Title: Recent measurements of dipolar-octupolar rare earth pyrochlores

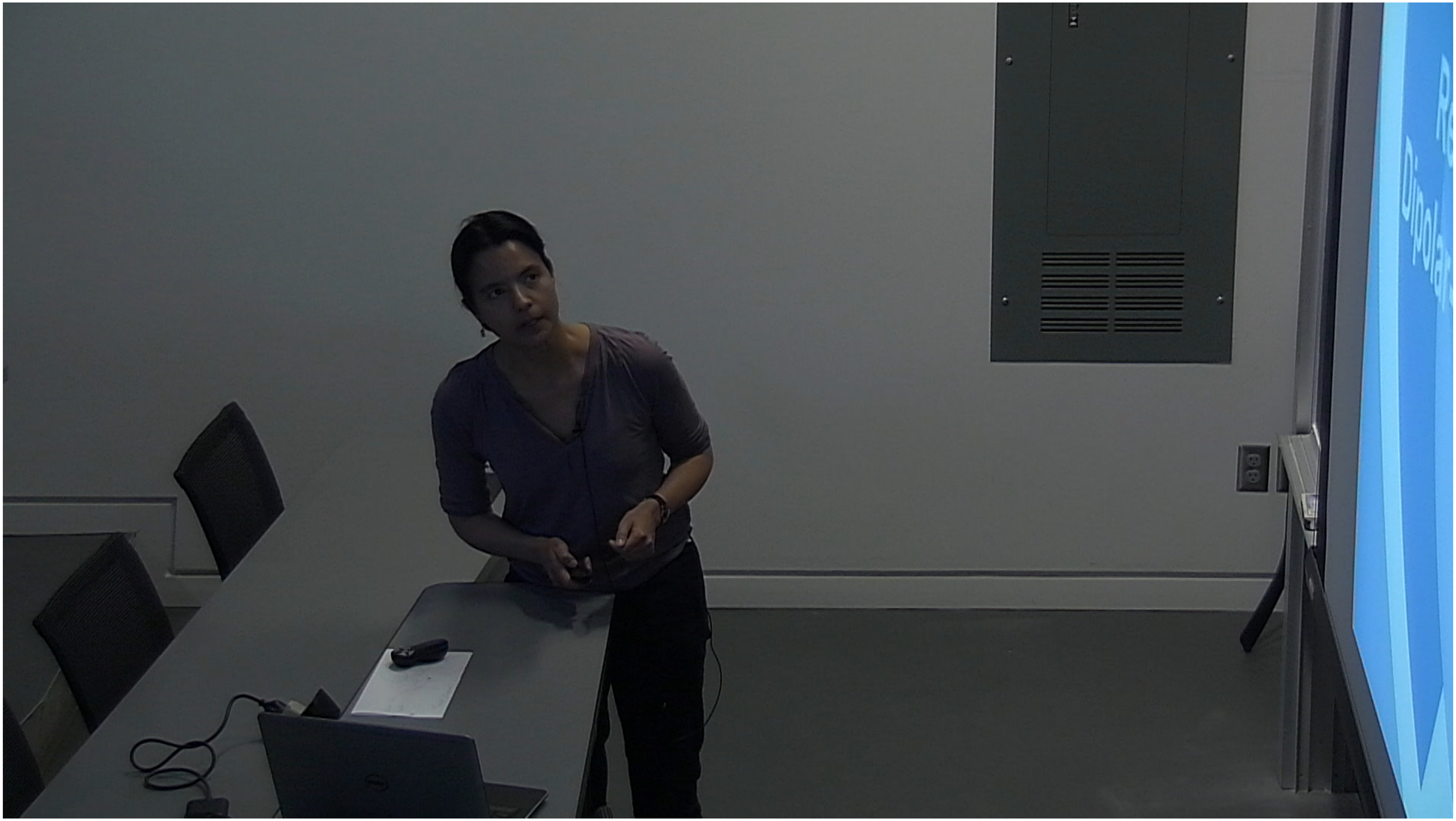
Date: Jun 08, 2017 10:30 AM

URL: http://pirsa.org/17060041

Abstract: This talk will outline recent measurements on the dipolar-octupolar rare earth pyrochlores Nd2Zr2O7 and Nd2Hf2O7. Measurements of their crystal field excitations allows the wavefunction of their ground state Kramerâ€™s doublet to be determined. Both compounds develop long-range magnetic order and their Hamiltonians are extracted by comparing inelastic neutron scattering data to spin-wave theory at low temperatures. The Hamiltonians are used to qualitatively explain AC magnetization measurements as well as neutron data collected in an applied magnetic field. Both system as predicted to lie close to a U(1) spin liquid and the excitation spectrum above the NÃ©el temperature is compared to calculations for bosonic many body quantum spin ice.

Recent Measurements of Dipolar-Octupolar Rare Earth Pyrochlores

Bella Lake
Helmholtz-Zentrum Berlin



Collaborators

HZB Helmholtz Zentrum Berlin

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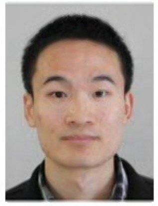

Jianhui Xu
PhD thesis

Alexandros Samartzis

Nazmul Islam
Vivek Kumar Anand
Crystal growth

Andre Sokolowski
AC susceptibility

Akito Sakai
Heat Capacity

UNA Universität Augsburg University

Forschungs-Neutronenquelle Heinz Maier-Leibnitz (FRM II)

Yixi Su, DNS, FRM2
Diffuse neutron scattering

Thomas Herrmannsdörfer
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Jacob Hornung
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Neutron Diffraction
Nicola Casati
X-ray diffraction

PAUL SCHERRER INSTITUT PSI

Clemens Ritter, D20
Neutron diffraction

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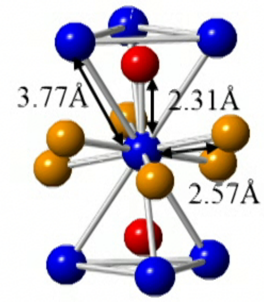
Nd Based Pyrochlores

Ground state

Nd^{3+} $^4I_{9/2}$ S=3/2; L=6; J=9/2

Crystal Field

Local uniaxial symmetry short Nd-O oxide bond along <111>
D_{3d} symmetry about local <111>
dipolar – octupolar doublet



$Nd_2Zr_2O_7$

Nd Based Pyrochlores

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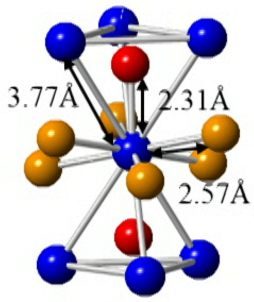

$Nd_2Zr_2O_7$

Interactions

Exchange Interaction: Short range, *Antiferromagnetic*
Dipolar Interaction: Long range, *Ferromagnetic*

balance between **dipole interaction exchange interaction.**

$$\mathcal{H}_{\mathrm{int}} = -\frac{1}{2}\sum_{(i,j)} \mathcal{J}_{ij}\,\mathbf{J}_i\cdot\mathbf{J}_j + \left(\frac{\mu_0}{4\pi}\right)\frac{(g_{\mathrm{L}}\mu_{\mathbf{B}})^2}{2{r_{\mathrm{nn}}}^3}\sum_{(i,j)}\frac{(\mathbf{J}_i\cdot\mathbf{J}_j - 3\mathbf{J}_i\cdot\hat{r}_{ij}\hat{r}_{ij}\cdot\mathbf{J}_j)}{(r_{ij}/r_{\mathrm{nn}})^3}$$

Ratio of exchange to dipole interaction is stronger in light than heavy RE

Outline

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- $Nd_2Zr_2O_7$ - Sample Preparation
- Crystal field excitations - dipolar/octupolar doublet
- Magnetic interactions and order – T_N=0.4K
- Magnetic structure – all-in-all-out order
- Excitation – gapped flat pinch point mode & dispersive excitations
- Spin-wave theory – Hamiltonian
- Excitations above T_N
- Some results for $Nd_2Hf_2O_7$

