Spin Excitations in Stoichiometric $Yb_2Ti_2O_7$

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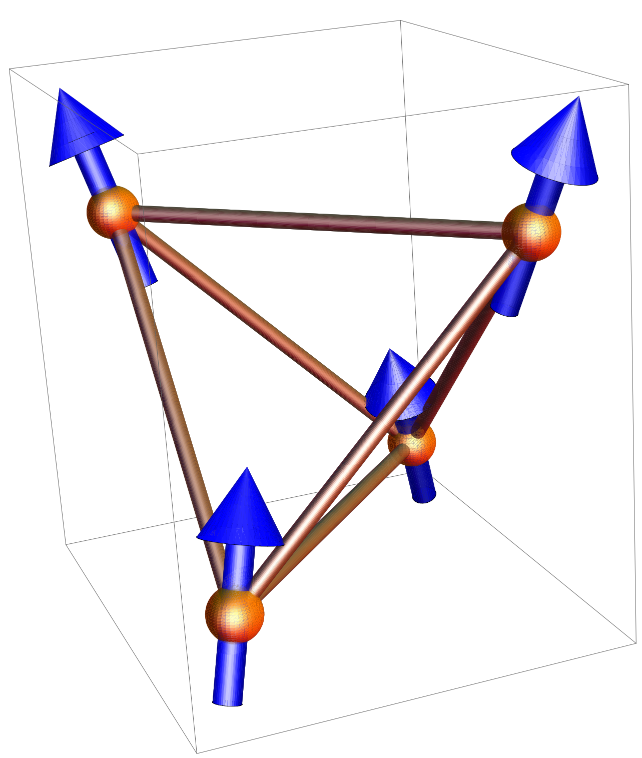
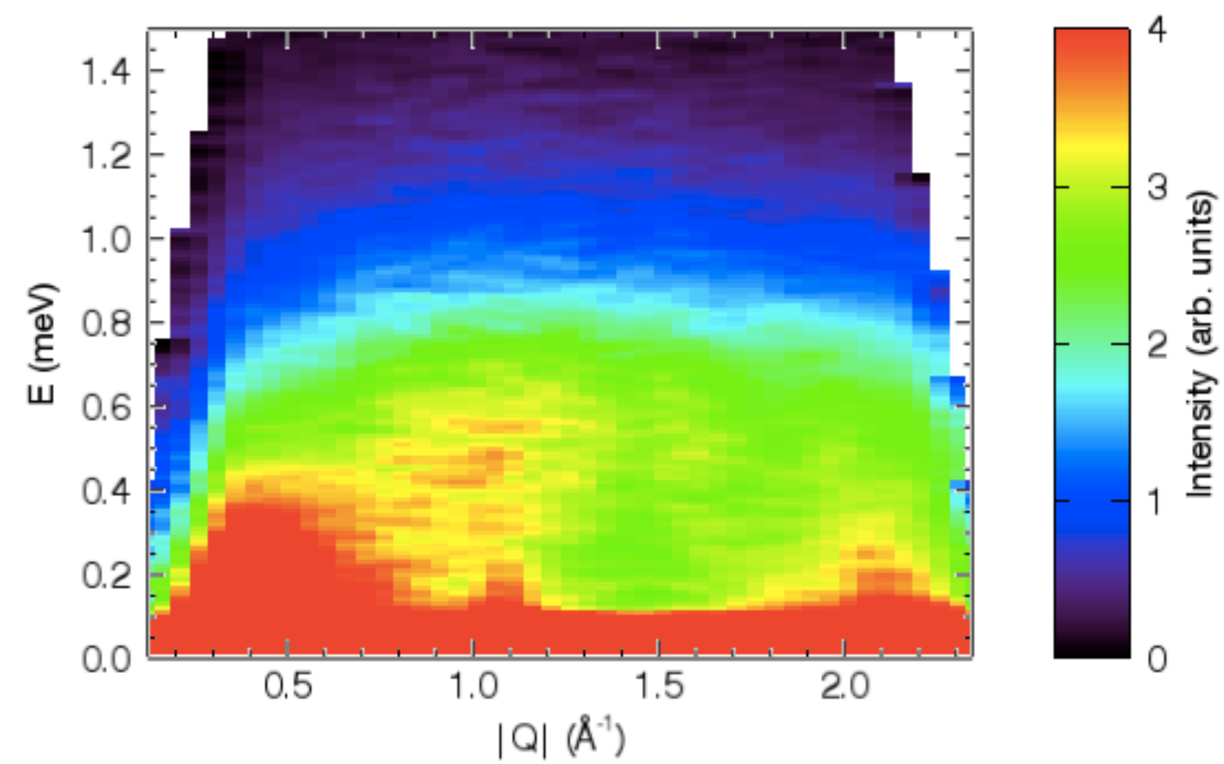


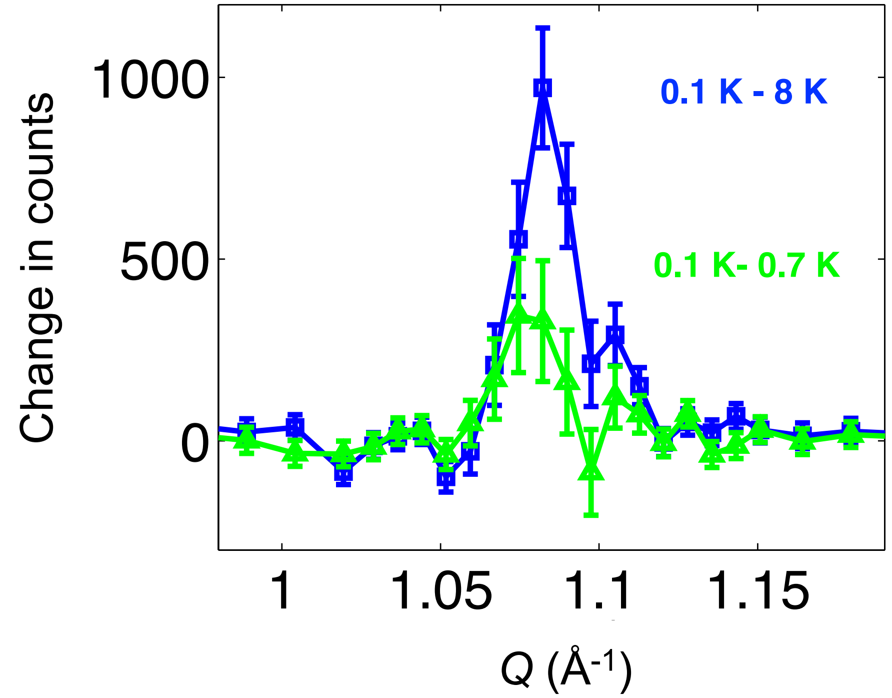
$Yb_2Ti_2O_7$ 100 mK



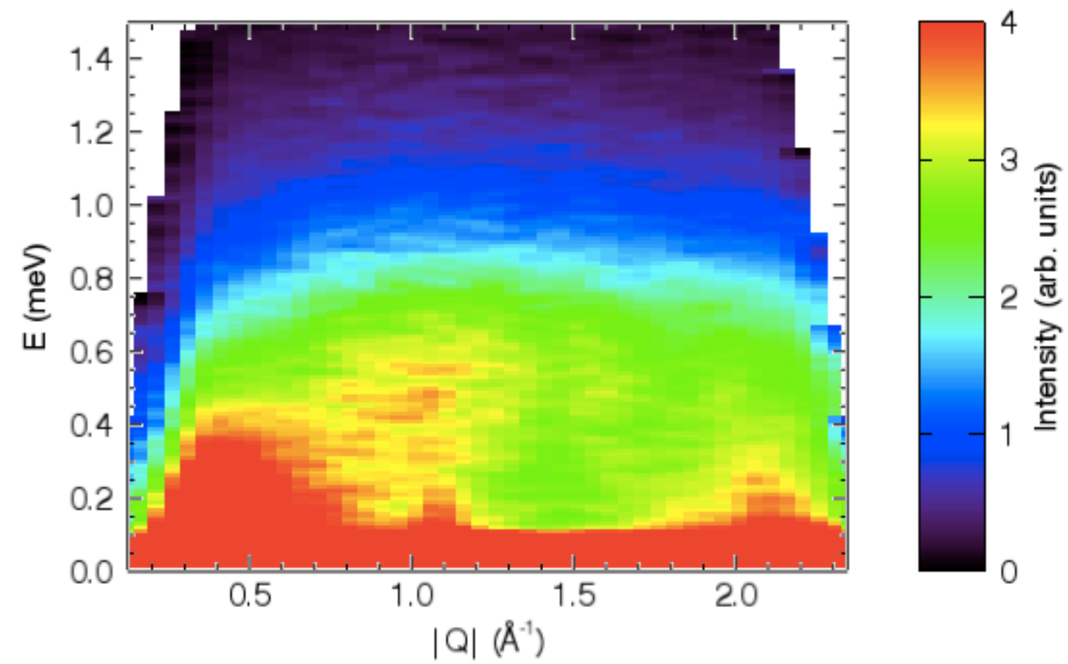
KITP
July 2015

Outline

- Anisotropic exchange in rare-earth pyrochlores
- "Stoichiometric" vs. "Stuffed" $Yb_2Ti_2O_7$
- Elastic and inelastic neutron scattering on stoichiometric $Yb_2Ti_2O_7$ powders
    - development of **50% long range ordered moment**
    - Unconventional, **gapless spin excitations**
- Comparison to excitations in crystals and $Yb_2Sn_2O_7$



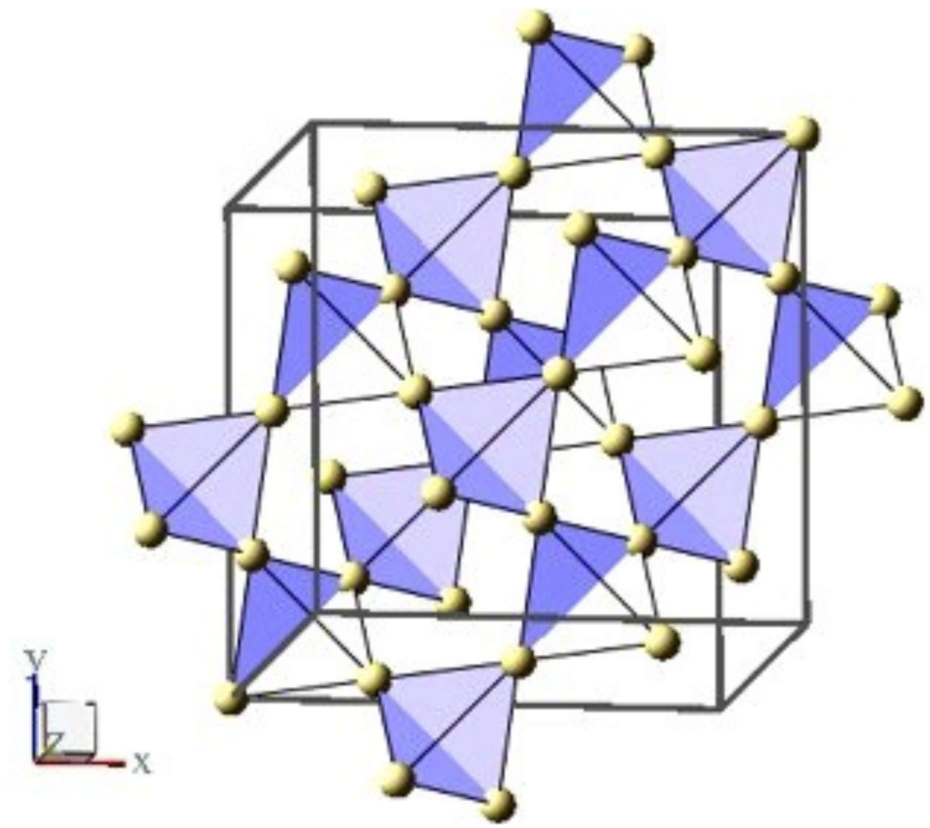
Yb2Ti2O7 100 mK

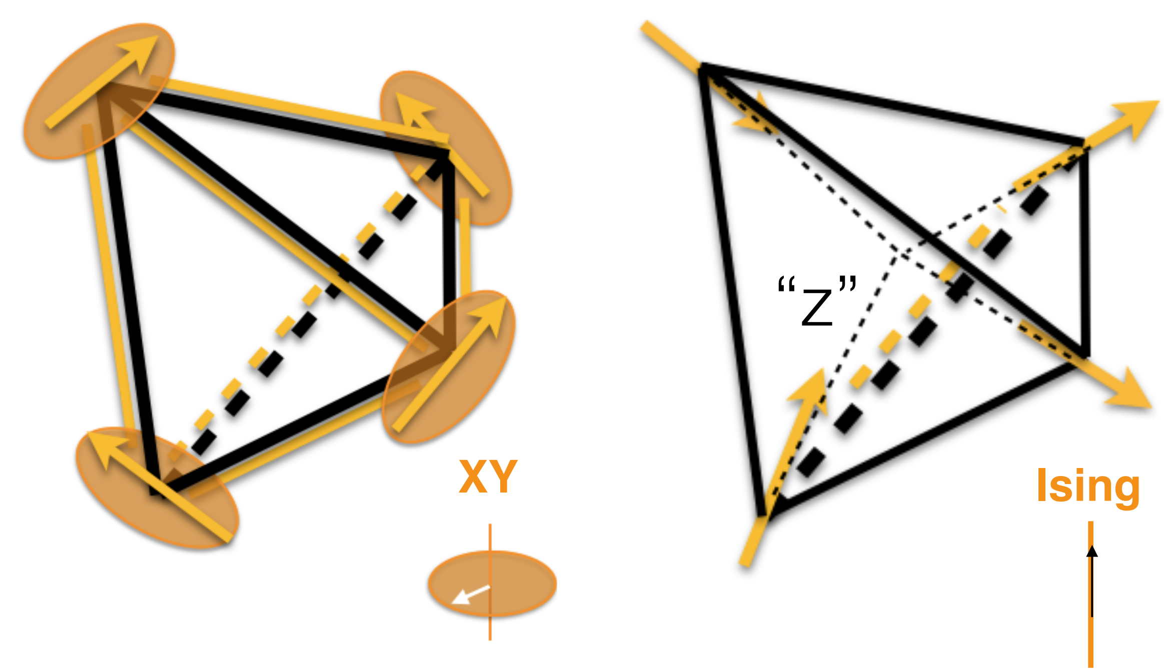


Anisotropic Exchange in Pyrochlores

$R_2Ti_2O_7$ (R = Yb^{3+}, Er^{3+}, Ho^{3+}, Dy^{3+} etc.)

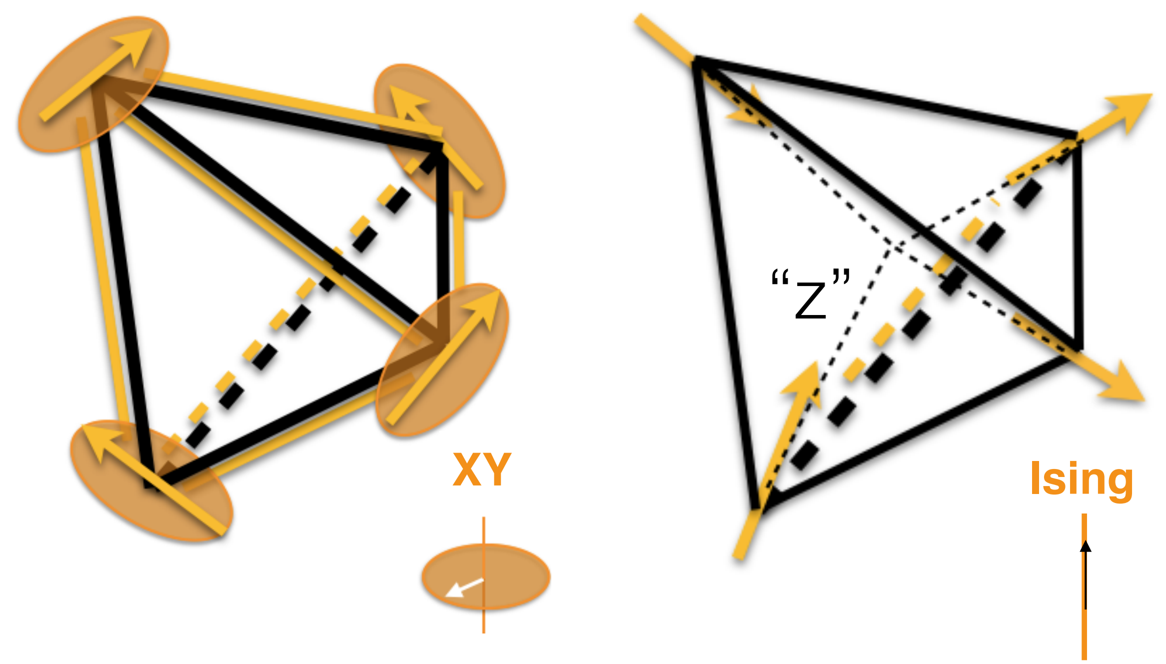
- Rare-Earth Oxides: dominant spin orbit coupling
- Moments described by total angular momentum, **J**
- low temperatures: anisotropic g-tensors and exchange interactions
- Can get anything from long range AFM order to spin ice depending on details





