g^0 \rightarrow A_{1g}$ CEF transition in $Pr_2Zr_2O_7$ in H||[100]



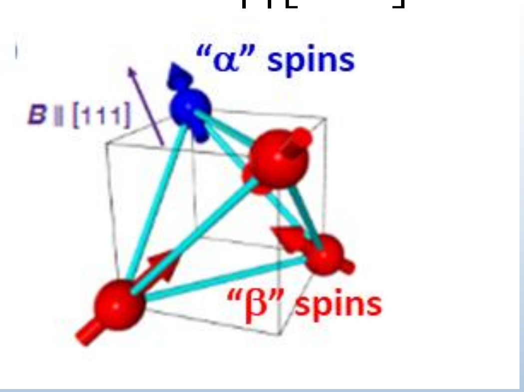




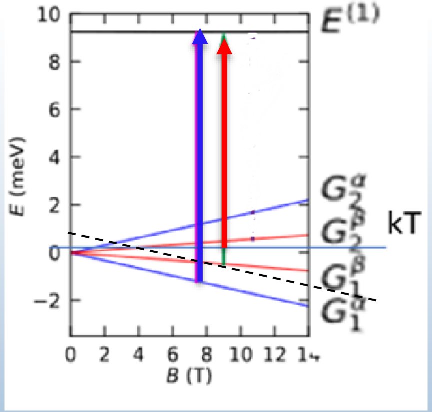
Transition $E_g \rightarrow A_{1g}$
probes Zeeman splitting of the ground state

H||[111]: Zeeman splitting?

Expected: Fully polarized state under H||[111]

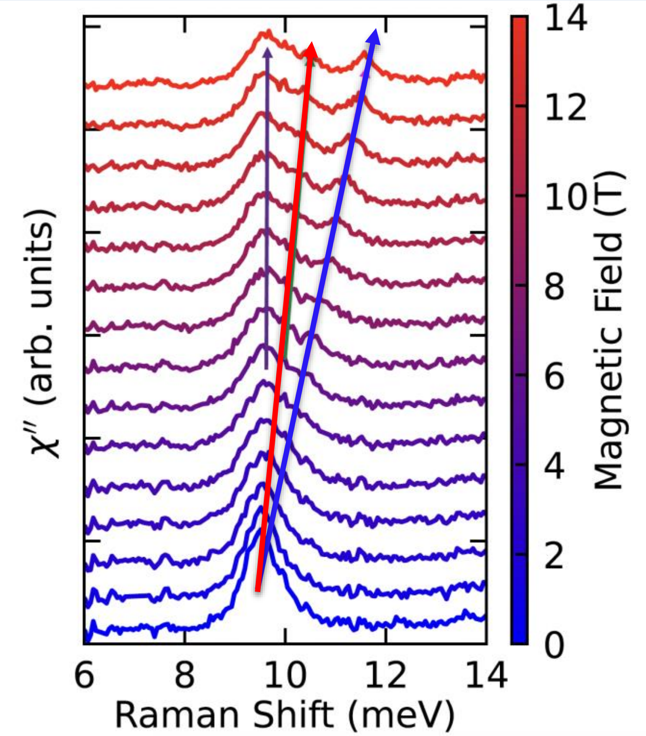


Different splitting for "α" and "β" spins

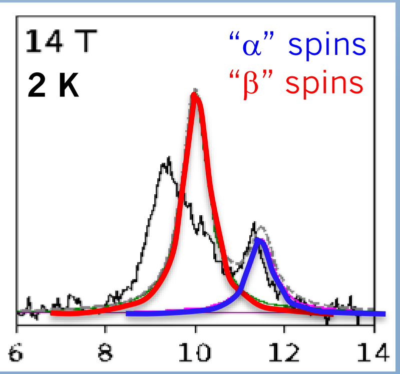


- Zeeman splitting for β spins is lower than expected
- The line is broadened with H-field:

2 K χ'' vs H||[111]



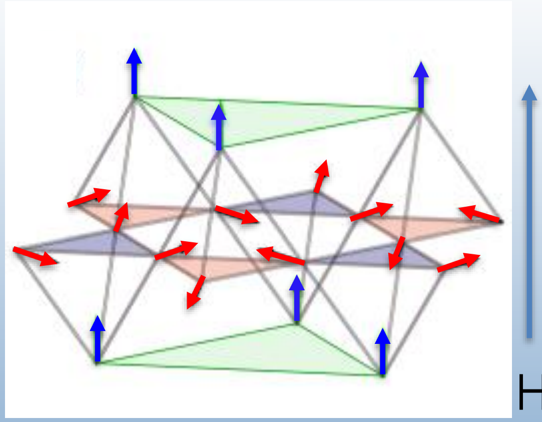
Data vs model: Zeeman splitting for α spins is reproduced, Splitting for "β" spins is smaller