Sample Preparation – $Nd_2Zr_2O_7$

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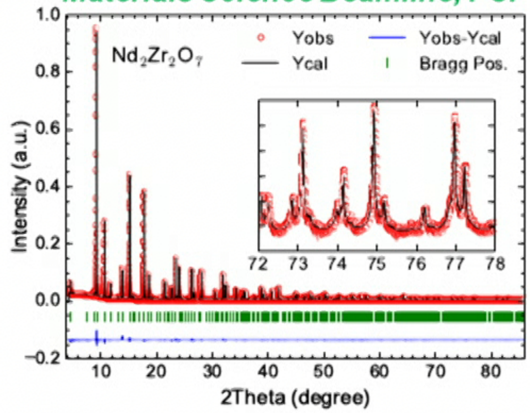
Powder: Solid State Reaction
Nd_2O_3 & ZrO_2 mill, and anneal

	$Nd_2Zr_2O_7$
Annealing in air	1200˚c,2d 1350˚c,2d 1500˚c,3d
Structure	Pyrochlore

Synchrotron X-ray Diffraction
Materials Science Beamline, PSI



a=10.6605Å, x_{O1}=0.335
Site mixing <0.5%

Sample Preparation – $Nd_2Zr_2O_7$

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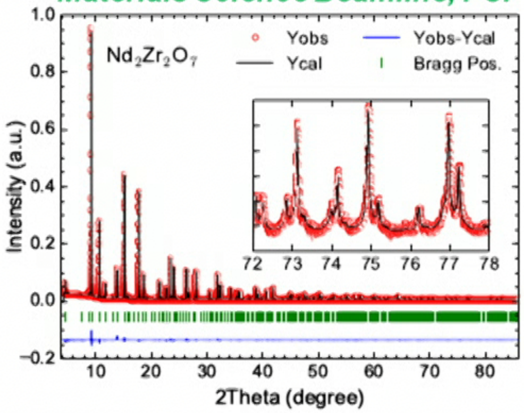
Nd_2O_3 & ZrO_2 mill, and anneal

	$Nd_2Zr_2O_7$
Annealing in air	1200°c,2d 1350°c,2d 1500°c,3d
Structure	Pyrochlore

Single Crystal: Optical floating zone furnace

Ground Nd_2O_3 and ZrO_2 for 1h
Anneal at 1200°c for 12h
↓
Re-mill for 1h and make rods
Anneal at 1200°c for 12h
↓
Grow single crystal
Using traveling technique | Crystal Laboratory at HZB

Synchrotron X-ray Diffraction
Materials Science Beamline, PSI



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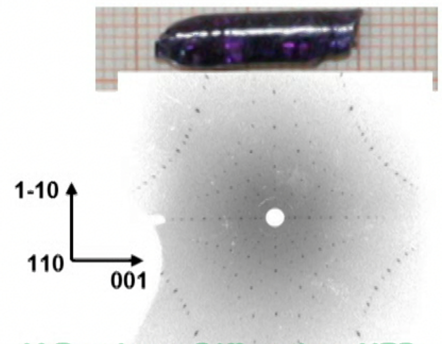

X-Ray Laue Diffraction, HZB

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Crystal Field Excitations – $Nd_2Zr_2O_7$

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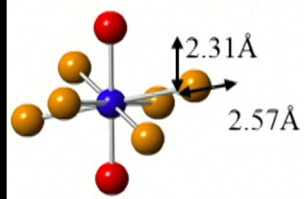
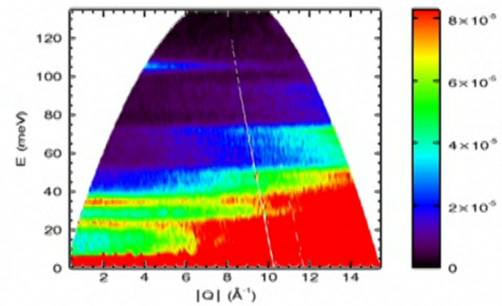
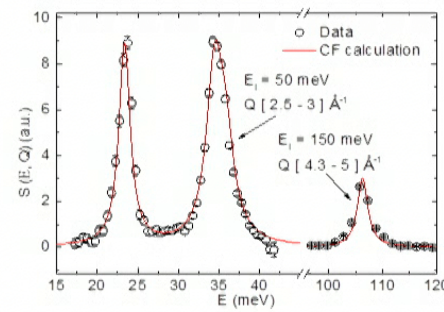
5 Kramers doublets expected for J=9/2 ground multiplet



Powder Neutron Inelastic Scattering
ARCS, ORNL, 150meV, 5K



Big gap: 23.5meV~270K – well isolated ground state
3 magnetic excitations observed – 23, 35, 106 meV



Crystal Field Excitations – $Nd_2Zr_2O_7$

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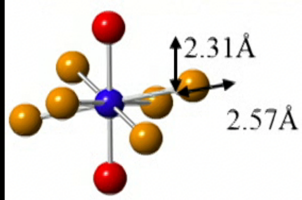

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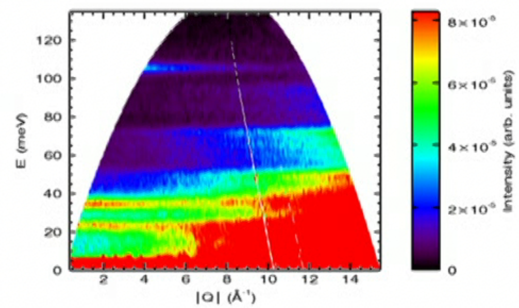

Big gap: 23.5meV~270K – well isolated ground state
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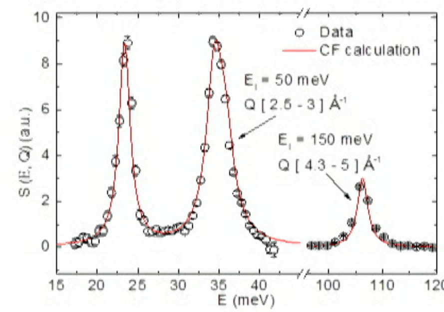
Levels	E_{obs} (meV)	E_{cal} (meV)	I_{obs}	I_{cal}
Γ_{56}^+	0	0	-	2.5
Γ_4^+	23.4(2)	23.36	0.58(5)	0.558
Γ_{56}^+	34.4(4)	34.44	] 1	0.655
Γ_4^+	35.7(4)	35.81	]	0.345
Γ_4^+	106.2(5)	106.28	0.60(8)	0.525



CEF Hamiltonian (z//<111>): D_{3d} symmetry (tensor operators)

$$H_{\mathrm{CEF}} = B_0^2 C_0^2 + B_0^4 C_0^4 + B_3^4 (C_{-3}^4 + C_3^4) + B_0^6 C_0^6 + B_3^6 (C_{-3}^6 + C_3^6) + B_6^6 (C_{-6}^6 + C_6^6)$$

CEF Ground State: Kramers' Doublet

$$\Gamma_{56}^+ = 0.899|^4I_{9/2}, \pm 9/2\rangle \mp 0.252|^4I_{9/2}, \pm 3/2\rangle + 0.330|^4I_{9/2}, \mp 3/2\rangle \mp 0.112|^4I_{11/2}, \pm 9/2\rangle$$

Ground State Doublet

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Mostly J = 9/2, small mixing of J = 11/2 multiplet; m_J = 9/2,3/2 terms only

Dipolar Moment

$\mu_z = \langle GS|J_z|GS\rangle = 2.65\mu_B$ free ion value of $3.27\mu_B$

$\mu_x = \langle GS|J_x|GS\rangle = 0\mu_B$; $\mu_y = \langle GS|J_y|GS\rangle = 0\mu_B$;

Pseudo spin-1/2 model

$\mu_z = g_{zz}\mu_B\tau_z$; $g_{zz} = 5.30$; $g_{xx} = 0$; $g_{yy} = 0$; $\rightarrow$ Ising!