Anisotropic Exchange in Pyrochlores

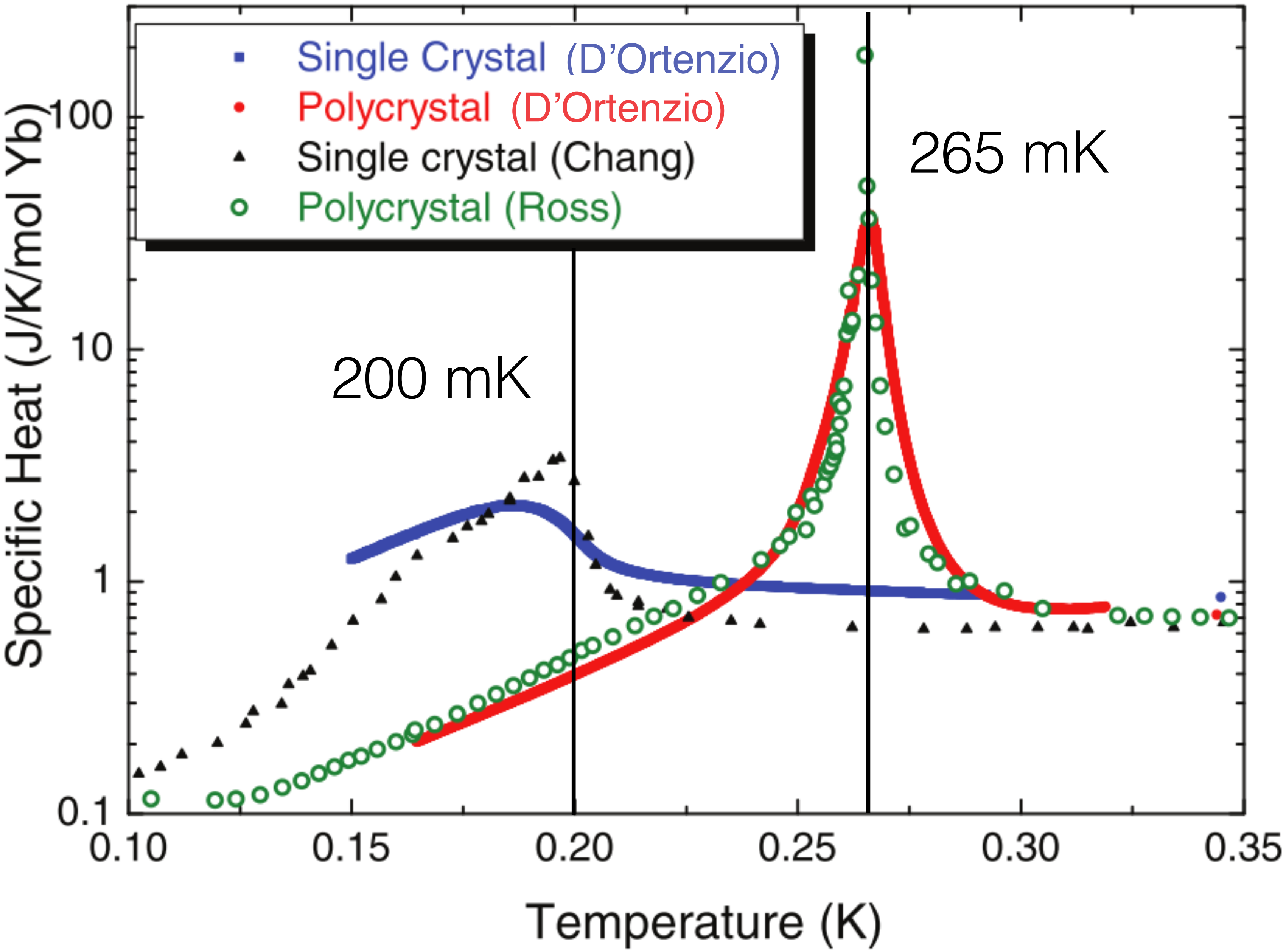
- Pyrochlore: 4 Symmetry-allowed exchange parameters (J_{zz}, $J_{\pm}$, $J_{\pm\pm}$, $J_{z\pm}$)
- **$Yb_2Ti_2O_7$: XY-like g-tensor, but strong exchange coupling with Ising components**



$$H = \sum_{\langle ij \rangle} \Big\{ J_{zz} \mathsf{S}_i^z \mathsf{S}_j^z - J_{\pm}(\mathsf{S}_i^+ \mathsf{S}_j^- + \mathsf{S}_i^- \mathsf{S}_j^+) + J_{++}\left[\gamma_{ij}\mathsf{S}_i^+\mathsf{S}_j^+ + \gamma_{ij}^*\mathsf{S}_i^-\mathsf{S}_j^-\right] + J_{z\pm}\left[\mathsf{S}_i^z(\zeta_{ij}\mathsf{S}_j^+ + \zeta_{ij}^*\mathsf{S}_j^-) + i \leftrightarrow j\right] \Big\}$$

K.A. Ross, L. Savary, B. D. Gaulin, and L. Balents, Phys. Rev. X **1**, 021002 (2011)

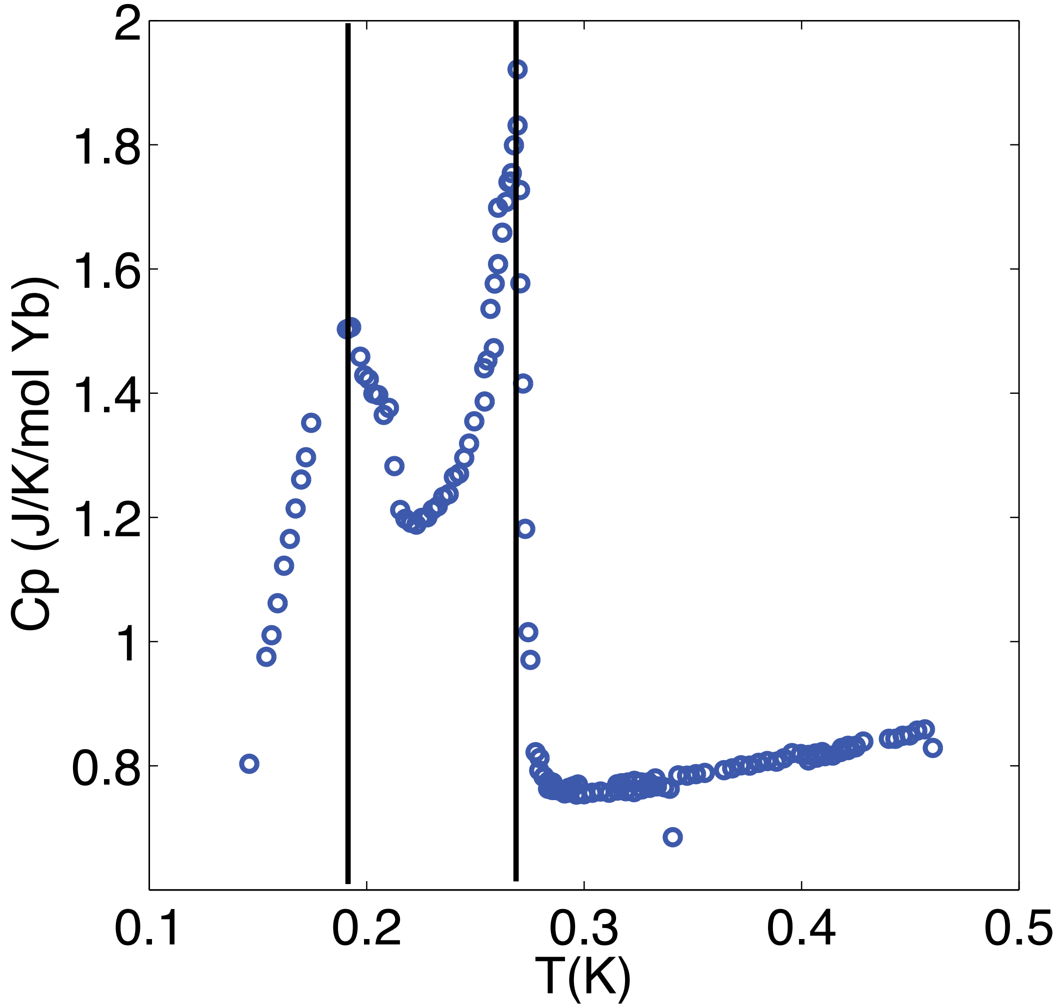
$Yb_2Ti_2O_7$ Specific Heat

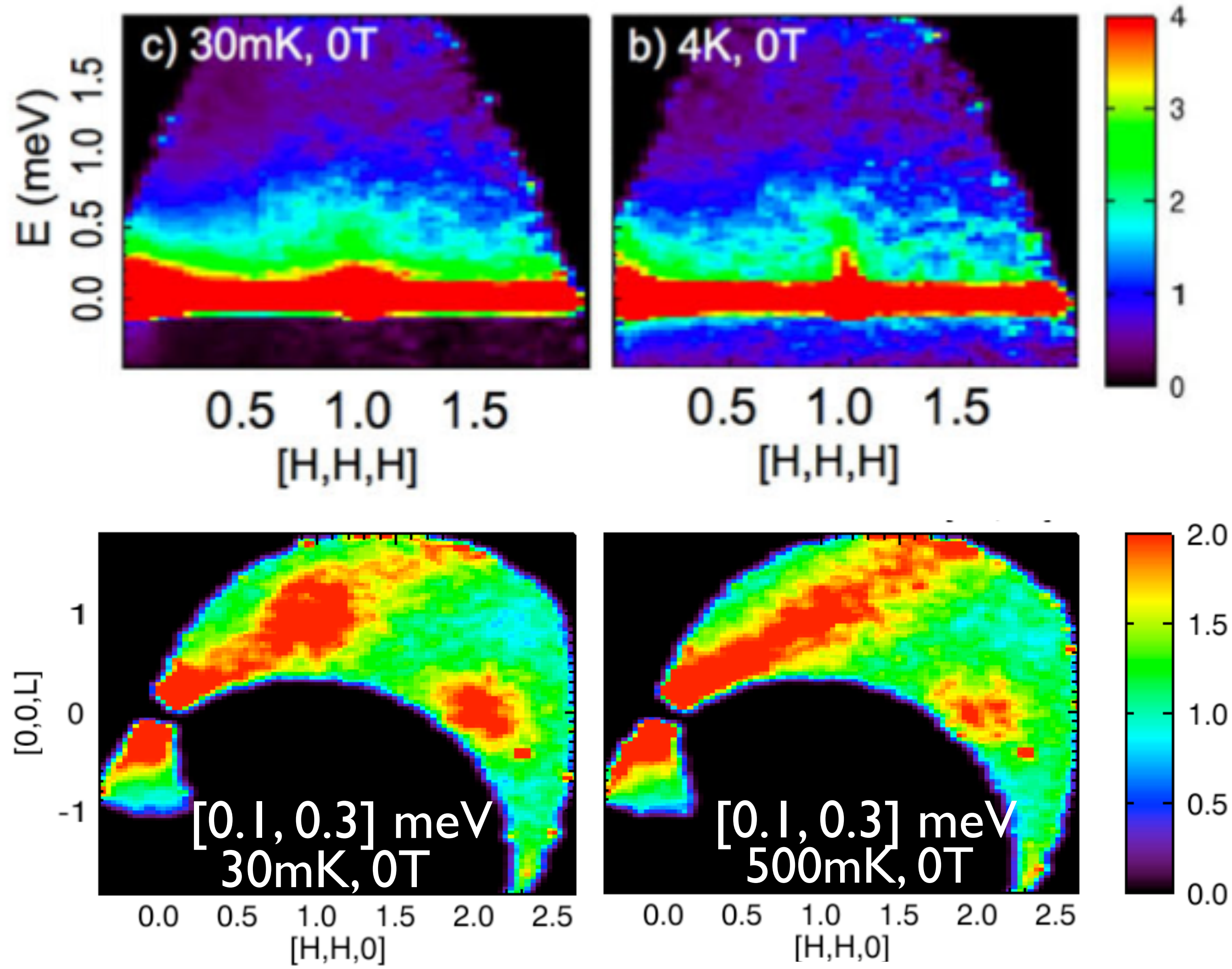


D'Ortenzio et al, PRB **88**, 134428 (2013)

- **Powder samples** usually have sharper, higher temperature anomalies
- Some samples show signs of ferromagnetic order below T_C, others do not
  - Ordered moment size quoted from 0.8 to 1.1 μB **(47 - 64% of total moment)**
- Evidence for hysteresis at the sharper transitions: **first order**

Some single crystals do not order

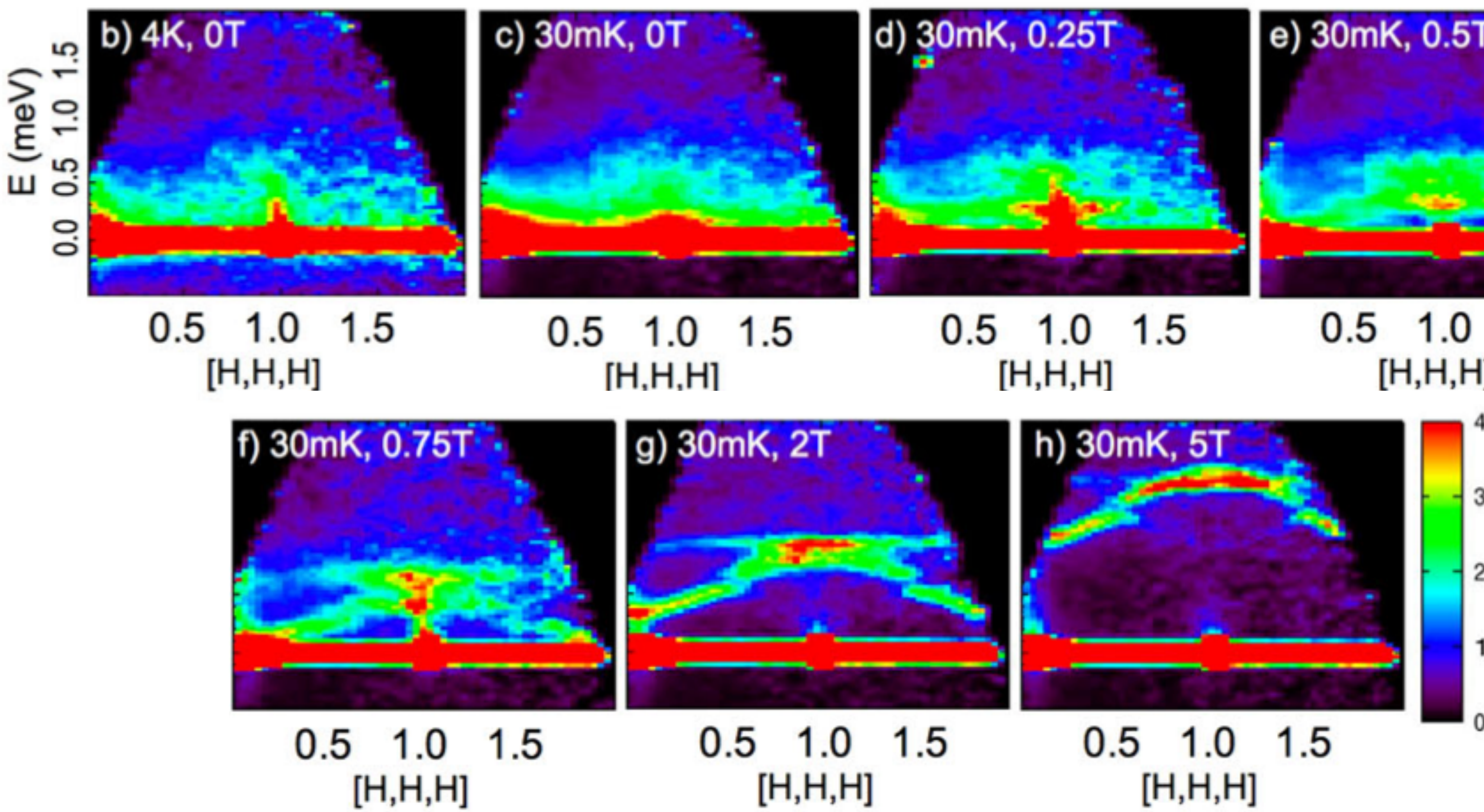




K. A. Ross et al, PRL **103** 227202 (2009)

- Diffuse continuum-like scattering at low temperatures and low fields
- No sharp magnetic Bragg scattering below Tc