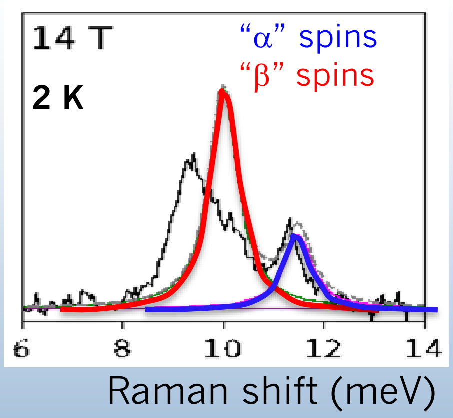
Raman shift (meV)

H||[111]: Spins in Kagome planes

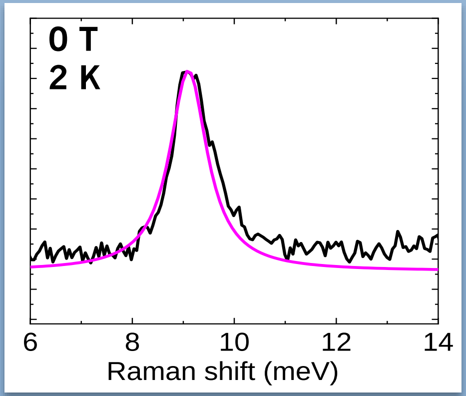
In H||[111] pyrochlore structure "separates" triangular lattice planes and Kagome plains



Not a "frozen" structural disorder:
It should increase the line shape for α and β spins.

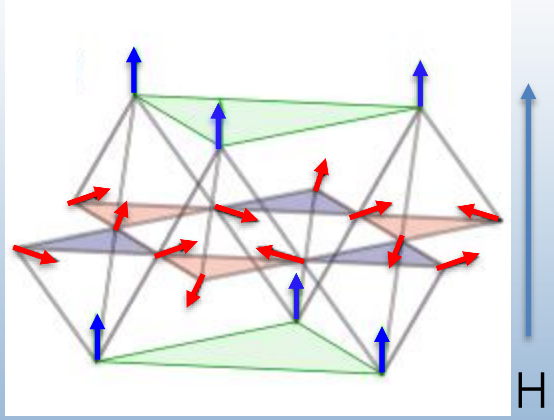


Line width @ 0T is larger than for α spins @14T



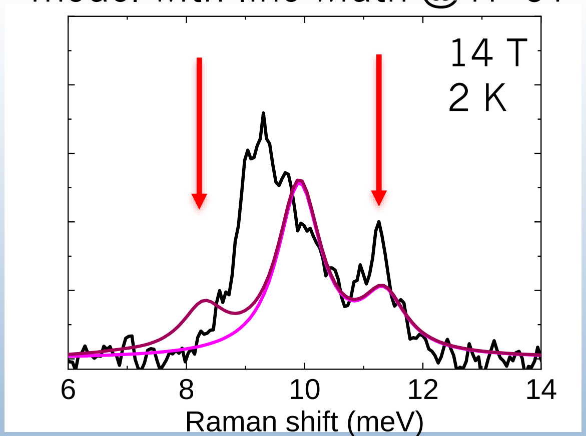
H||[111]: Spins in Kagome planes

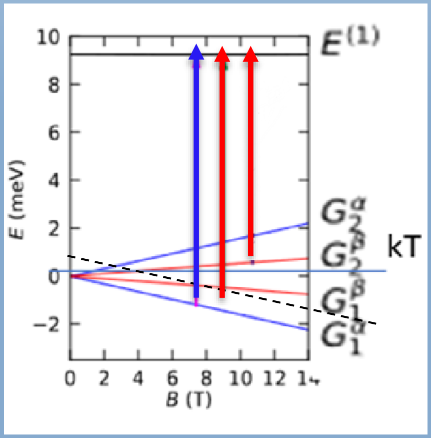
In H||[111] pyrochlore structure "separates" triangular lattice planes and Kagome plains