Ground State Doublet

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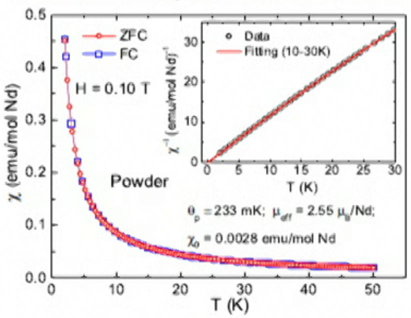


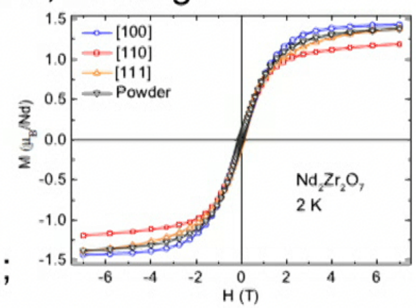
Powder DC susceptibility

Curie-Weiss $\mu_{eff} = 2.55\mu_B$

Magnetization at T=2K

Saturation μ_{100}=1.43(1.53)μ_B

μ_{111}=1.38(1.32)μ_B; μ_{110}=1.19(1.08)μ_B;



Ground State Doublet

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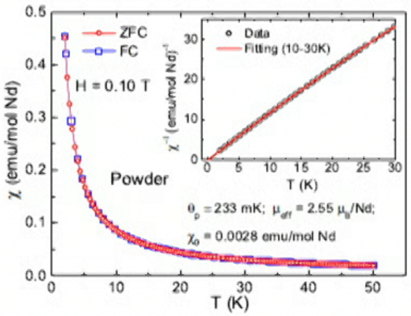

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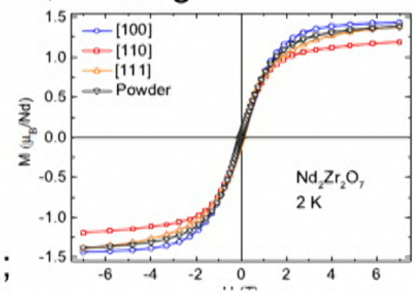
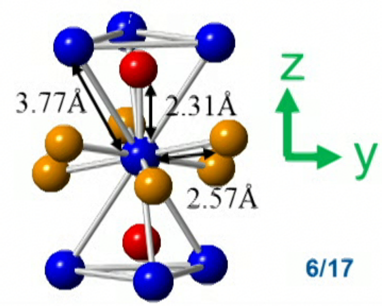


Octupolar moment $T = i(J_+^3 - J_-^3)$ $\mu_{oct} = \langle GS|i(J_+^3 - J_-^3)|GS\rangle \neq 0$

Pseudo spin-1/2 model τ_x & τ_z transform as magnetic dipole

τ_y transforms as octupolar moment



Y.-P. Huang, G. Chen, M. Hermele Phys. Rev. Lett. 112 167203 (2014)

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Interactions and Magnetic Order

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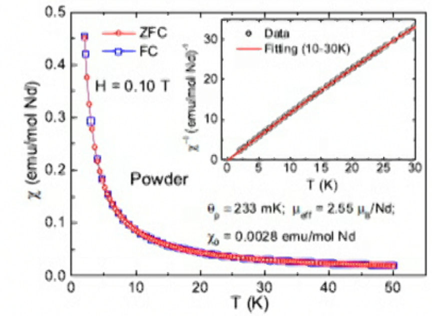
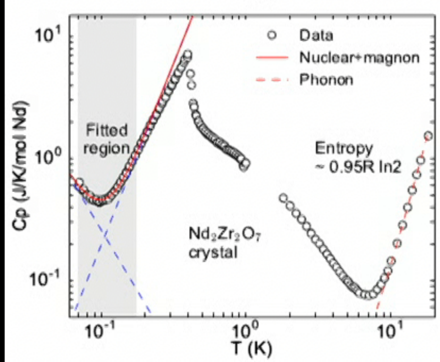
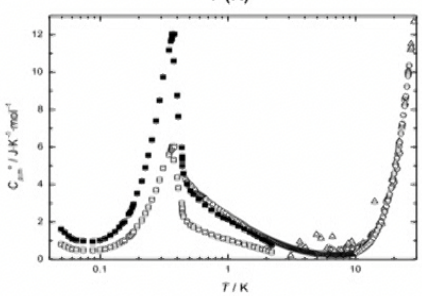


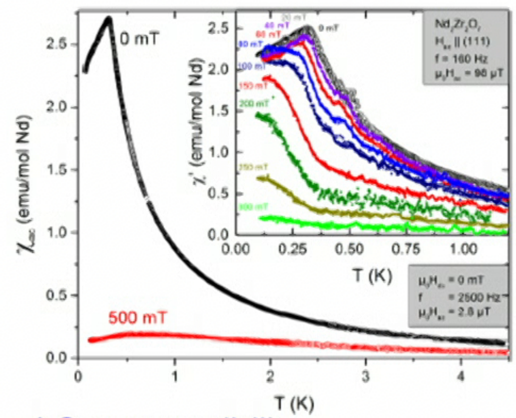
DC susceptibility
Curie-Weiss
10K<T<30K
T_{CW} = +0.233K



specific heat
Crystal 1
T_N=0.311K



specific heat
powder
T_N=0.40K



AC susceptibility
crystal 2
T_N=0.40K

Blöte et al Physica 43, 549 (1969)

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Interactions and Magnetic Order

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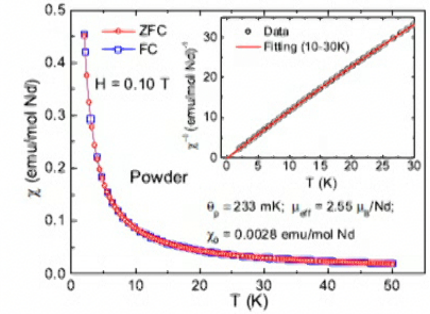

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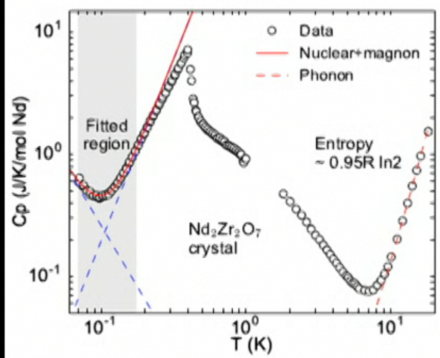

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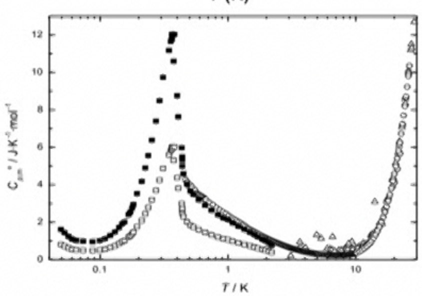

specific heat

powder

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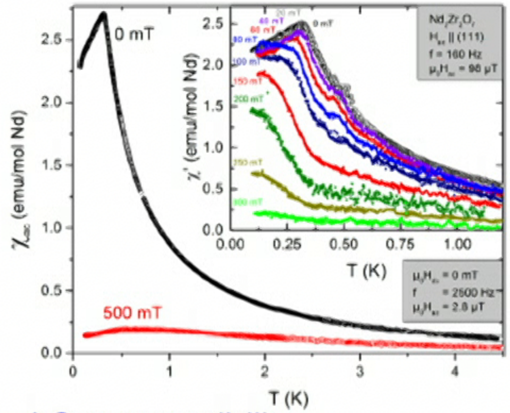
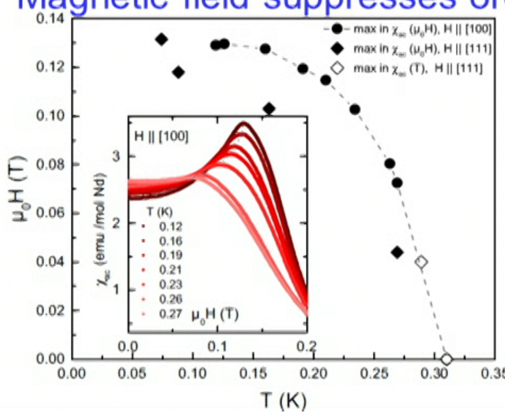