Evolution of S(Q,ω) in **Single Crystals**

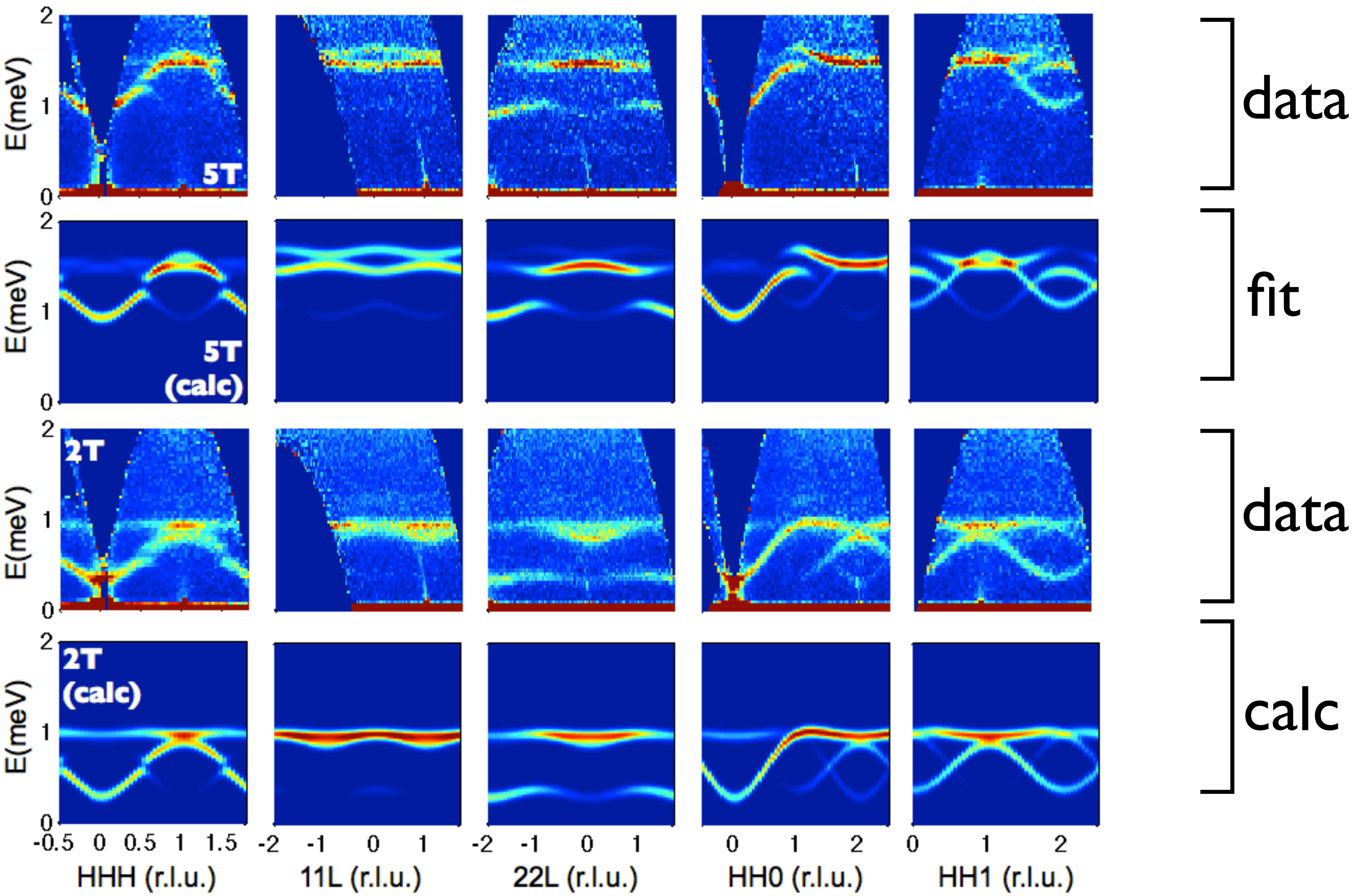


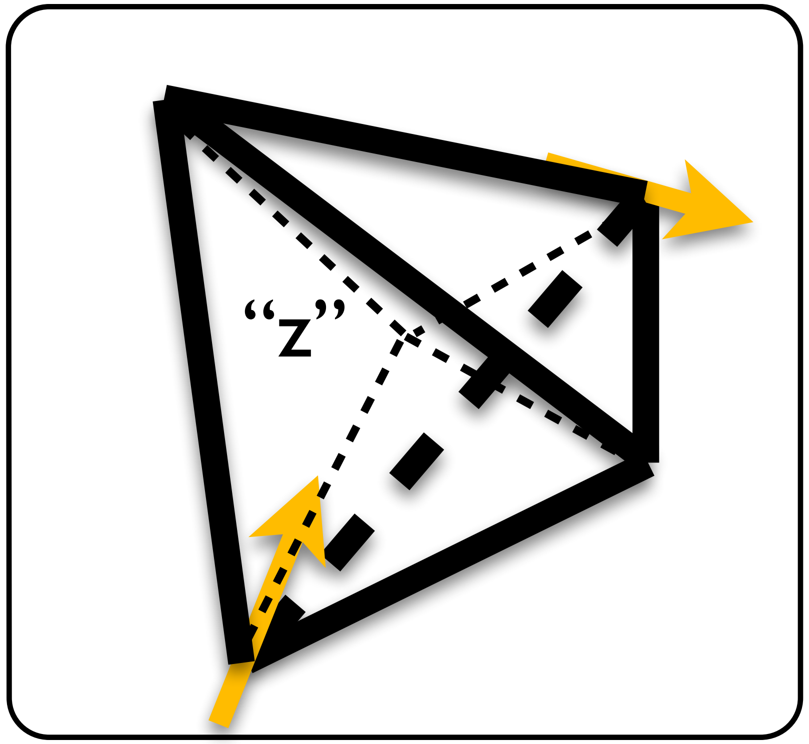
Broad scattering develops into sharp magnons with increasing field (H || [1,-1,0])

K. A. Ross et al, PRL **103** 227202 (2009)

"Quantum Spin Ice" Exchange Parameters for $Yb_2Ti_2O_7$

H along $[1\bar{1}0]$ $g_z = 1.8$, $g_{xy} = 4.32$





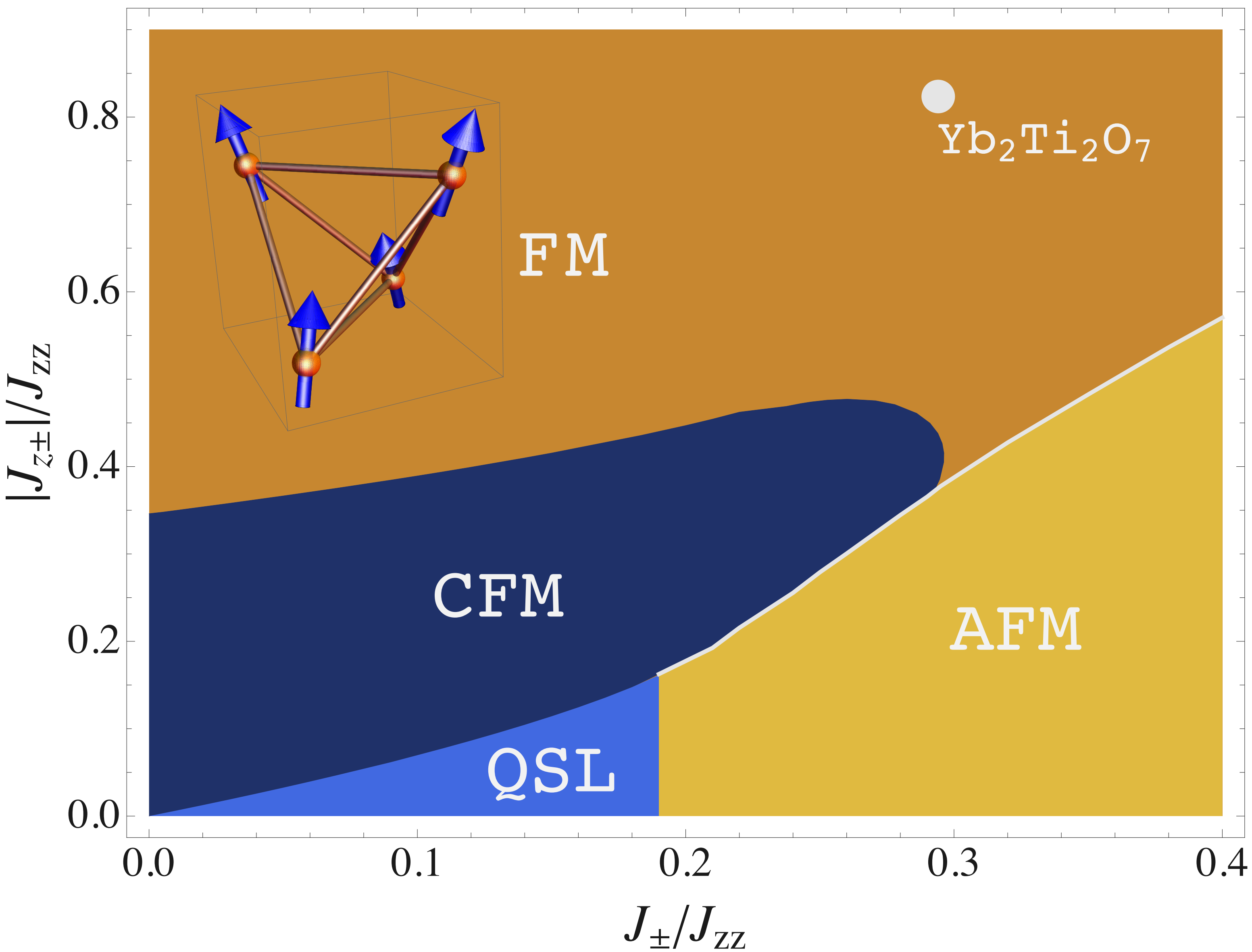
Spin Ice plus
quantum fluctuations

$$J_{zz} = 0.17 \pm 0.04, \; J_{\pm} = 0.05 \pm 0.01, \; J_{\pm\pm} = 0.05 \pm 0.01, \; J_{z\pm} = -0.14 \pm 0.01 \quad \text{(meV)}$$

K.A. Ross, L. Savary, B. D. Gaulin, and L. Balents, Phys. Rev. X **1**, 021002 (2011)

What is the ground state?

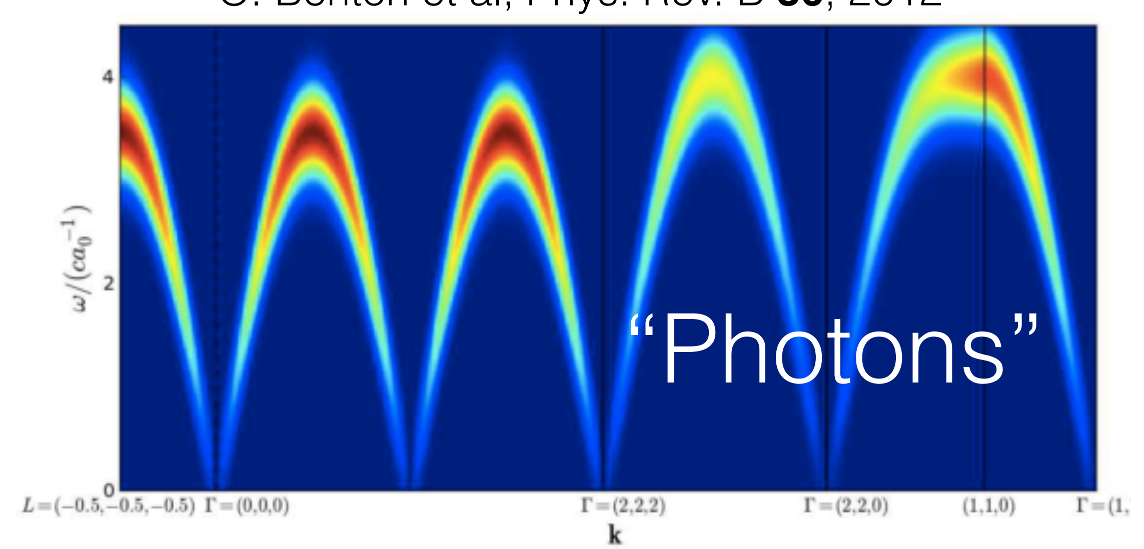
Exchange parameters for $Yb_2Ti_2O_7$ compared to "Gauge Mean Field" phase diagram



L. Savary, L. Balents, Phys. Rev. Lett. 108, 037202 (2012)

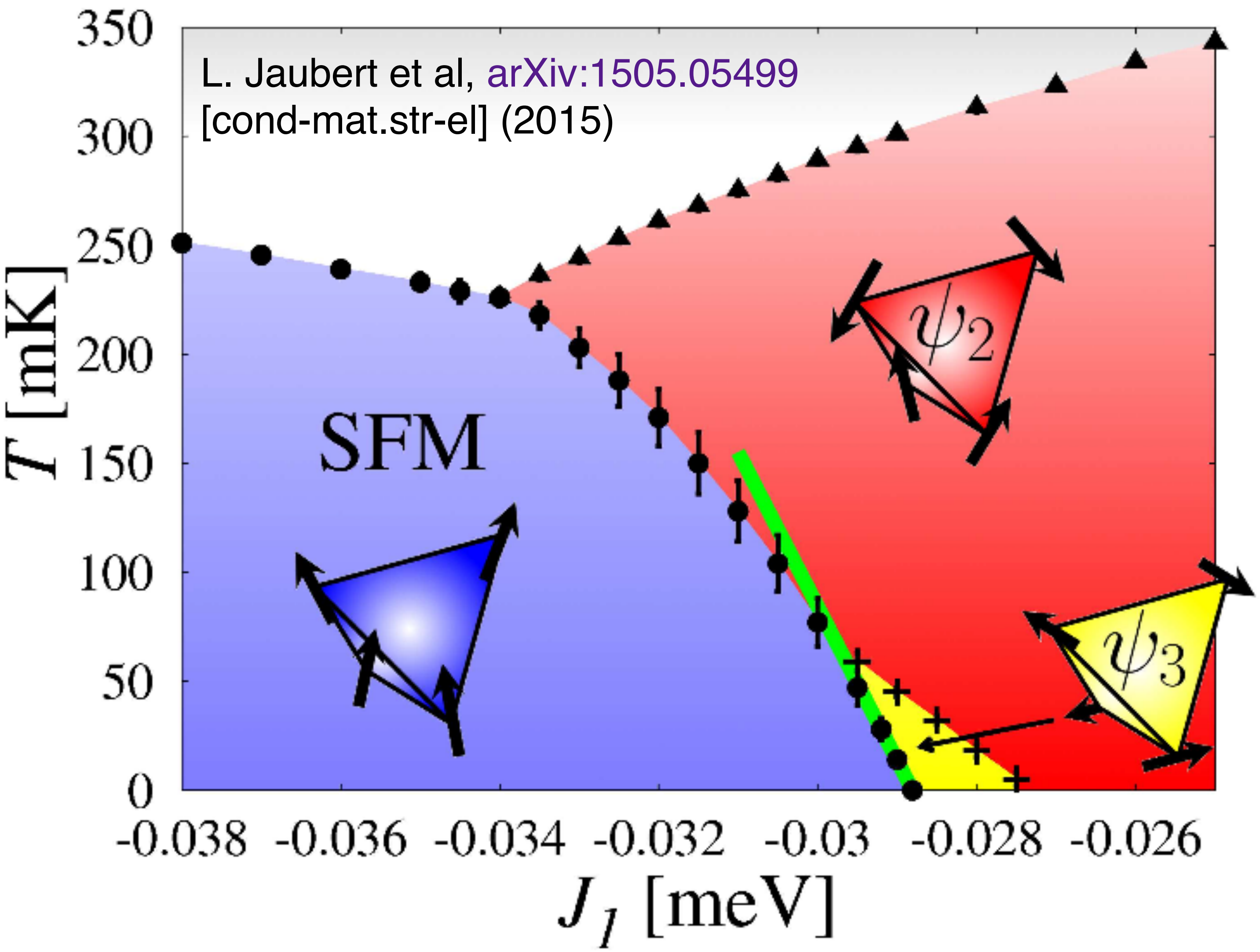
- $Yb_2Ti_2O_7$ "close to" the boundary with exotic phases of matter
- **Quantum Spin Liquid** (QSL)
  - supports emergent electrodynamics
  - Monopoles, Photons, Electrons
- **Coulomb Ferromagnet** (CFM)
  - **partially polarized phase**
  - supports emergent electrodynamics

O. Benton et al, Phys. Rev. B **86**, 2012



What is the ground state?

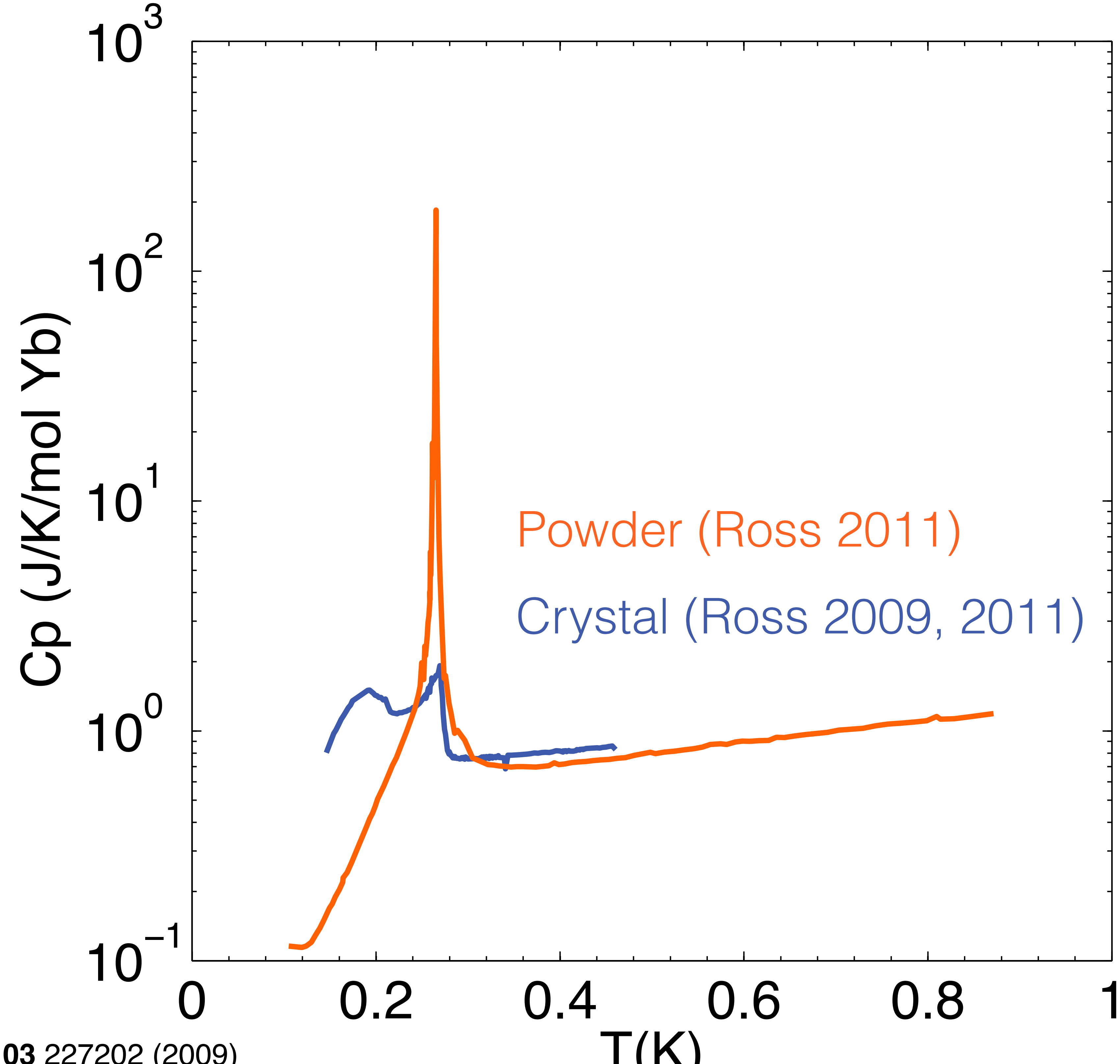
$Yb_2Ti_2O_7$ also lies “close” to a phase boundary between AFM and FM states