Not spins "frozen" in 2I2O/3I1O in Kagome plains

model with line width @ H=0T



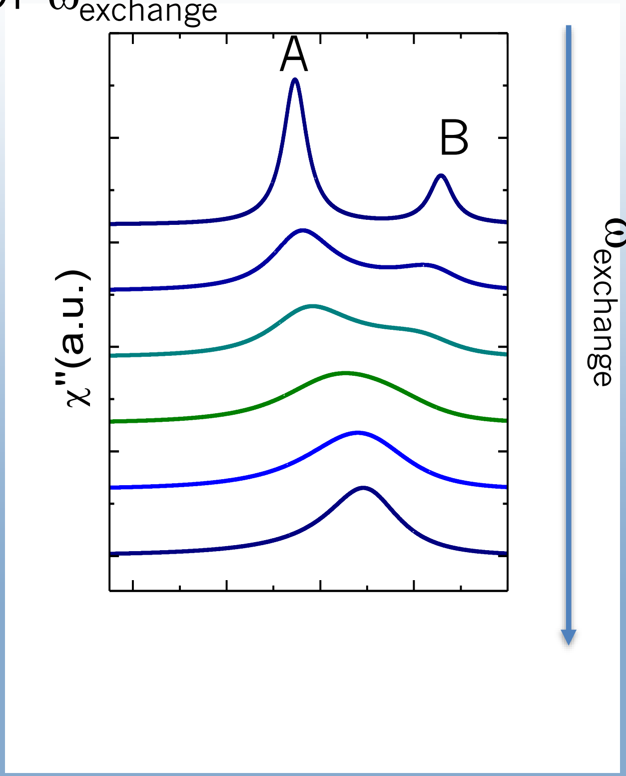


Line shape of a fluctuating system

- Levels A and B
- $\Delta\omega = \omega_A - \omega_B$
- $\omega_{exchange}$

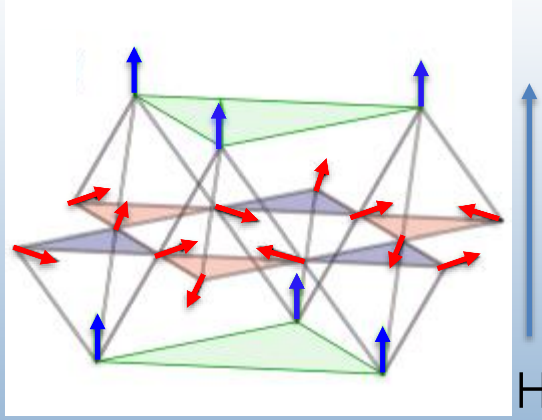
$$I(\omega) \propto Re[\begin{pmatrix} a_R, & a_P \end{pmatrix} \begin{pmatrix} i(\omega - (\omega_{1/2} - \Delta)) + \omega_{EX}/2 + \Gamma/2 & -\omega_{EX}/2 \\ -\omega_{EX}/2 & i(\omega - (\omega_{1/2} - \Delta)) + \omega_{EX}/2 + \Gamma/2 \end{pmatrix}$$

Line shape changes on an increase Of $\omega_{exchange}$



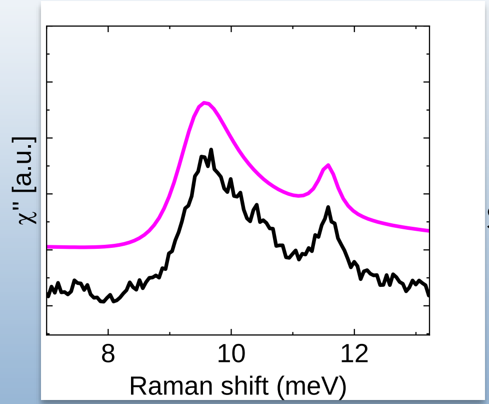
H||[111]: fluctuations of Kagome ice

In H||[111] pyrochlore structure "separates" triangular lattice planes and Kagome plains



Example: $Nd_2Zr_2O_7$
L'Hotel et al Nat. Com. 9, 3786 (2018)

The simplest model:
α- moments described as static
β moments are fluctuating between IN and OUT

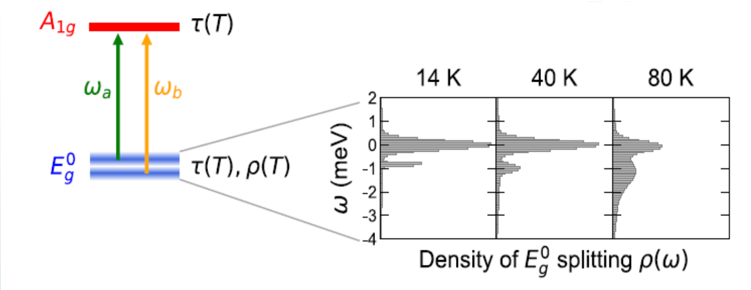


ω(H=0) experimental value
$\Delta\omega$ (H)= $-\mu_B g_J$ (**B J**), g_J=4/5
ω_{ex}= 2meV=0.5THz
N(DOWN)/N(UP)=1/5

Fluctuation of moments in Kagome planes
No spin-polarized state up to 14 T @ 2K

Conclusions

- In H=0: E_g ground state in $Pr_2Zr_2O_7$ shows T-dependent splitting, which suggest a double potential for Pr environment



Y. Xu et al PRB 104, 075125 (2021)

- In H||[111] line shape suggests static J||[111], fluctuating J_{kagome} No fully polarized state up to 14 T