AC susceptibility

crystal 2

T_N=0.40K

Magnetic field suppresses order



Magnetic Structure of $Nd_2Zr_2O_7$

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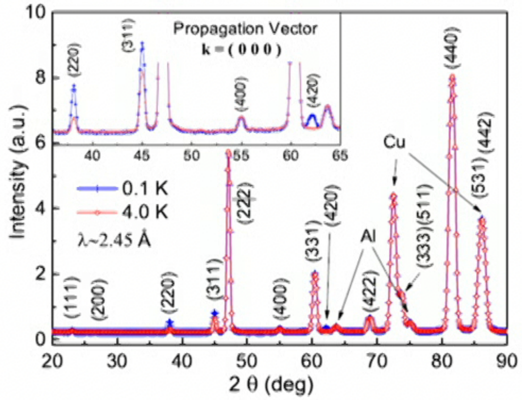
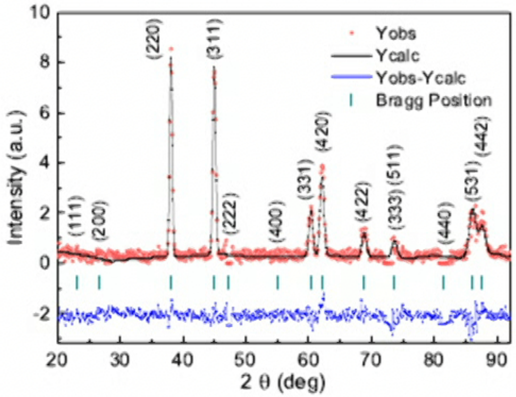
Powder neutron diffraction
DMC PSI





J. Xu, Phys. Rev. B 92, 224430 (2015).

Magnetic Structure of $Nd_2Zr_2O_7$

Powder neutron diffraction
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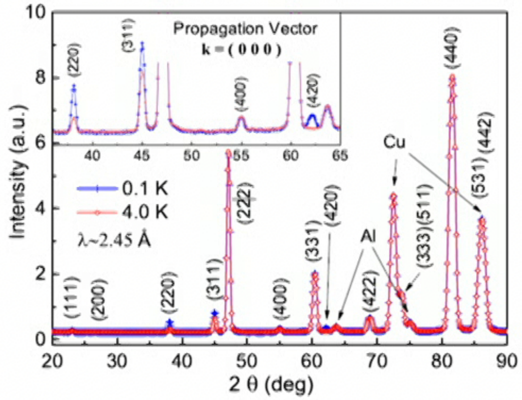

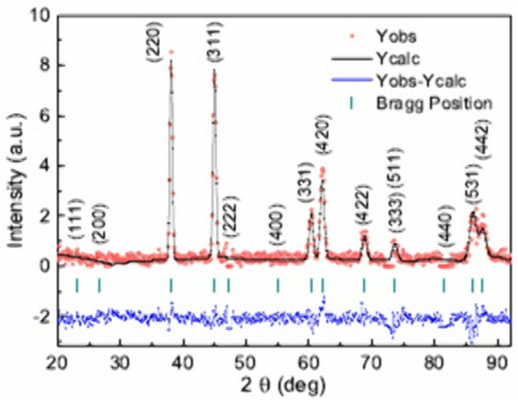

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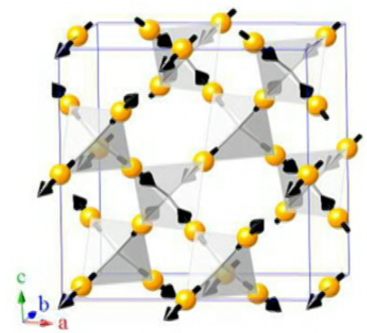
Neutron diffraction:

***k* = 0 structure**

Γ_3^1 'all-in-all-out' antiferromagnetic order



Magnetic Structure of $Nd_2Zr_2O_7$

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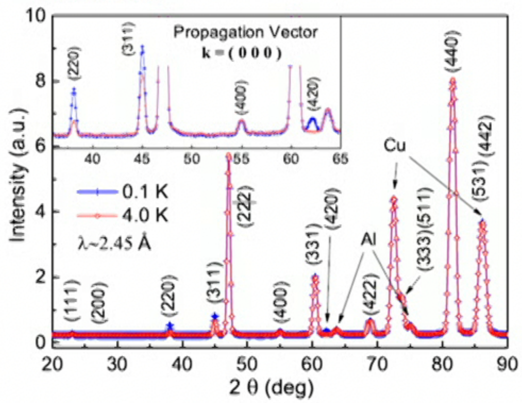
Powder neutron diffraction
DMC PSI



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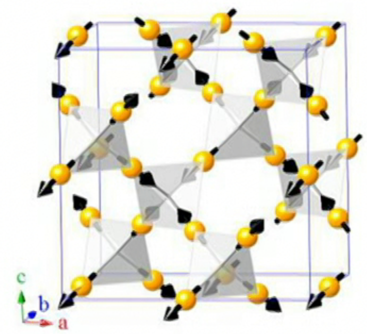
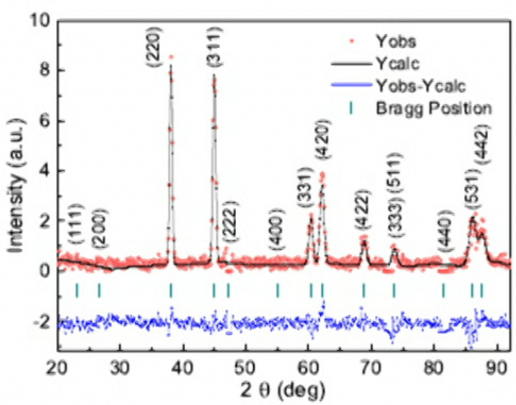
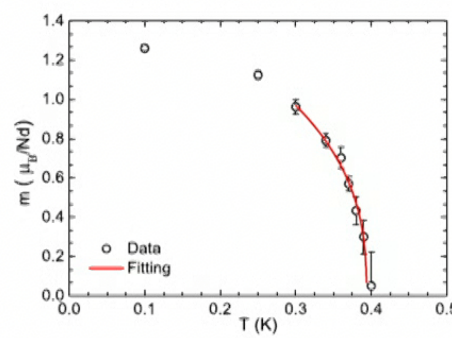




T_N=0.39K

$\mu_{order=}$=1.25μ_B /Nd
2.6 μ_B/Nd (from CEF)