- New proposed parameters from other groups[1,2] suggest $Yb_2Ti_2O_7$ is right on the edge of AFM order
- Do quantum fluctuations arise from proximity to AFM state?
- **What role does the known sample dependence play?**

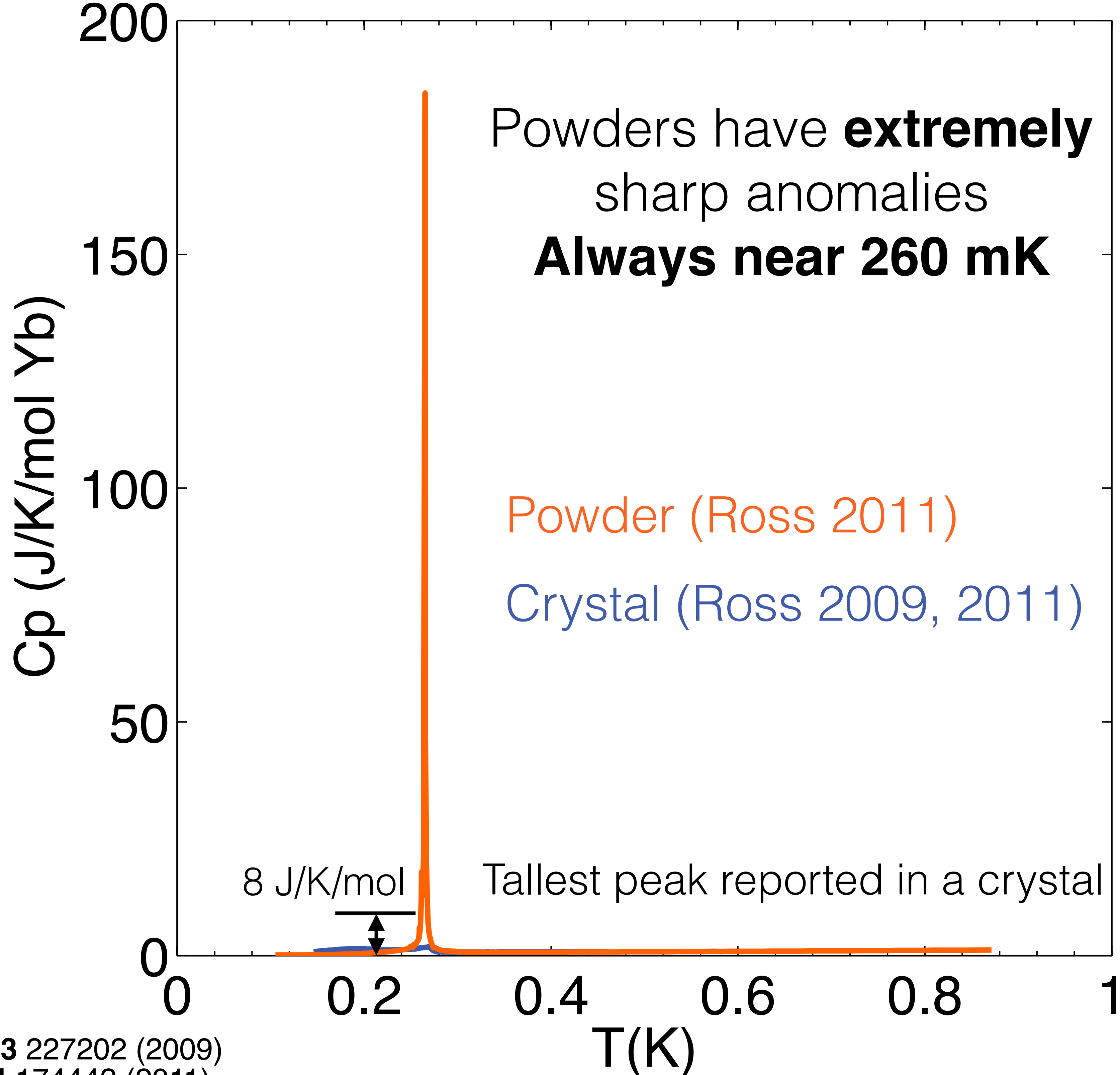
[1] J. Robert, arXiv:1506.01729 [cond-mat.str-el] (2015)
[2] R. Coldea, KITP talk (http://online.kitp.ucsb.edu/online/lsmatter15/coldea/)

Powder vs.crystal: Log scale



K. A. Ross et al, PRL **103** 227202 (2009)
K. A. Ross et al, PRB **84** 174442 (2011)

Powder vs.crystal: Linear scale

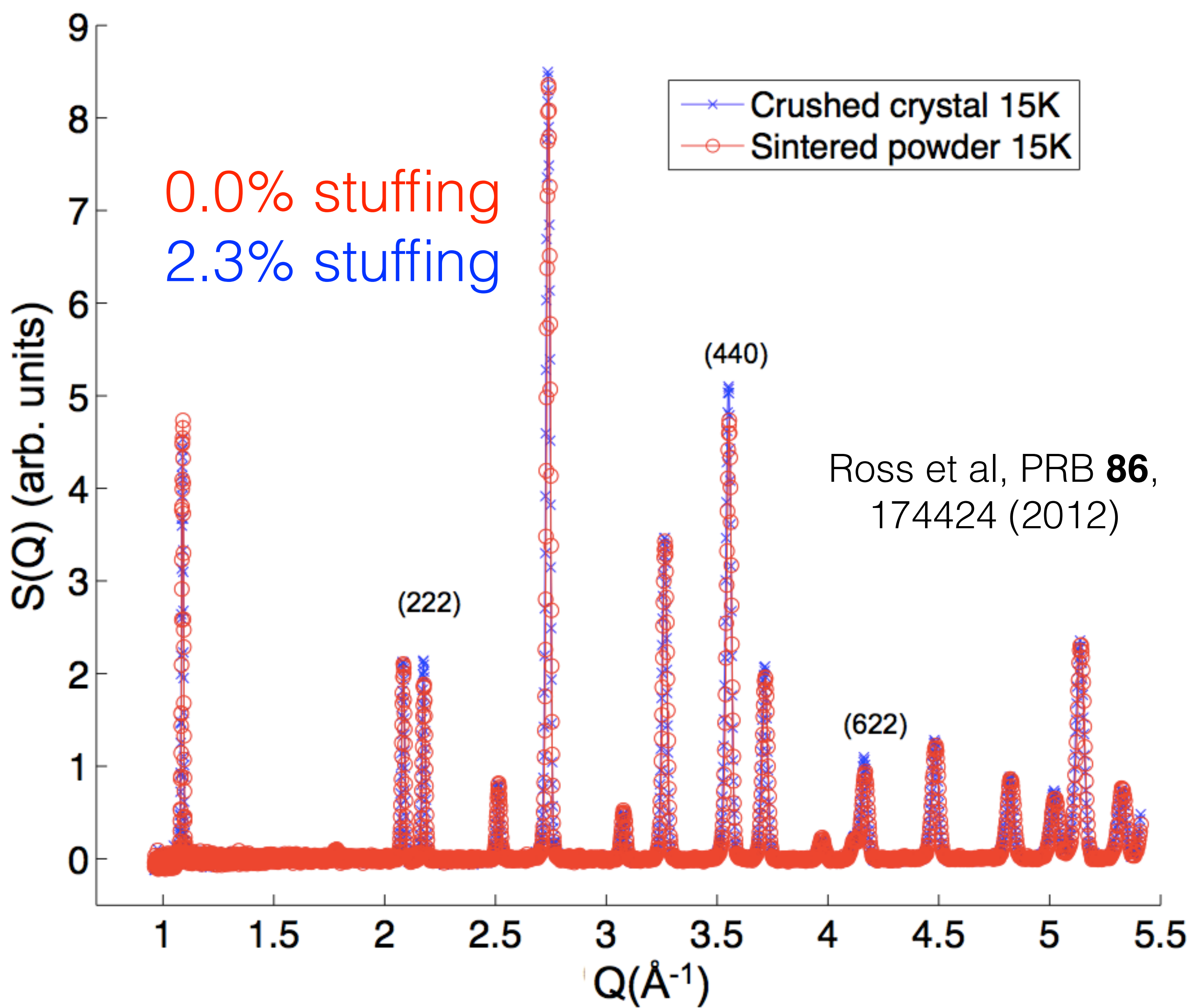


K. A. Ross et al, PRL **103** 227202 (2009)
K. A. Ross et al, PRB **84** 174442 (2011)

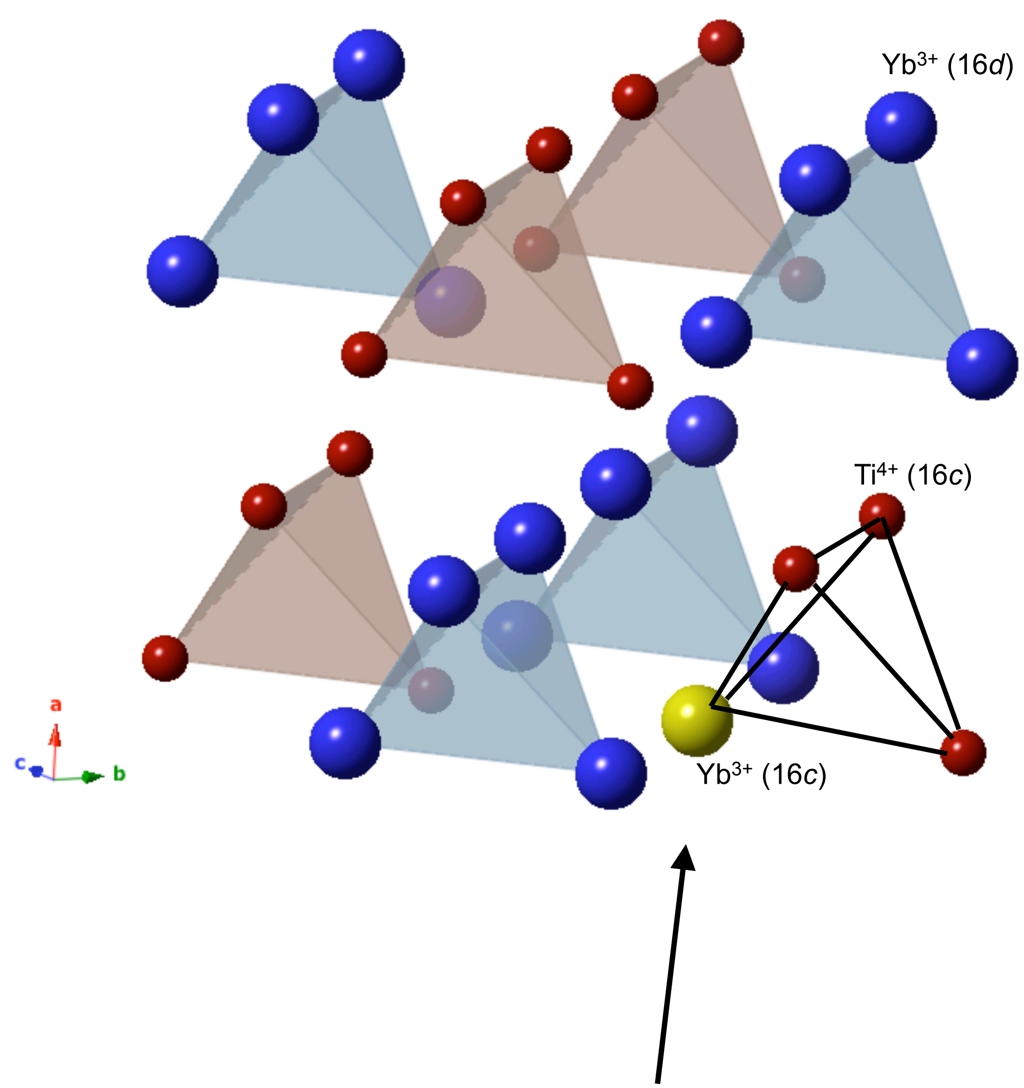
Crushed Crystal vs. Sintered Powder

Structural difference between crushed crystal and sintered powder can be attributed to “stuffing”

2.3% excess Yb^{3+} on Ti^{4+} sublattice

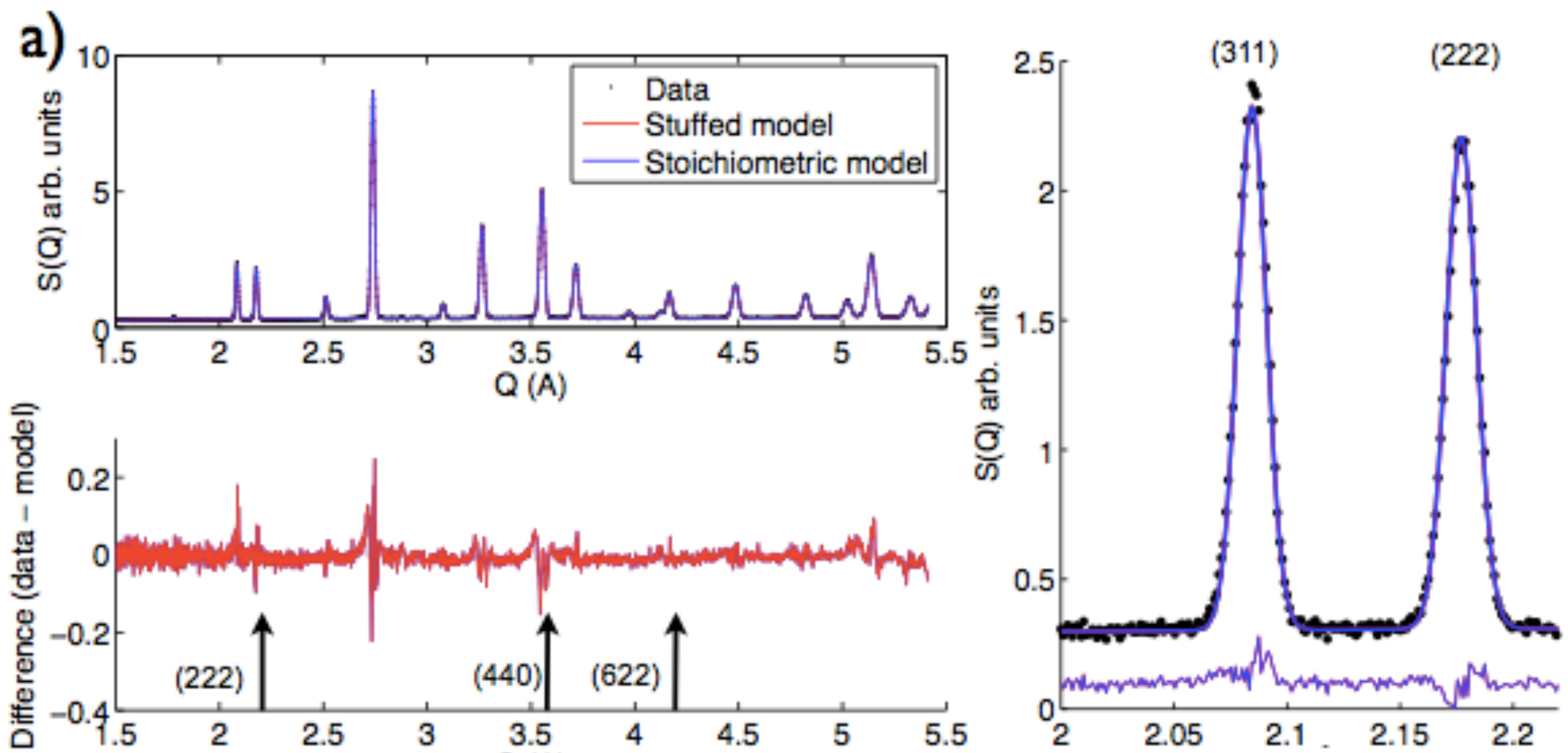


Ross et al, PRB **86**, 174424 (2012)

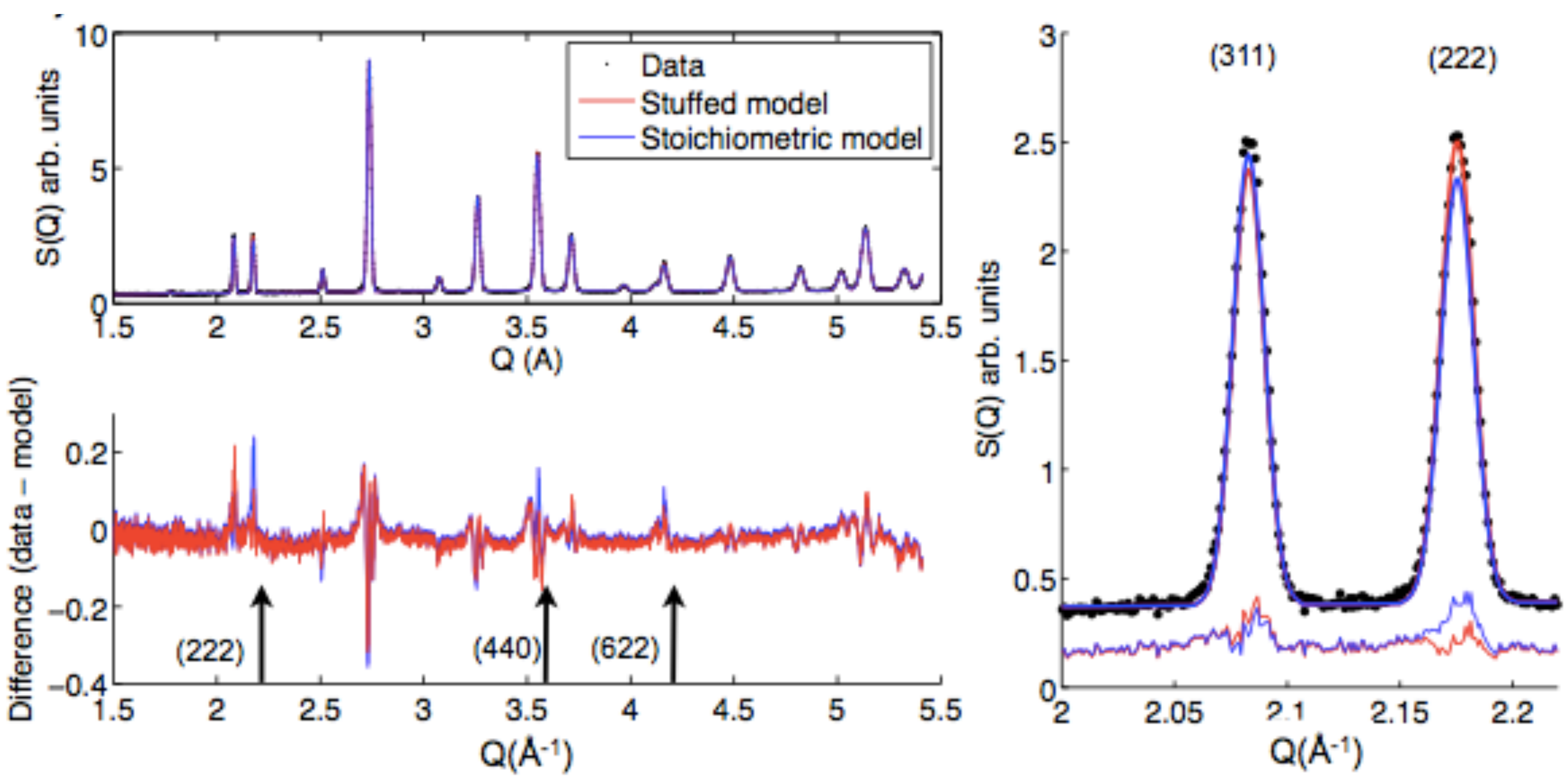


Stuffed Yb^{3+} in crushed crystal

Powder Diffraction data at 15 K



Stoichiometric powder

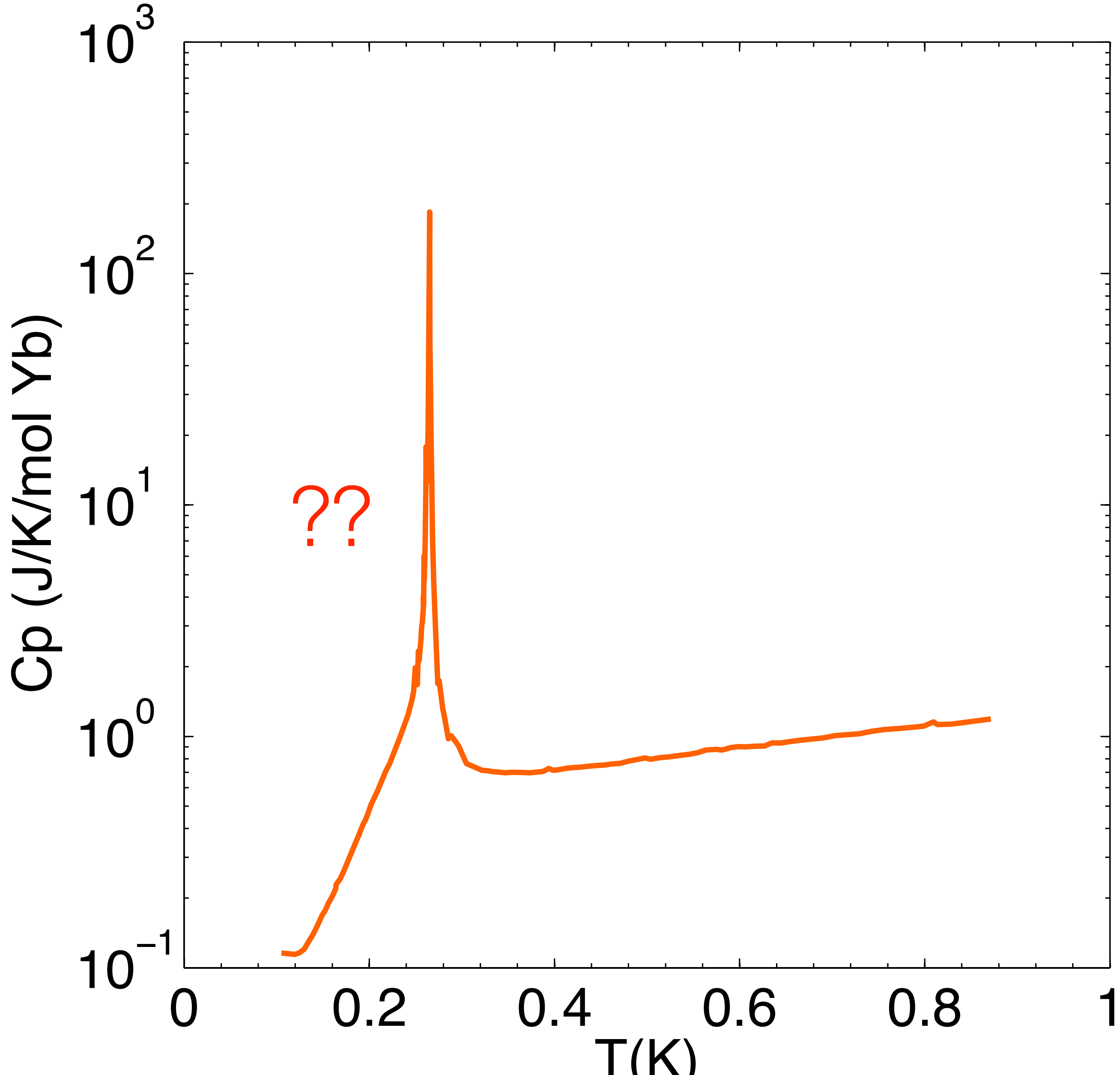


Crushed Crystal
2.3% stuffing

Ross et al, PRB **86**, 174424 (2012)

Crushed Crystal vs. Sintered Powder

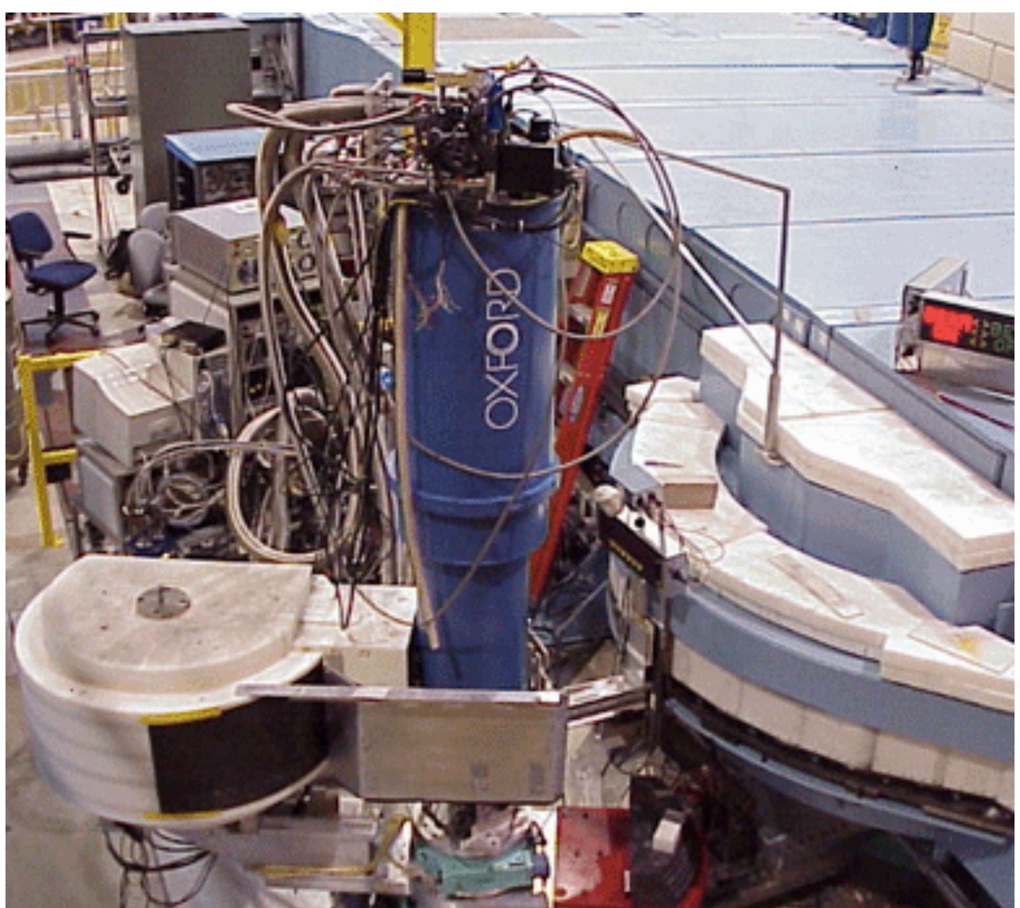
What are the ground state magnetic properties of the stoichiometric powder?



Neutron Scattering Measurements at the NCNR

10 grams of the **previously studied stoichiometric powder sample**, sealed with 10 atm helium gas at room temperature

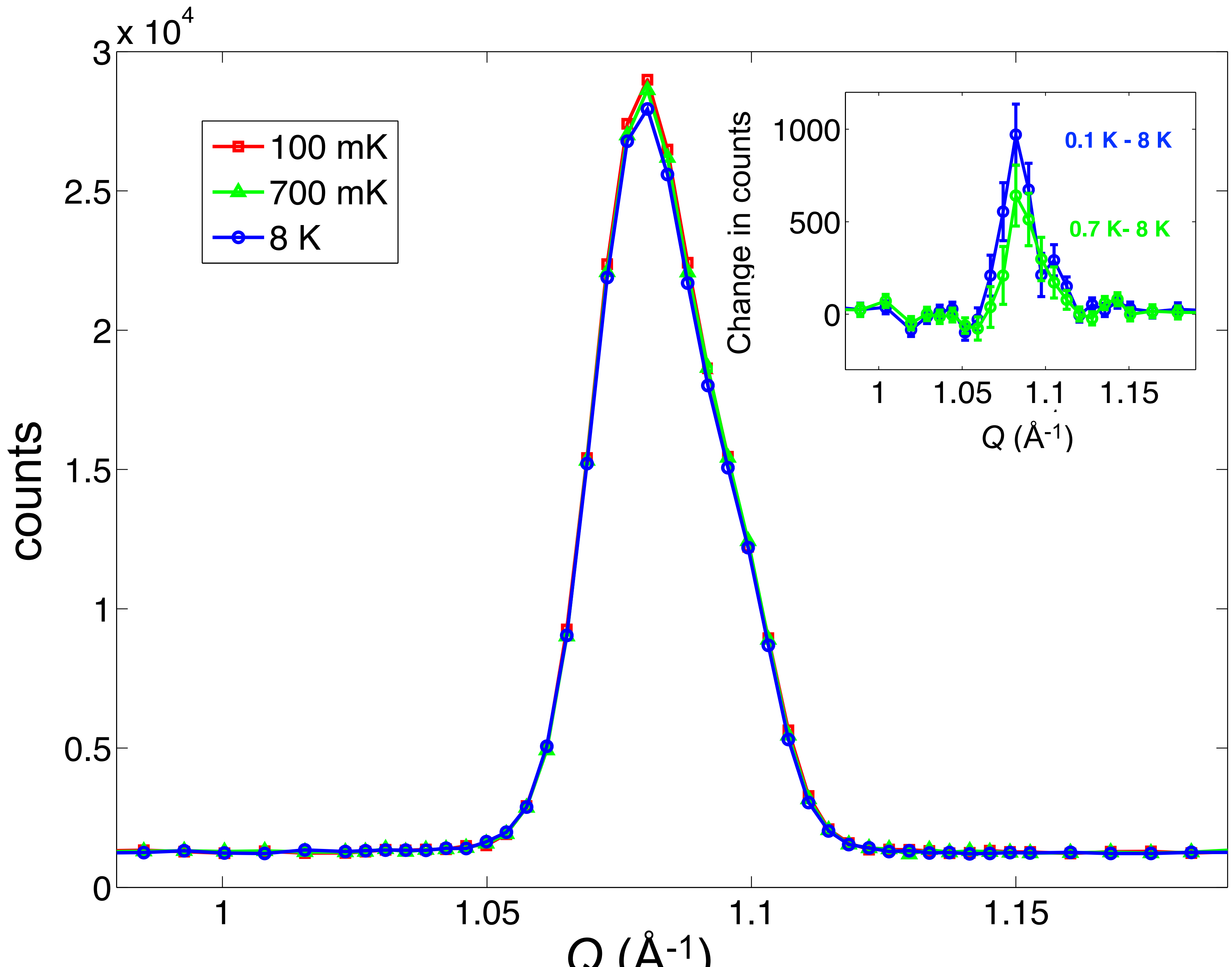
SPINS Triple Axis Spectrometer (Elastic)



Disk Chopper Spectrometer (inelastic)

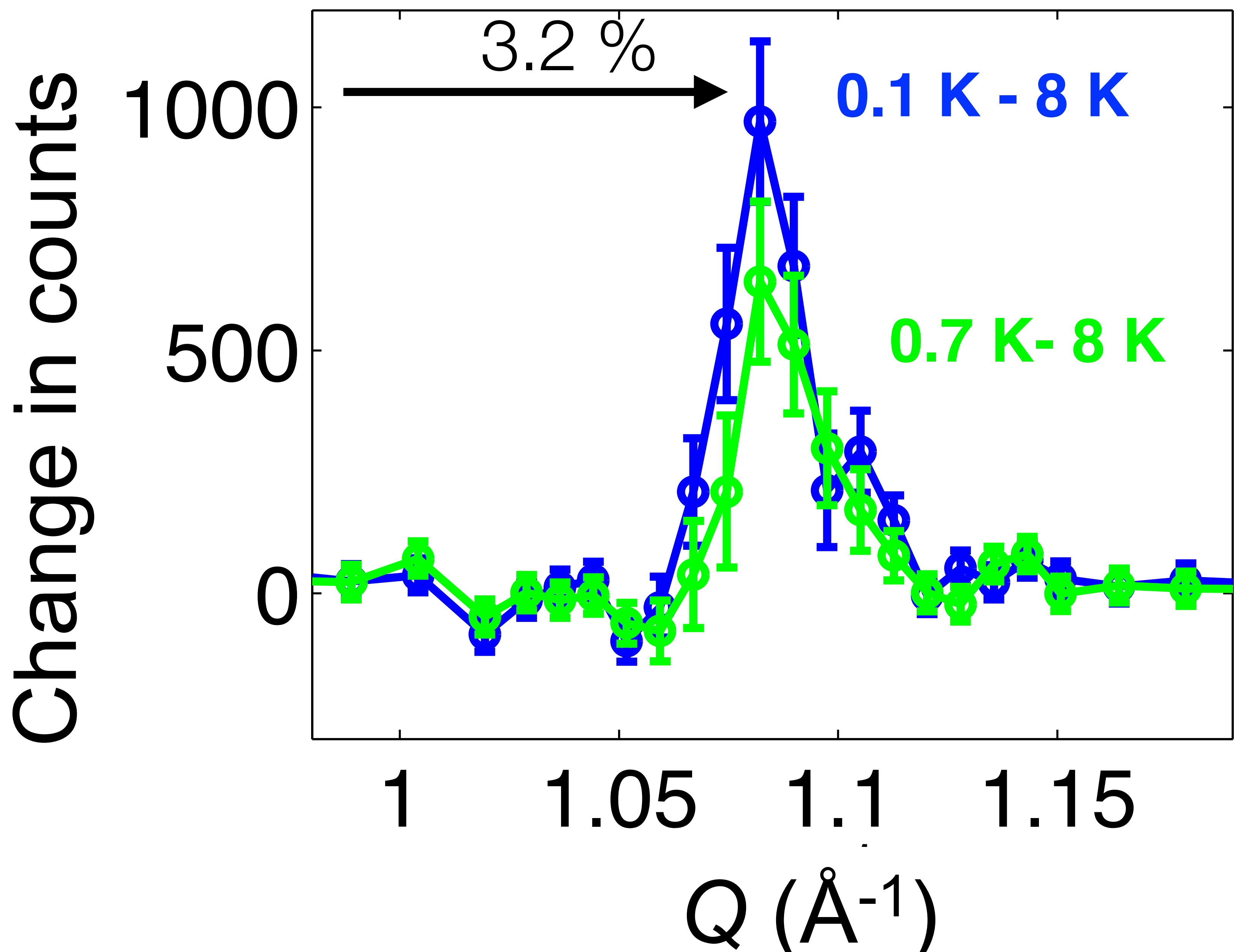
“Elastic” Scattering in a **Stoichiometric Powder**

- Elastic scattering, $E = 0.0 \pm 0.25$ meV, at (111) Bragg Peak
- Increase of intensity **visible in raw data**