J. Xu, Phys. Rev. B 92, 224430 (2015).

Single Crystal Neutron Scattering

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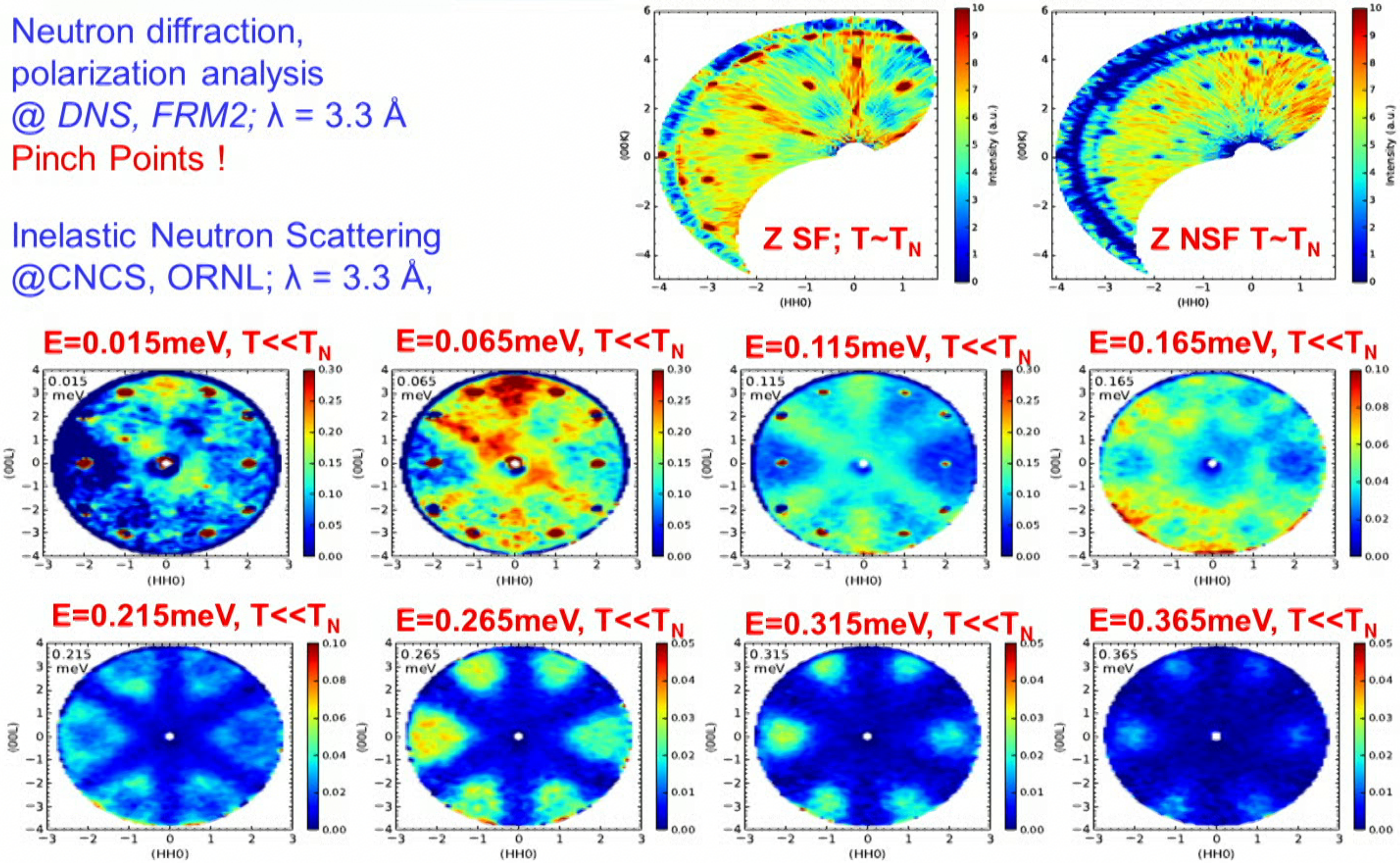
Neutron diffraction,
polarization analysis
@ *DNS, FRM2;* λ = 3.3 Å
Pinch Points !

Inelastic Neutron Scattering
@CNCS, ORNL; λ = 3.3 Å,



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Spin-Wave Theory

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General Hamiltonian for Nd^{3+} ions on a pyrochlore lattice (local frame) given by Y.-P. Huang, G. Chen, M. Hermele *Phys. Rev. Lett. 112 167203 (2014)*

$$\mathcal{H}_{\mathrm{ex}}^{\mathrm{DO}} = \sum_{\langle ij \rangle} \left[\mathsf{J}_x \tau_i^x \tau_j^x + \mathsf{J}_y \tau_i^y \tau_j^y + \mathsf{J}_z \tau_i^z \tau + \mathsf{J}_{xz} \left(\tau_i^x \tau_j^z + \tau_i^z \tau_j^x \right) \right].$$

Rotation about the y-axis gives $$\mathcal{H}_{XYZ}^{\mathrm{DO}} = \sum_{\langle ij \rangle} \left[\tilde{\mathsf{J}}_x \tilde{\tau}_i^{\tilde{x}} \tilde{\tau}_j^{\tilde{x}} + \tilde{\mathsf{J}}_y \tilde{\tau}_i^{\tilde{y}} \tilde{\tau}_j^{\tilde{y}} + \tilde{\mathsf{J}}_z \tilde{\tau}_i^{\tilde{z}} \tilde{\tau}_j^{\tilde{z}} \right].$$

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Spin-Wave Theory

HZB Helmholtz Zentrum Berlin

General Hamiltonian for Nd^{3+} ions on a pyrochlore lattice (local frame) given by Y.-P. Huang, G. Chen, M. Hermele *Phys. Rev. Lett. 112 167203 (2014)*

$$\mathcal{H}_{\text{ex}}^{\text{DO}} = \sum_{\langle ij \rangle} \left[\mathsf{J}_x \tau_i^x \tau_j^x + \mathsf{J}_y \tau_i^y \tau_j^y + \mathsf{J}_z \tau_i^z \tau + \mathsf{J}_{xz} \left(\tau_i^x \tau_j^z + \tau_i^z \tau_j^x \right) \right].$$

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Spin-wave calculation of O. Benton *Phys. Rev. B 94 104430 (2016)*
Comparison to data of S. Petit E. Lhotel et al *Nat. Phys. 10 1038 (2016)*

Best exchange constants $\tilde{\mathsf{J}}_x = 0.103\ \text{meV}, \quad \tilde{\mathsf{J}}_y = 0, \quad \tilde{\mathsf{J}}_z = -0.047\ \text{meV}.$

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Curie-Weiss temperature

$$T_{\mathrm{CW}} = \frac{1}{2k_B}(\tilde{\mathsf{J}}_z \cos^2(\vartheta) + \tilde{\mathsf{J}}_x \sin^2(\vartheta)),$$

$T_{cw} \sim 0.2\mathrm{K} \Rightarrow \vartheta = 47.6°$

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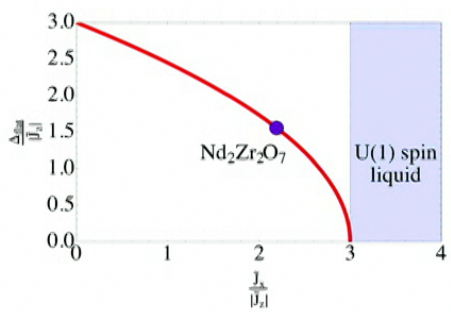
$T_{cw} \sim 0.2\mathrm{K} \Rightarrow \vartheta = 47.6°$

Flat gapped pinch point mode

$$\Delta_{\mathrm{flat}} = \sqrt{(3|\tilde{\mathrm{J}}_z| - \tilde{\mathrm{J}}_x)(3|\tilde{\mathrm{J}}_z| - \tilde{\mathrm{J}}_y)},$$



Gapless for $J_x/|J_z|=3$

Comparison of Data and SWT

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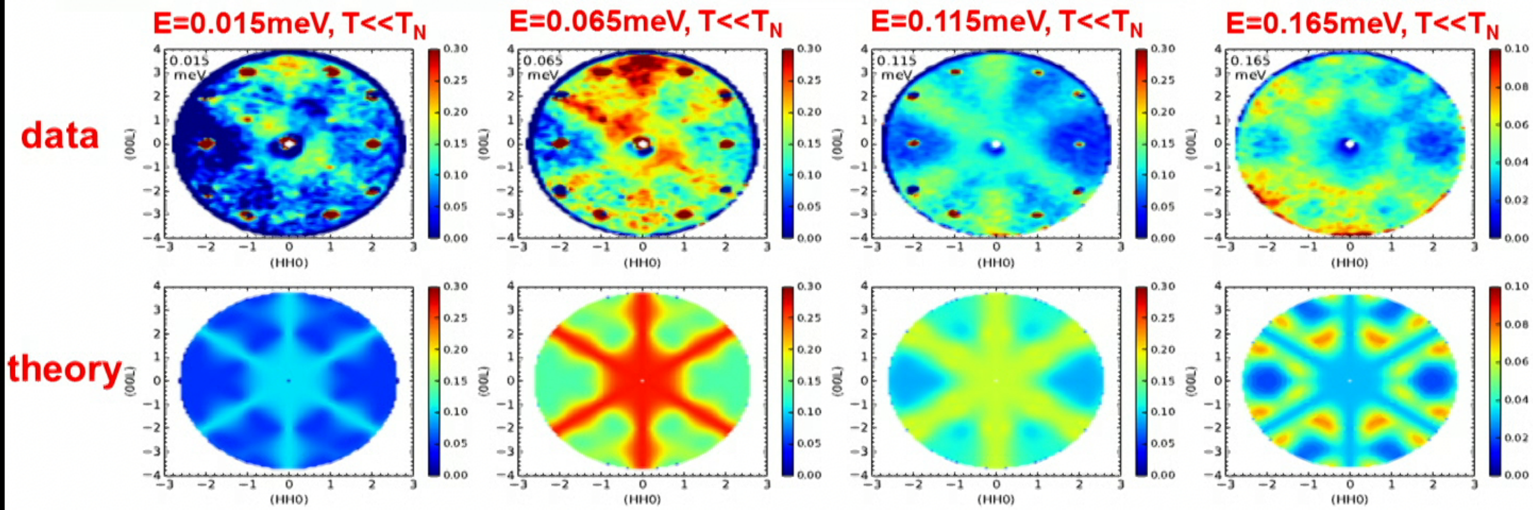