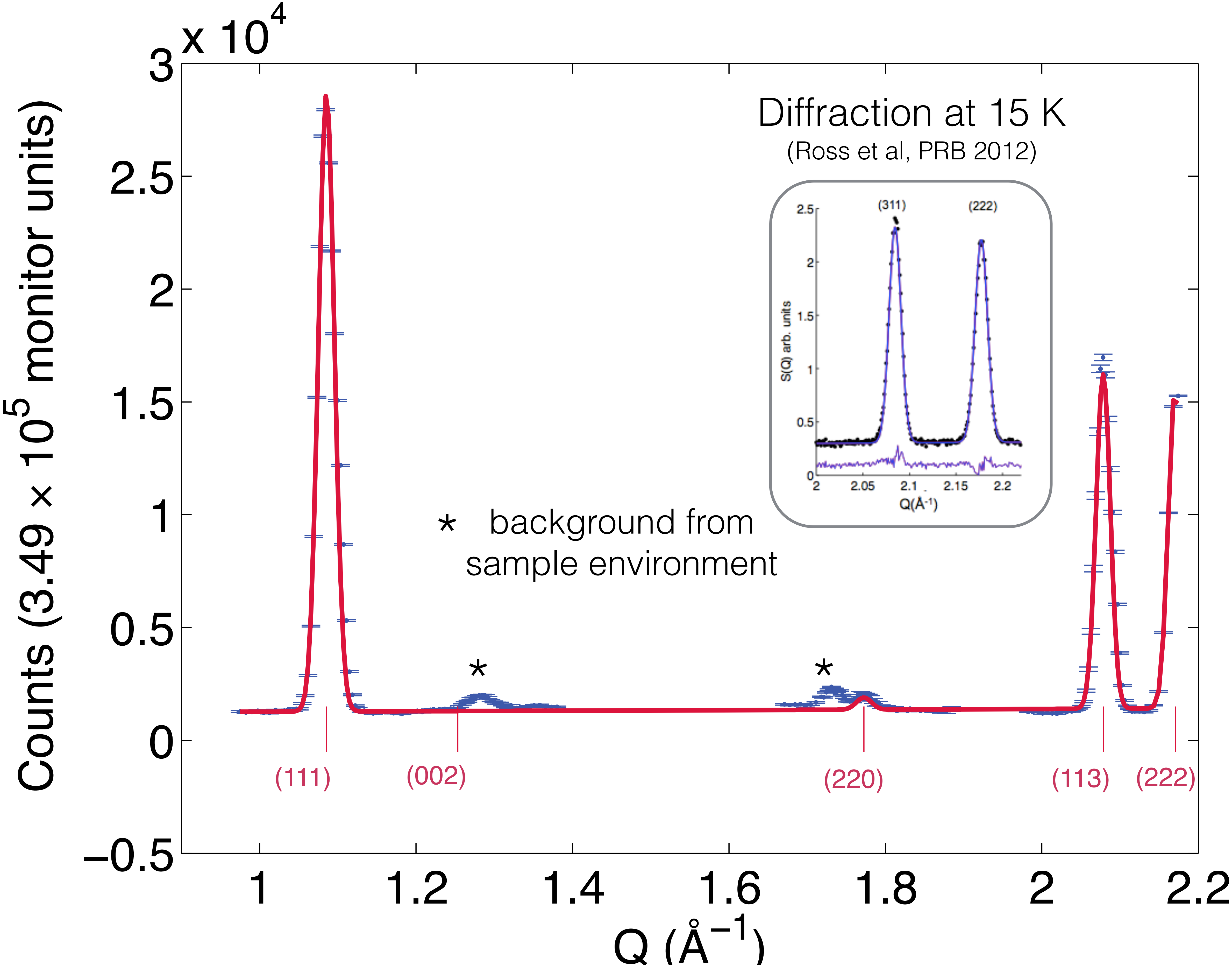


Stoichiometric Powder of $Yb_2Ti_2O_7$

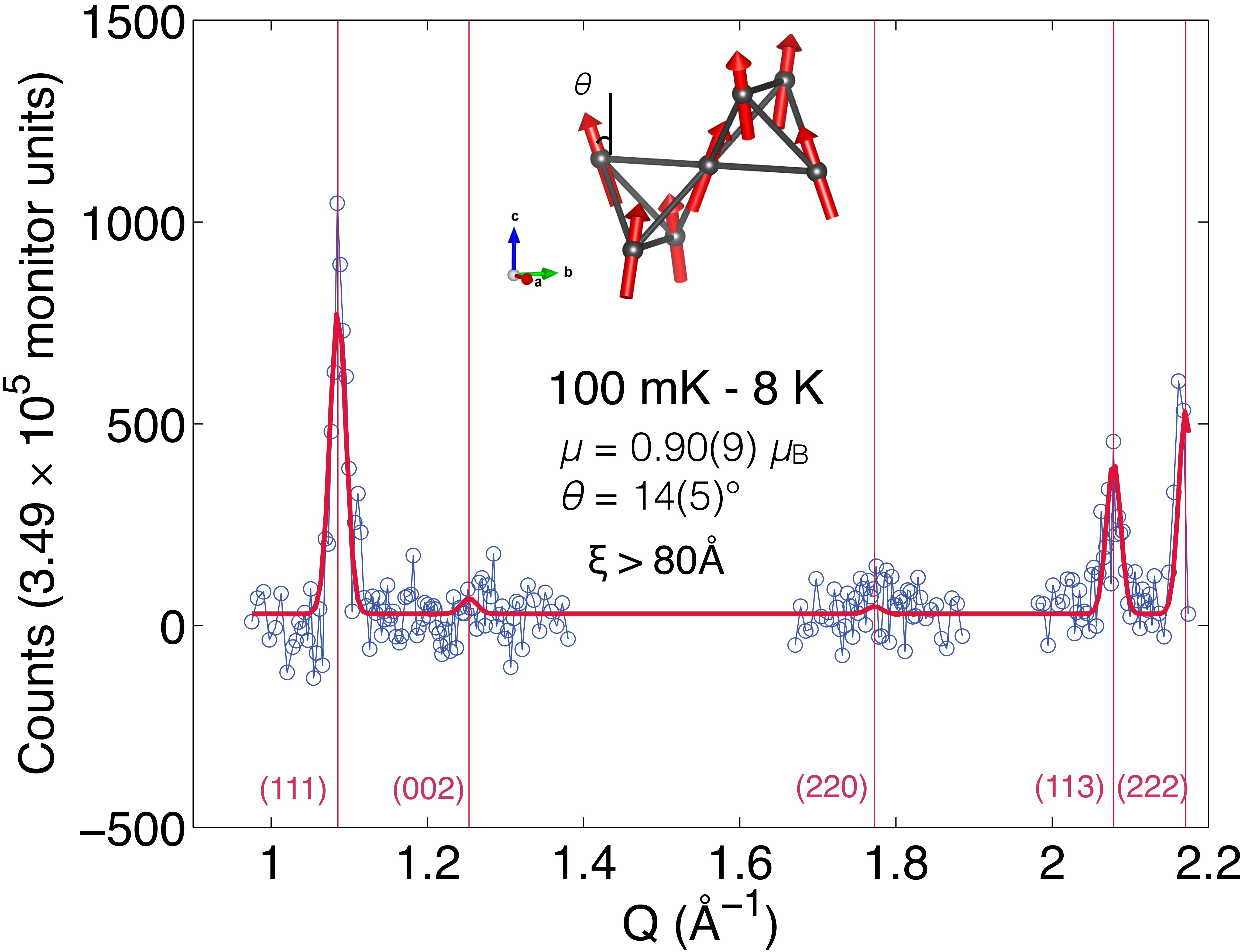
- (111) peak: 3.2 % increase from 8 K to 100 mK
- similarly, 3.3(5) % increase on (113) peak, 3.6(6)% increase on (222)



Fit to Stoichiometric Model at T= 8K



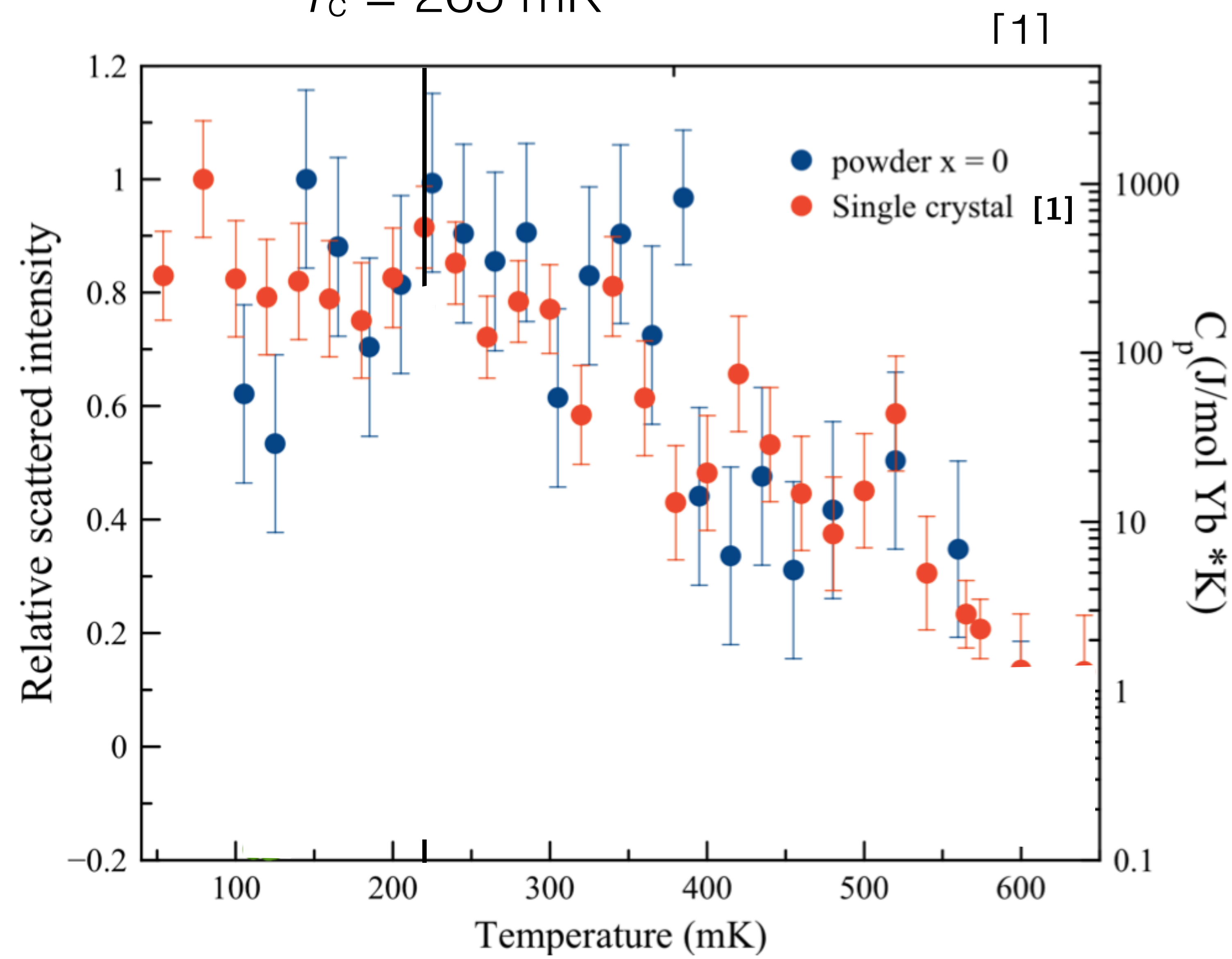
Resolution-Limited Elastic Scattering (100mK - 8K)



Temperature dependence of (111)

- **gradual decline of intensity** at (111) on warming (1 mK / min)
- Same temperature dependence as the diffuse scattering near (111) in our crystal
- No sharp changes at T_C

T_C = 265 mK

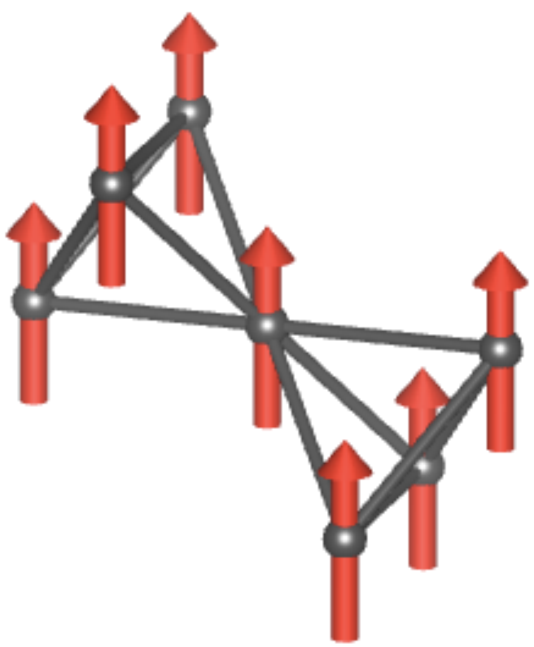


[1] K. Ross et al, PRB 84, 174442 (2011)

Splayed Ferromagnetism

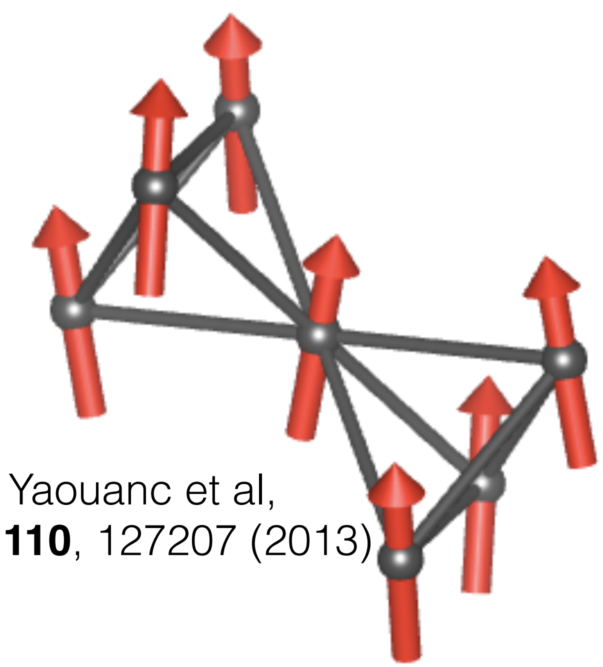
$\theta = 0$

Collinear Ferromagnet



$\theta < 0$

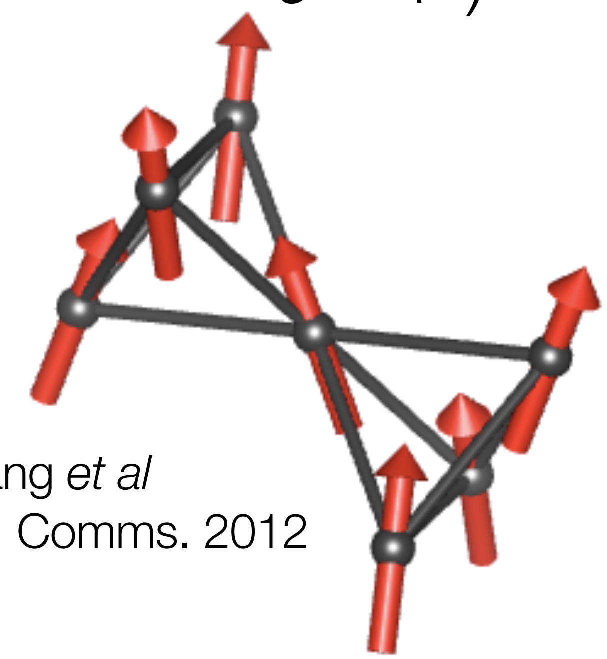
Splayed "XY" Ferromagnet
(i.e. $Yb_2Sn_2O_7$, θ = -10°)



Yaouanc et al,
PRL **110**, 127207 (2013)

$\theta > 0$

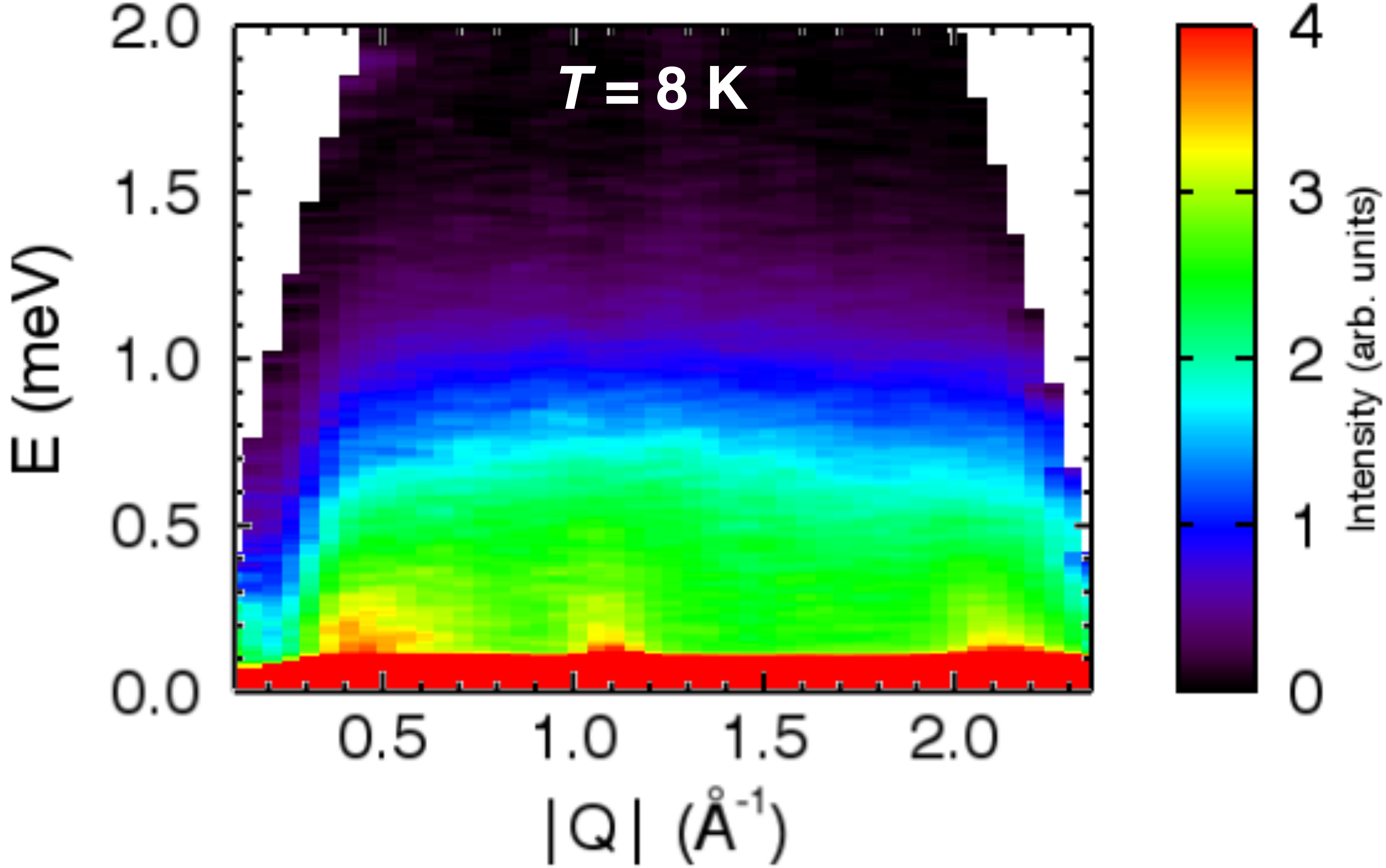
Splayed "Ice" Ferromagnet
(i.e. some xtals of $Yb_2Ti_2O_7$,
θ = 1°)