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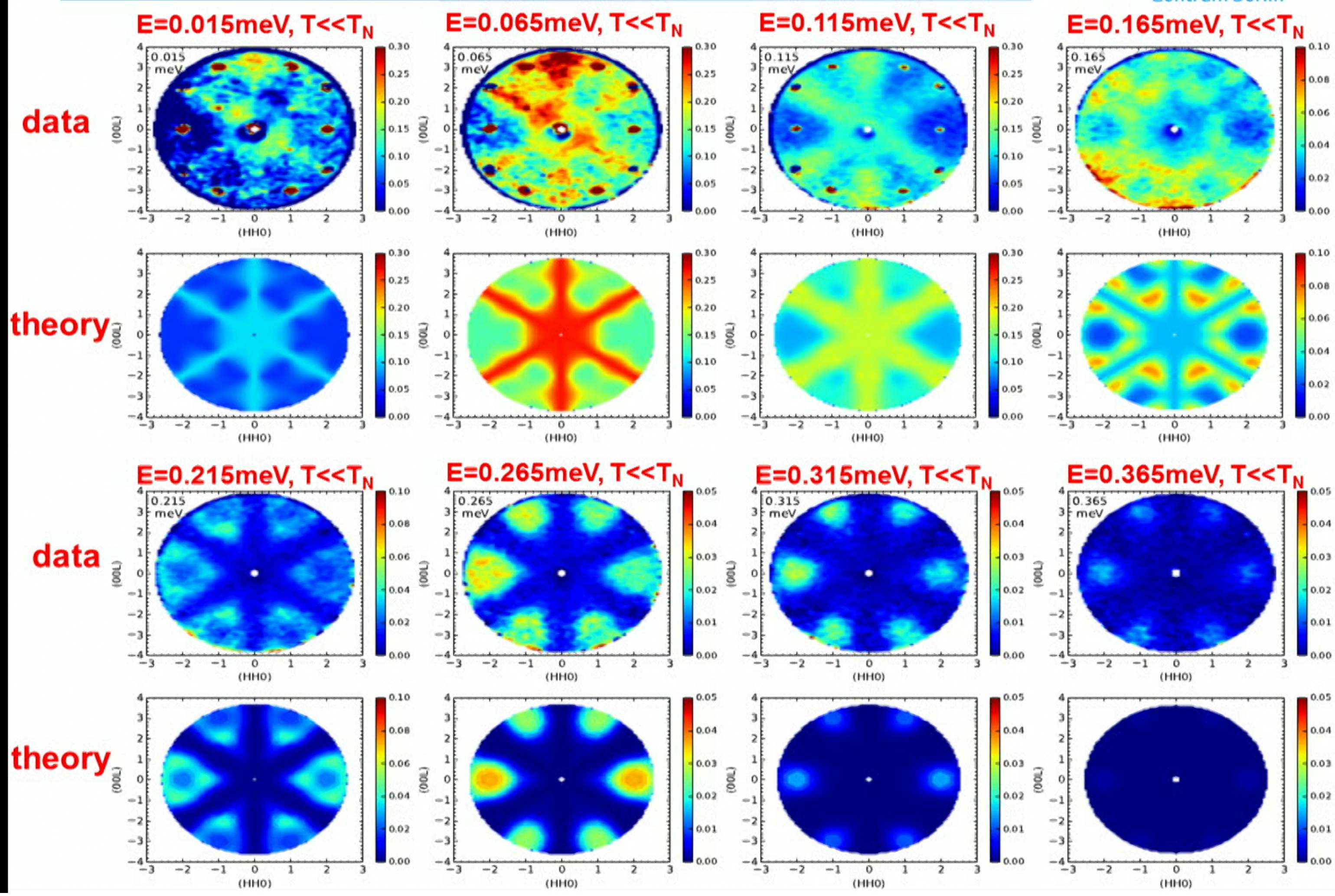
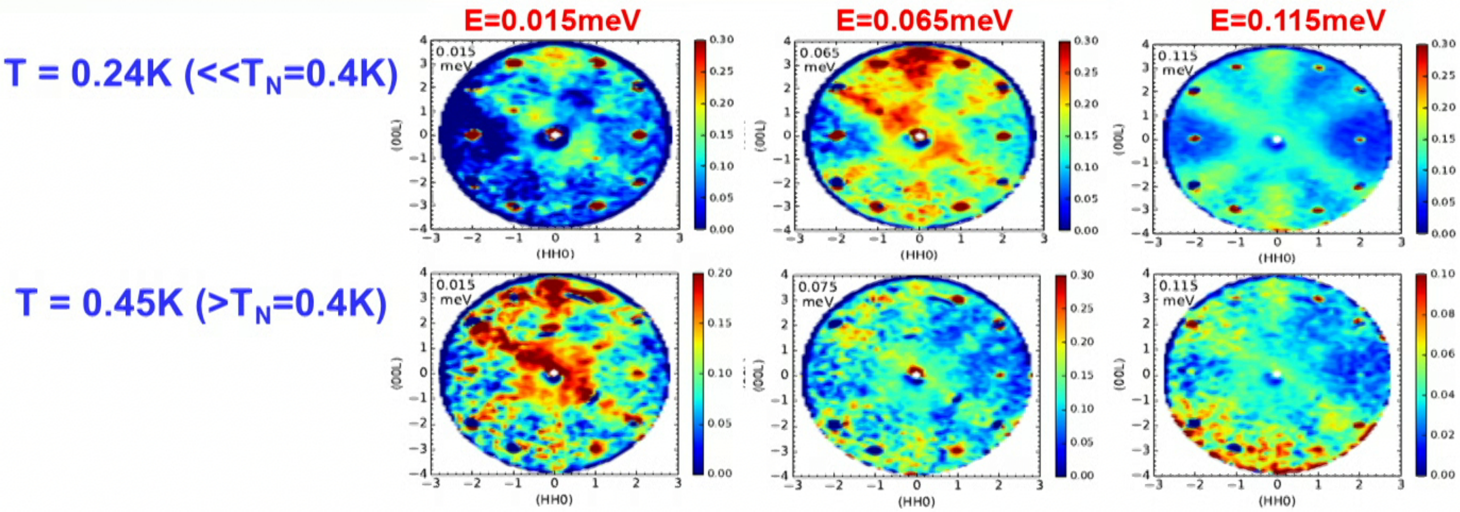
Comparison of Data and SWT



Excitations above T_N



Excitations above T_N

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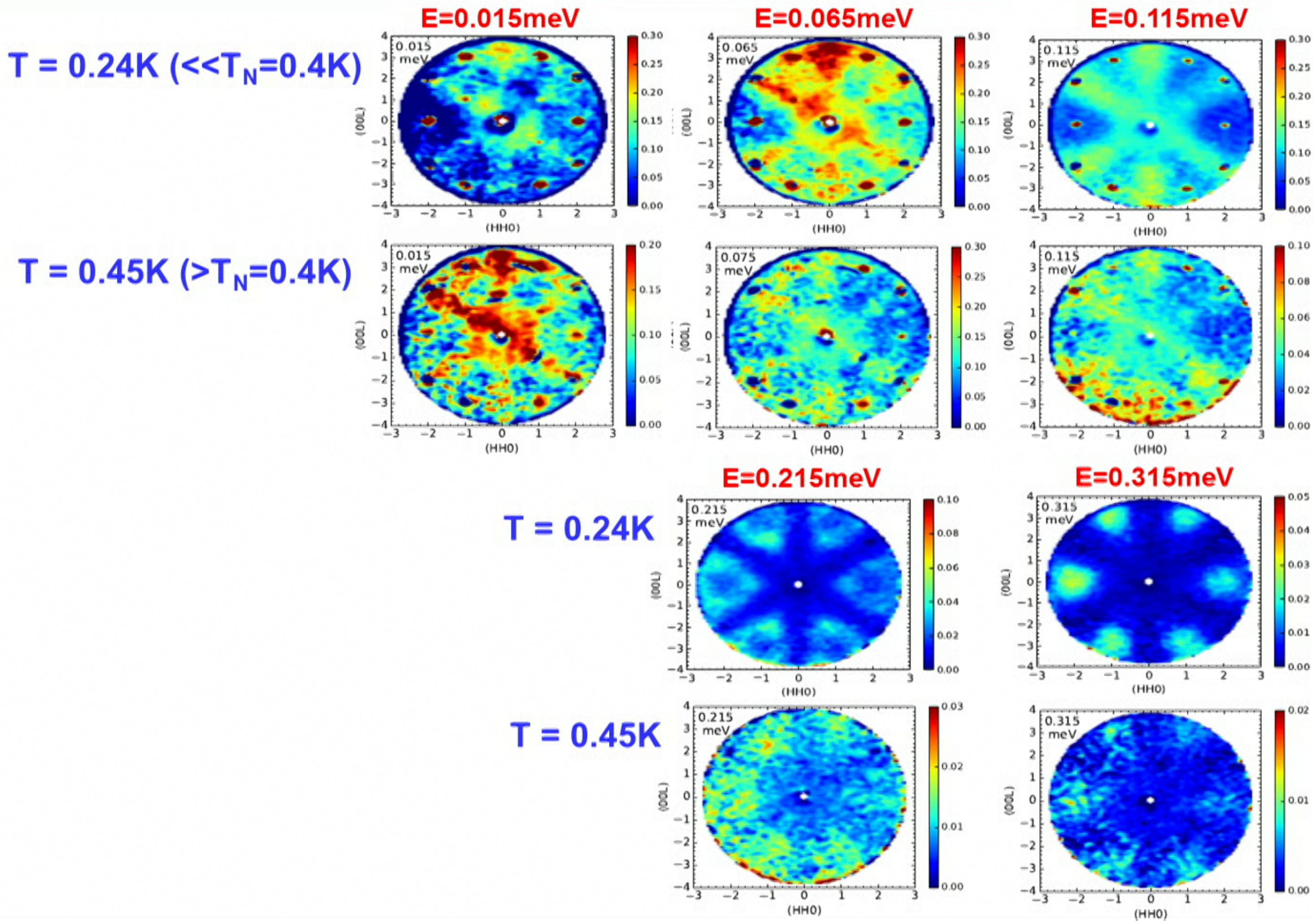

Excitations above T_N

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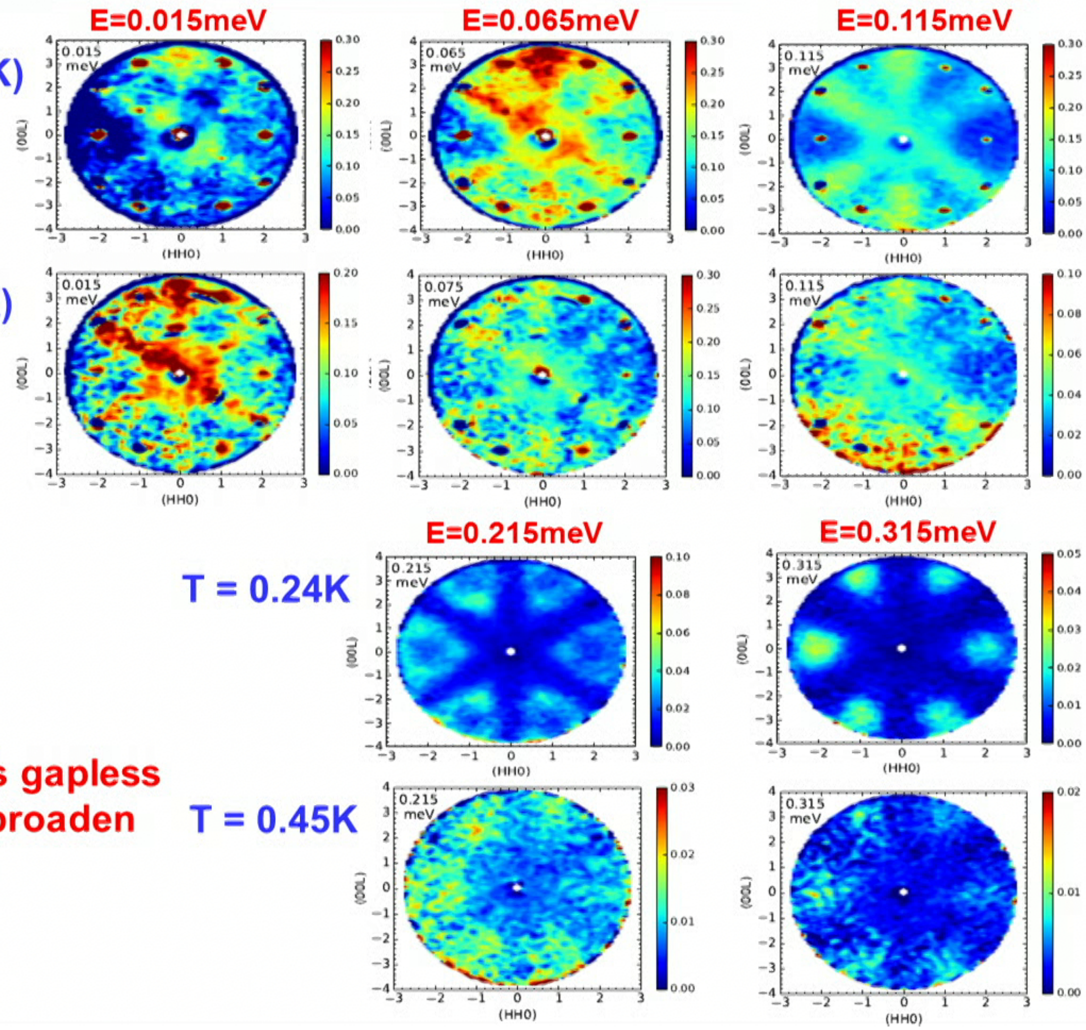
T = 0.24K (<<T_N=0.4K)

T = 0.45K (>T_N=0.4K)



Above T_N

- **Pinch point mode is gapless**
- **Dispersive modes broaden**
- **Features at (2,2,0)**

$Nd_2Hf_2O_7$

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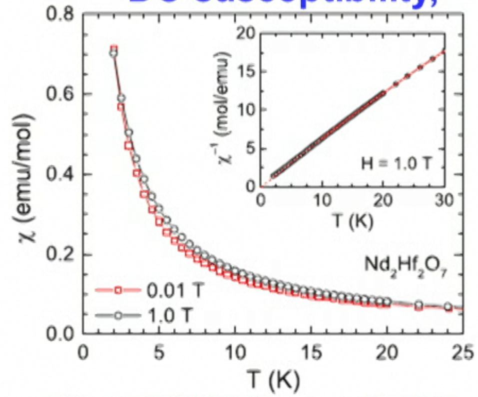
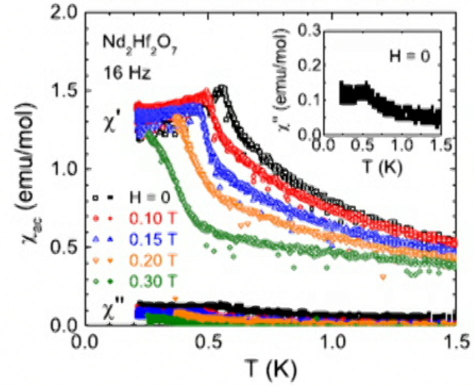
DC susceptibility,