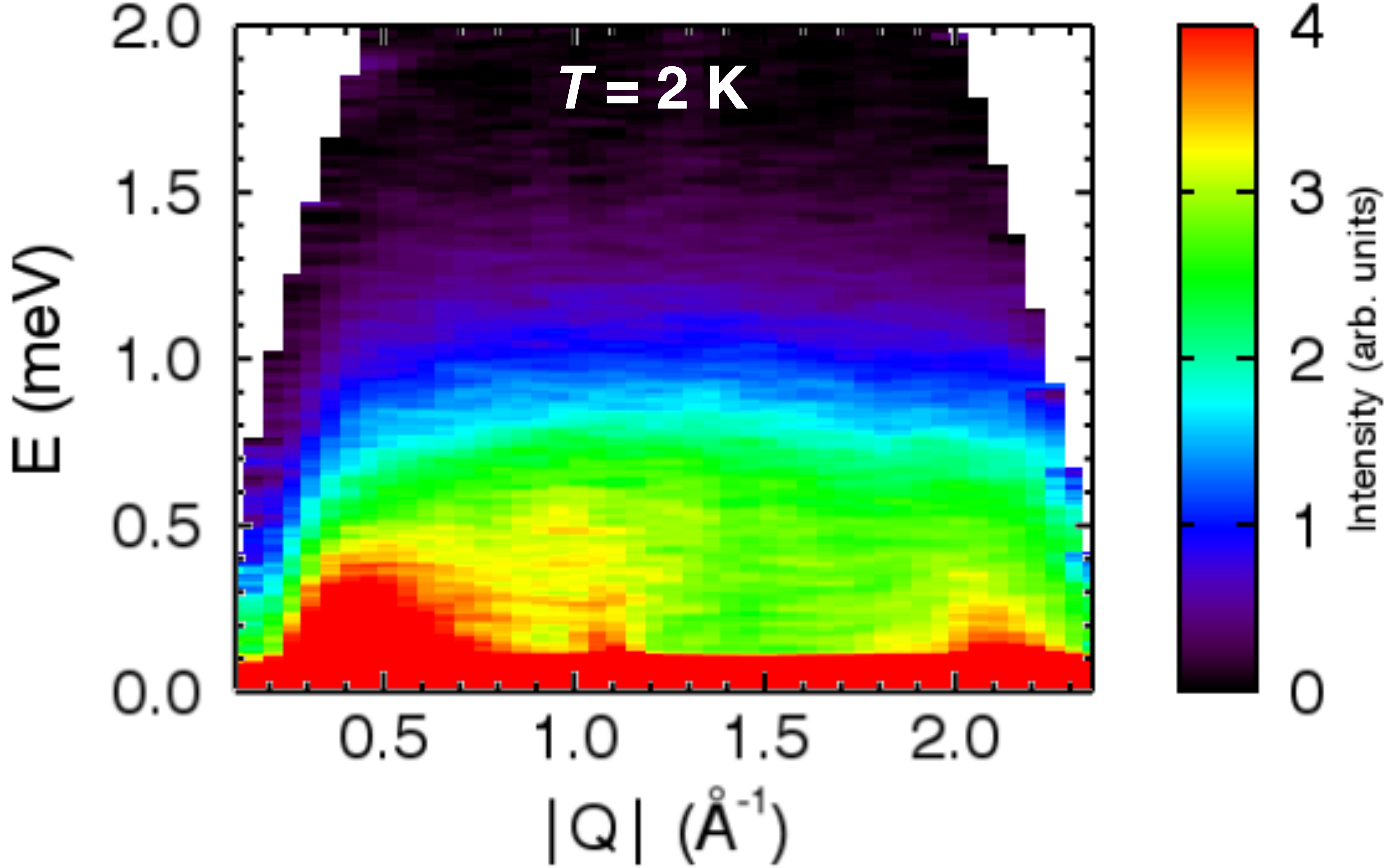
Chang *et al*
Nat. Comms. 2012

- Stoichiometric powder shows splayed "ice" ferromagnetism
- **0.90(6) μ_B** at 100 mK (compare to μ_{sat} = 1.7 μB, 53% ordered)
- **Large splaying angle of the moments**, **14°** (compare to spin ice, 54°)
- **No sharp onset at transition**

Temperature Dependence of S(Q,ω)



Temperature Dependence of $Yb_2Ti_2O_7$ Spectrum



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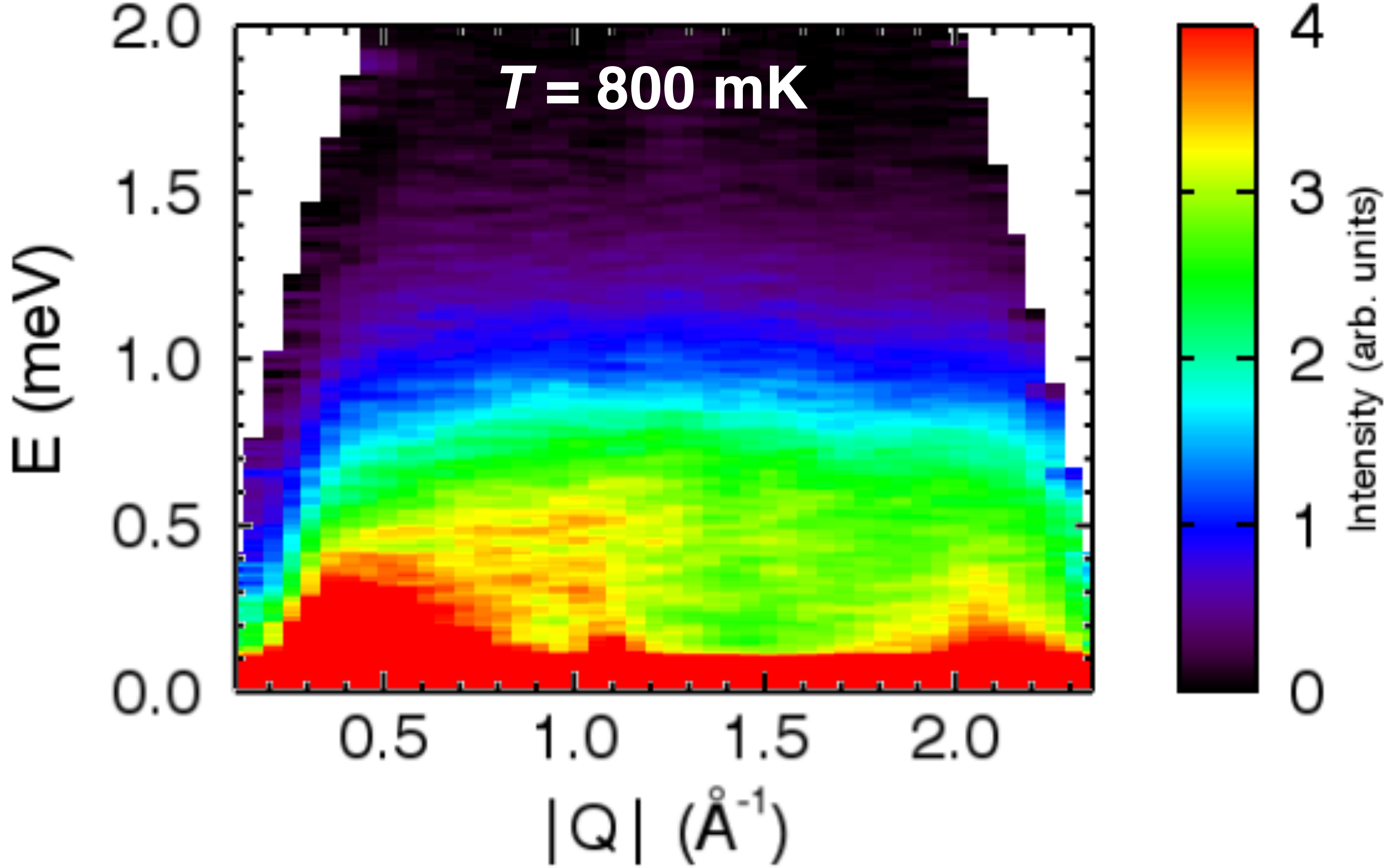

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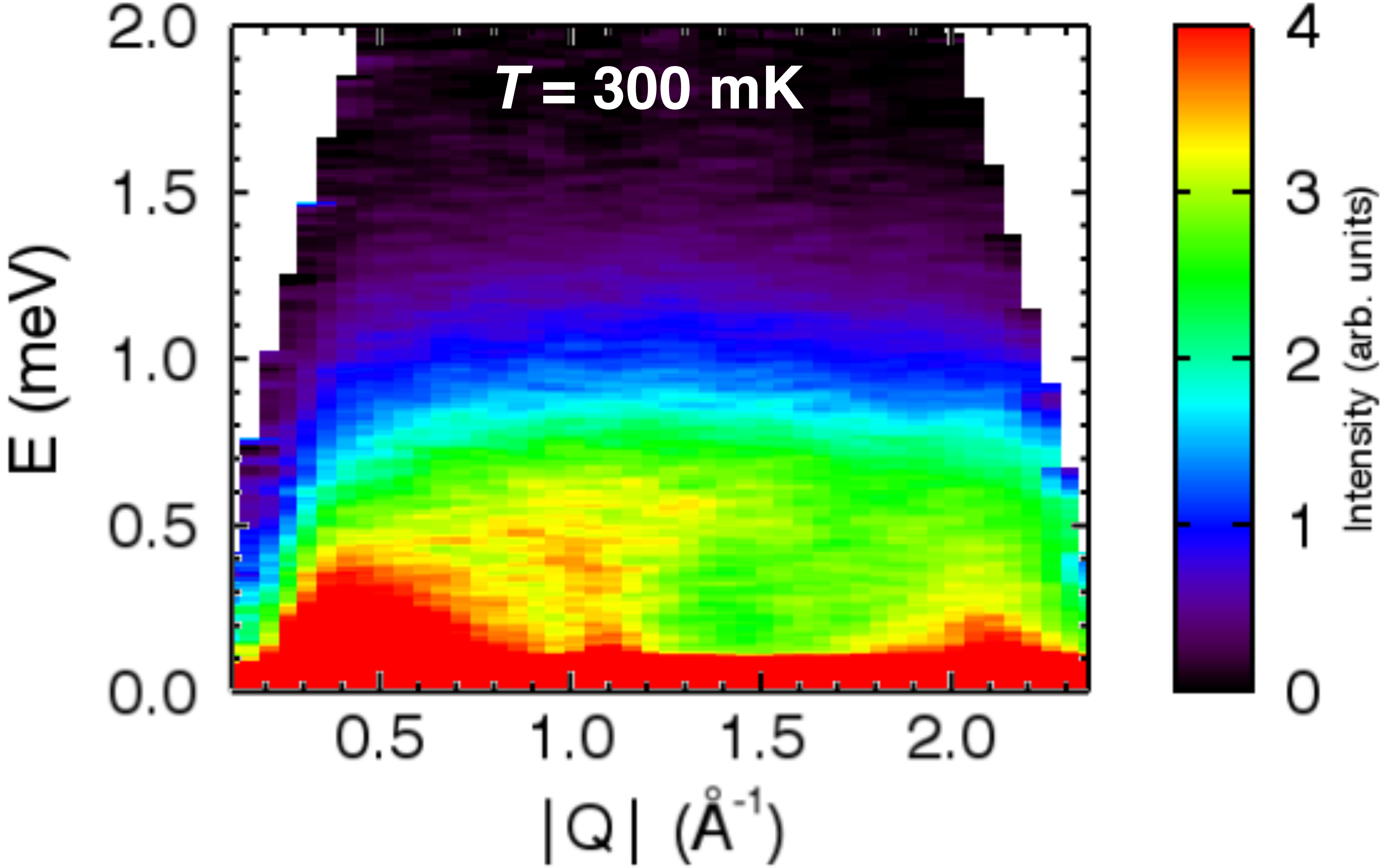
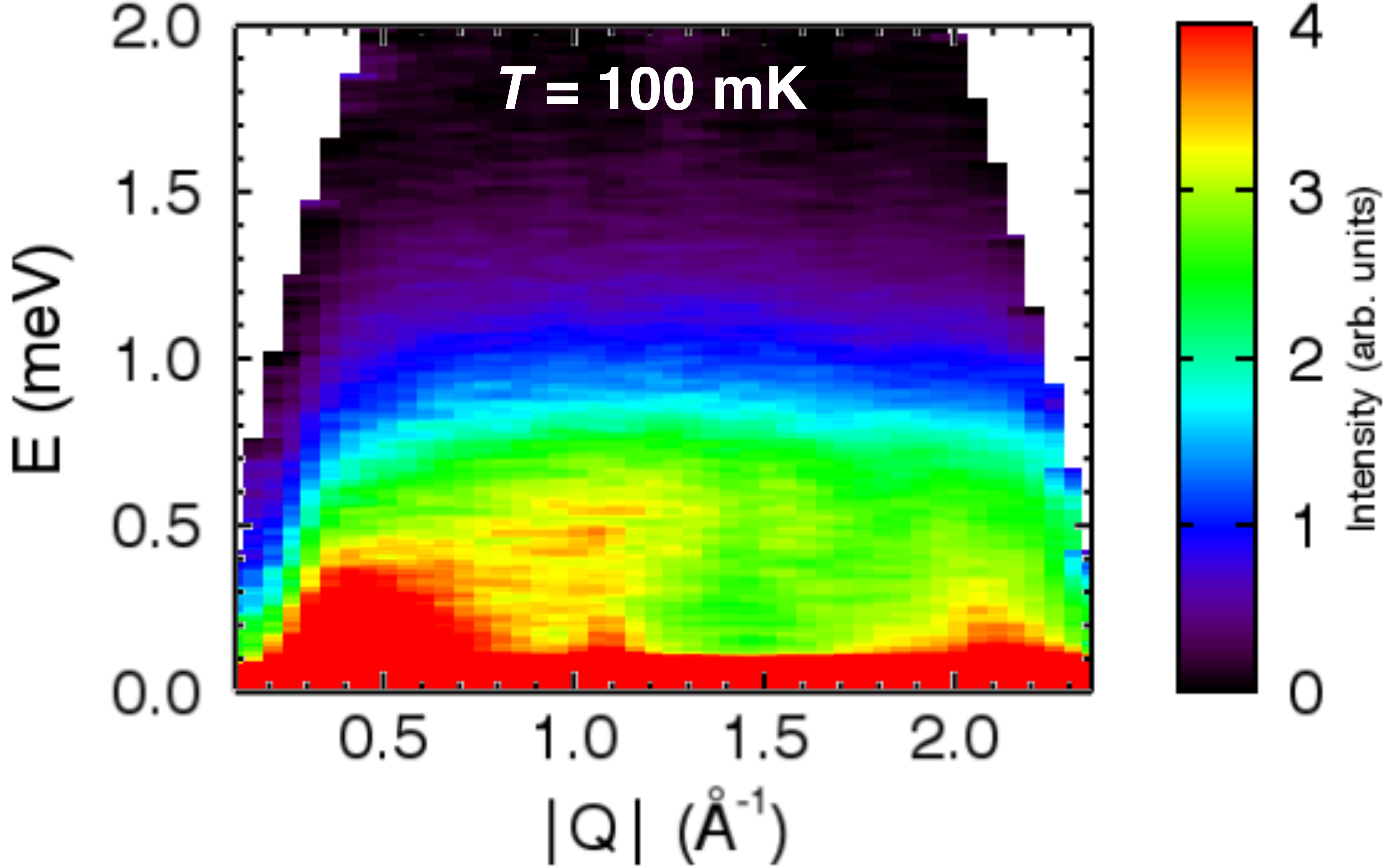
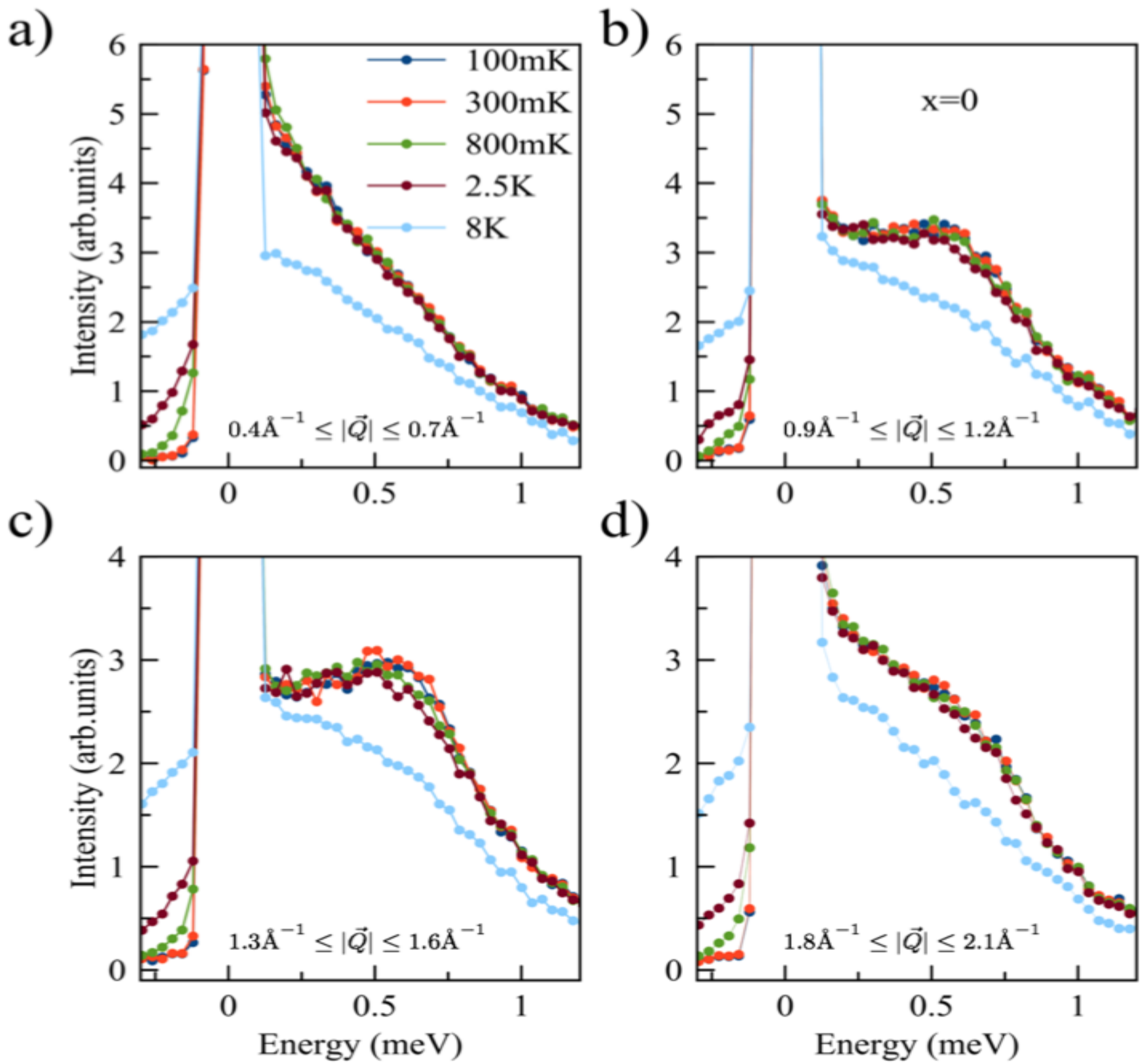