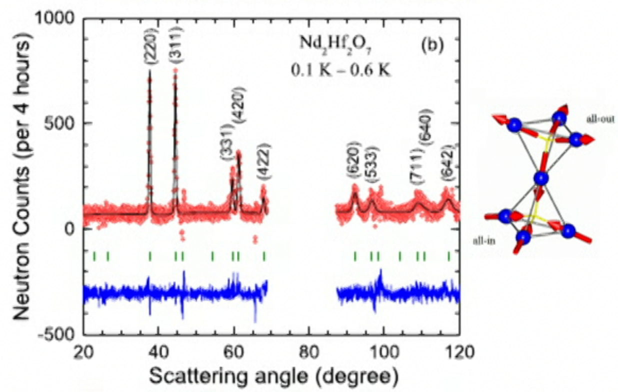
T_{CW}=+0.24K, μ_{eff}=2.45μ_B

AC susceptibility,



T_N=0.55K,
H dependence

Neutron Diffraction



T_N=0.53K, All-in-All-out order

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$Nd_2Hf_2O_7$

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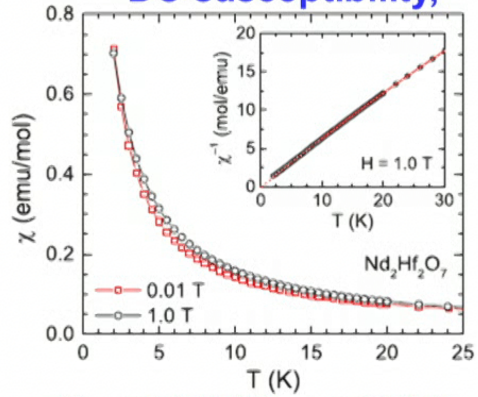
DC susceptibility,



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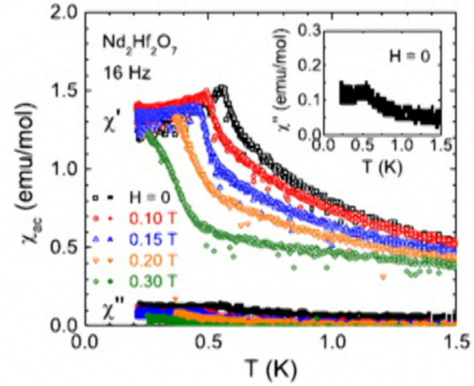
AC susceptibility,



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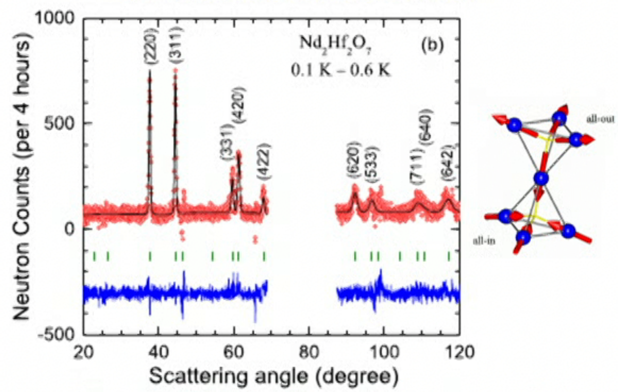
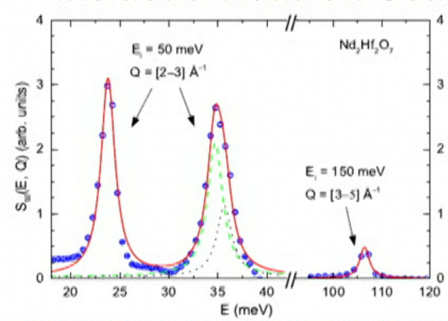
H dependence

Neutron Diffraction



T_N=0.53K, All-in-All-out order

Inelastic Neutron Scattering



μ_{eff}=2.53μ_B

Dipolar-octupolar ground state doublet

$$\Gamma_{56}^{+} = 0.903|^4I_{9/2}, \pm 9/2\rangle + 0.334|^4I_{9/2}, \mp 3/2\rangle \mp 0.232|^4I_{9/2}, \pm 3/2\rangle \mp 0.111|^4I_{11/2}, \pm 9/2\rangle + 0.045|^4I_{13/2}, \pm 9/2\rangle$$

$Nd_2Hf_2O_7$ – magnetic excitations

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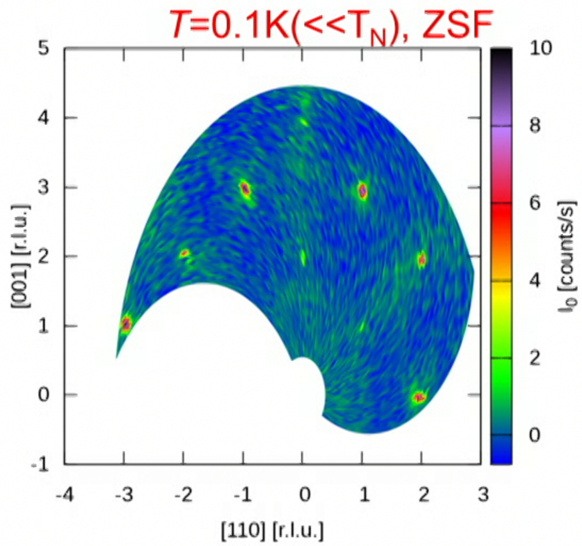
Neutron diffraction, polarization analysis @ *DNS, FRM2;* λ = 3.3 Å



Magnetic Bragg peaks
weak pinch point pattern

$Nd_2Hf_2O_7$ – magnetic excitations

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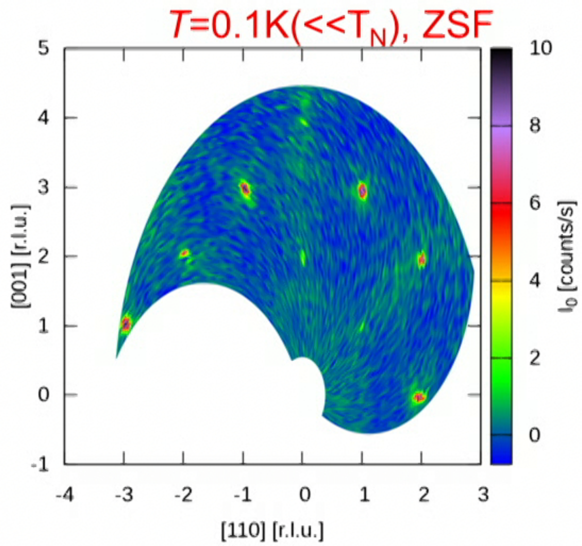

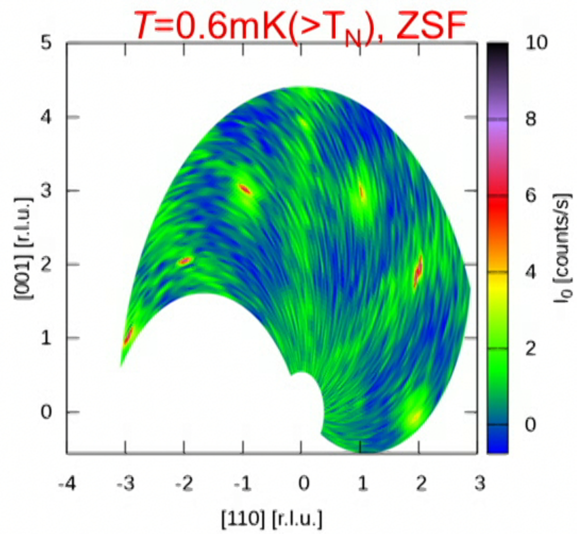

Magnetic Bragg peaks
weak pinch point pattern

Diffuse signal at Bragg peaks
strong pinch point pattern

The pinch point pattern becomes gapped and magnetic Bragg peaks grow below T_N

Summary

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- $Nd_2Zr_2O_7$ Crystal field excitations
  dipolar/octupolar ground state doublet μ_{eff}=2.65μ_B
- Magnetic interactions
  T_{CW}=0.233K and magnetic order at T_N=0.4K
- Magnetic structure
  all-in-all-out order $\mu_{order=}$=1.25μ_B/Nd
- Excitation
  gapped flat pinch point mode at 0.07meV
  dispersive excitations to 0.4meV
- Spin-wave theory
  Hamiltonian Jx=0.103, Jz=-0.047
- Excitations above T_N
  - Pinch point mode becomes gapless
  - Dispersive excitations broaden
- Similar feactures observed in $Nd_2Hf_2O_7$