Temperature Dependence of $Yb_2Ti_2O_7$ Spectrum



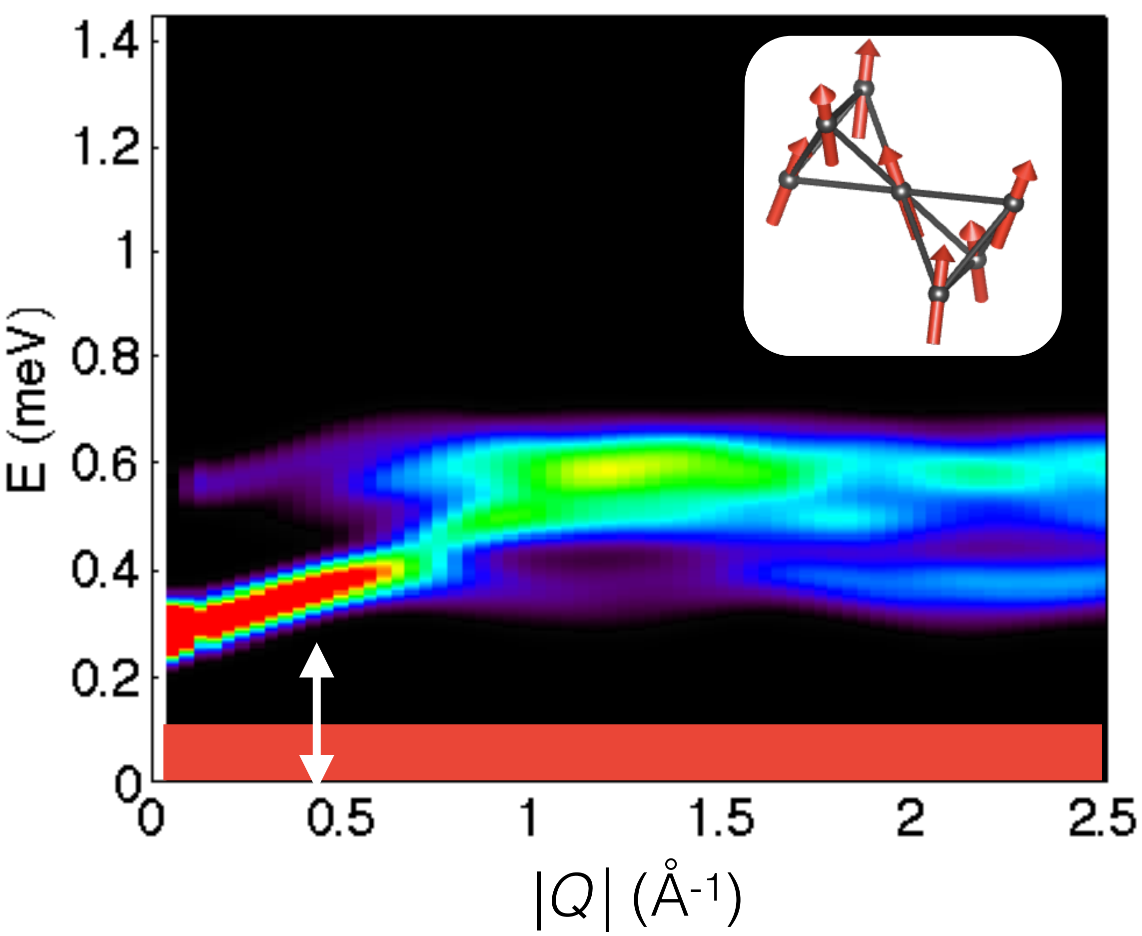
Temperature independence of spectrum



- Continuum of scattering extending to ~1.5 meV
- No change in S(Q,w) up to 2.5 K: **insensitive to the transition seen in the specific heat**

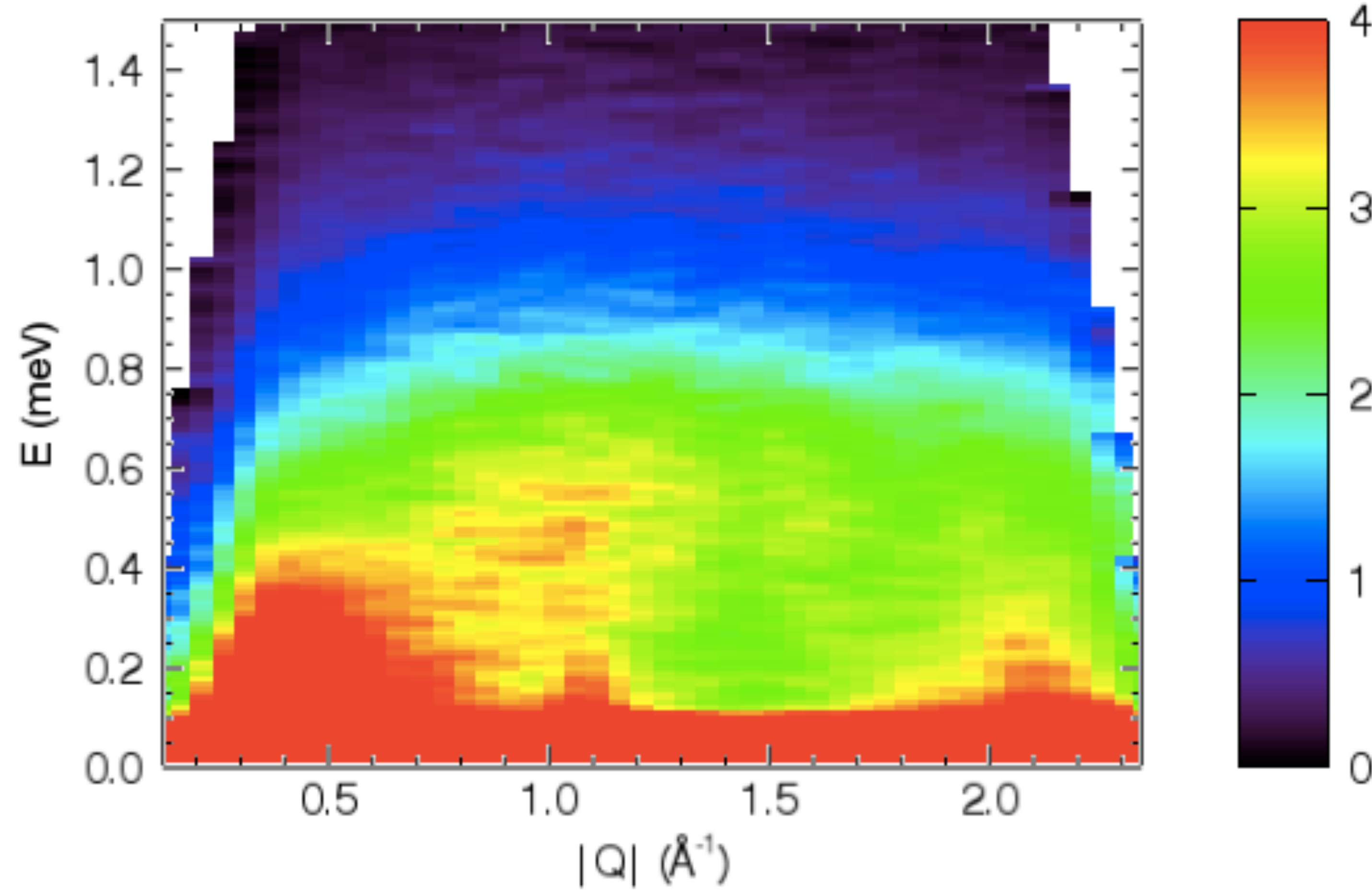
Gapless Excitations Not Expected

Calculated zero-field spin waves



Using Exchange parameters from Ross *et al*, Phys. Rev. X **1**, 021002 (2011)

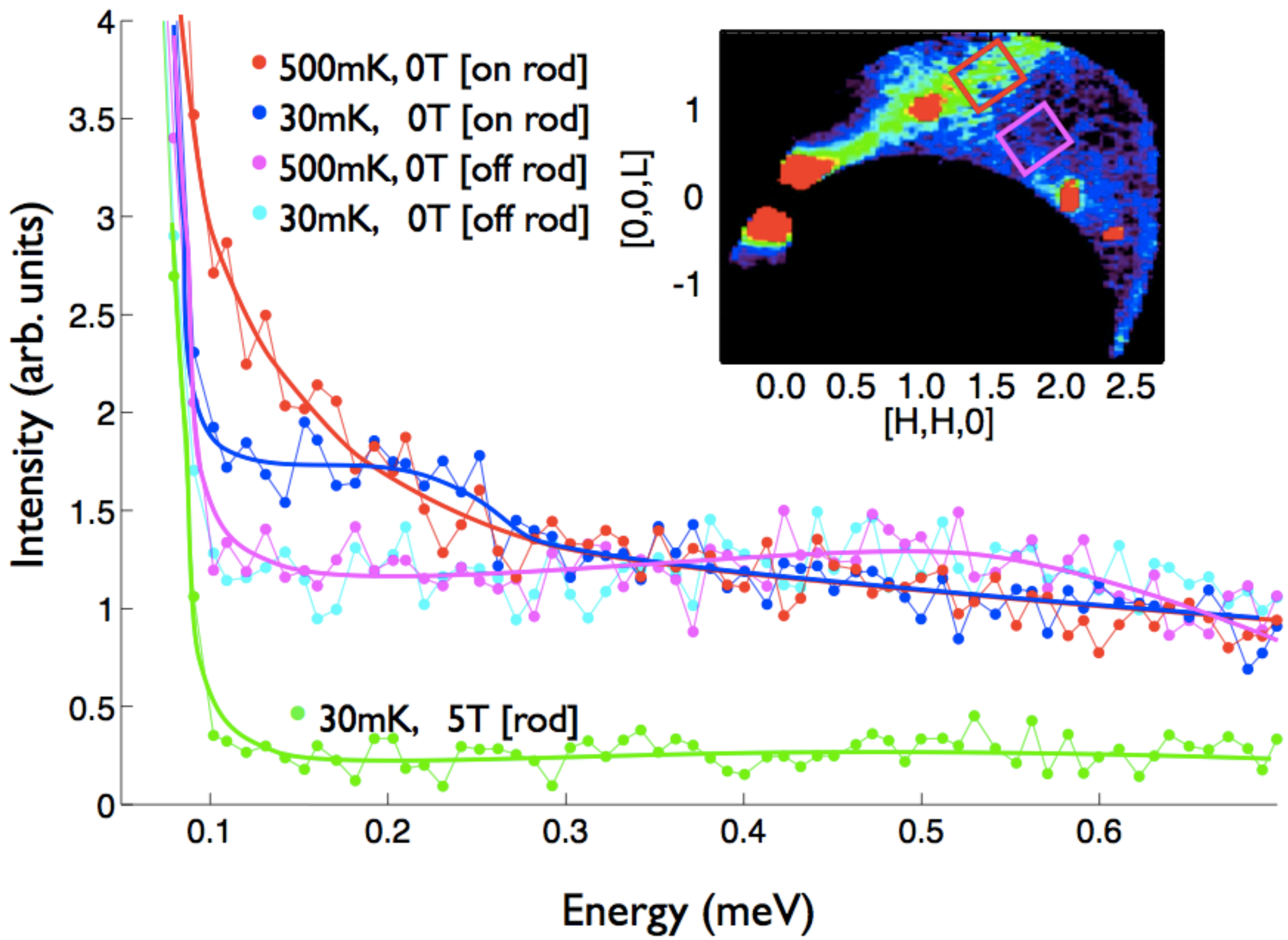
Measured $Yb_2Ti_2O_7$ 100 mK



Time-of-Flight Spectrometer (DCS)

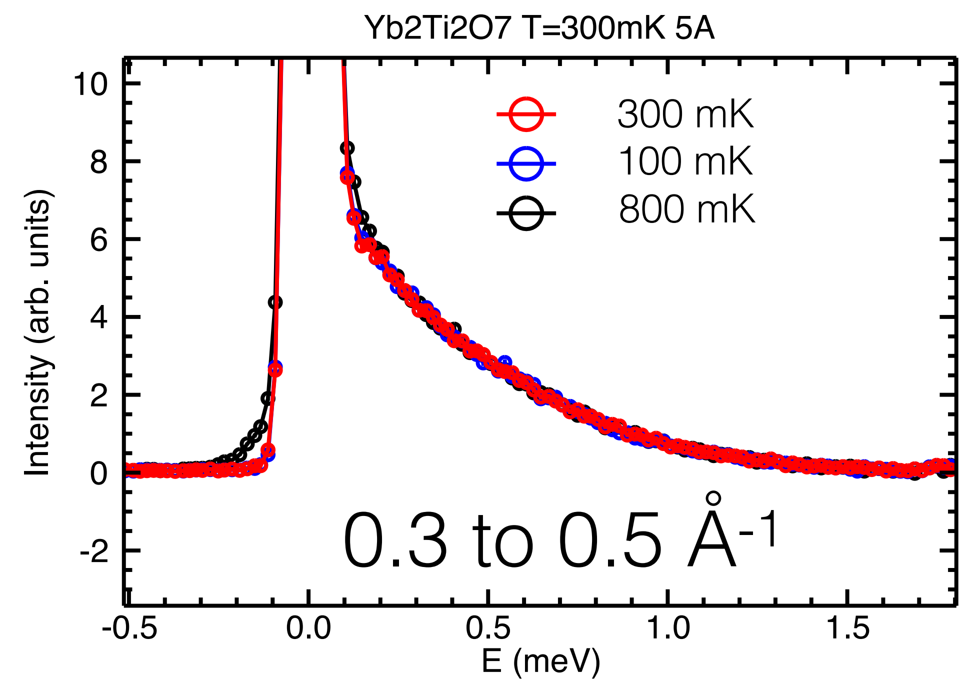
Comparison to Crystals

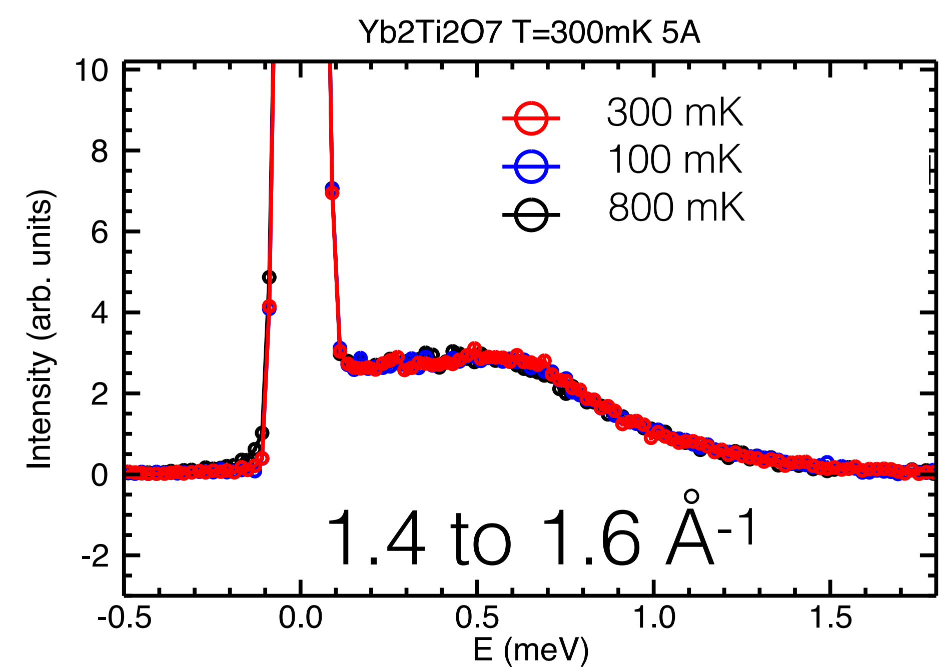
Crystal



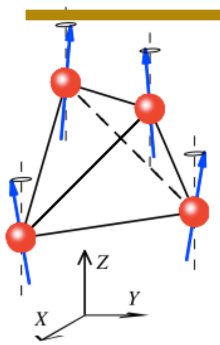
- Lineshapes are essentially the same as for crystals
- Is the spectrum insensitive to details of the transition seen in heat capacity?
- **Why is the spectrum insensitive to the presence or absence of order?**

Powder

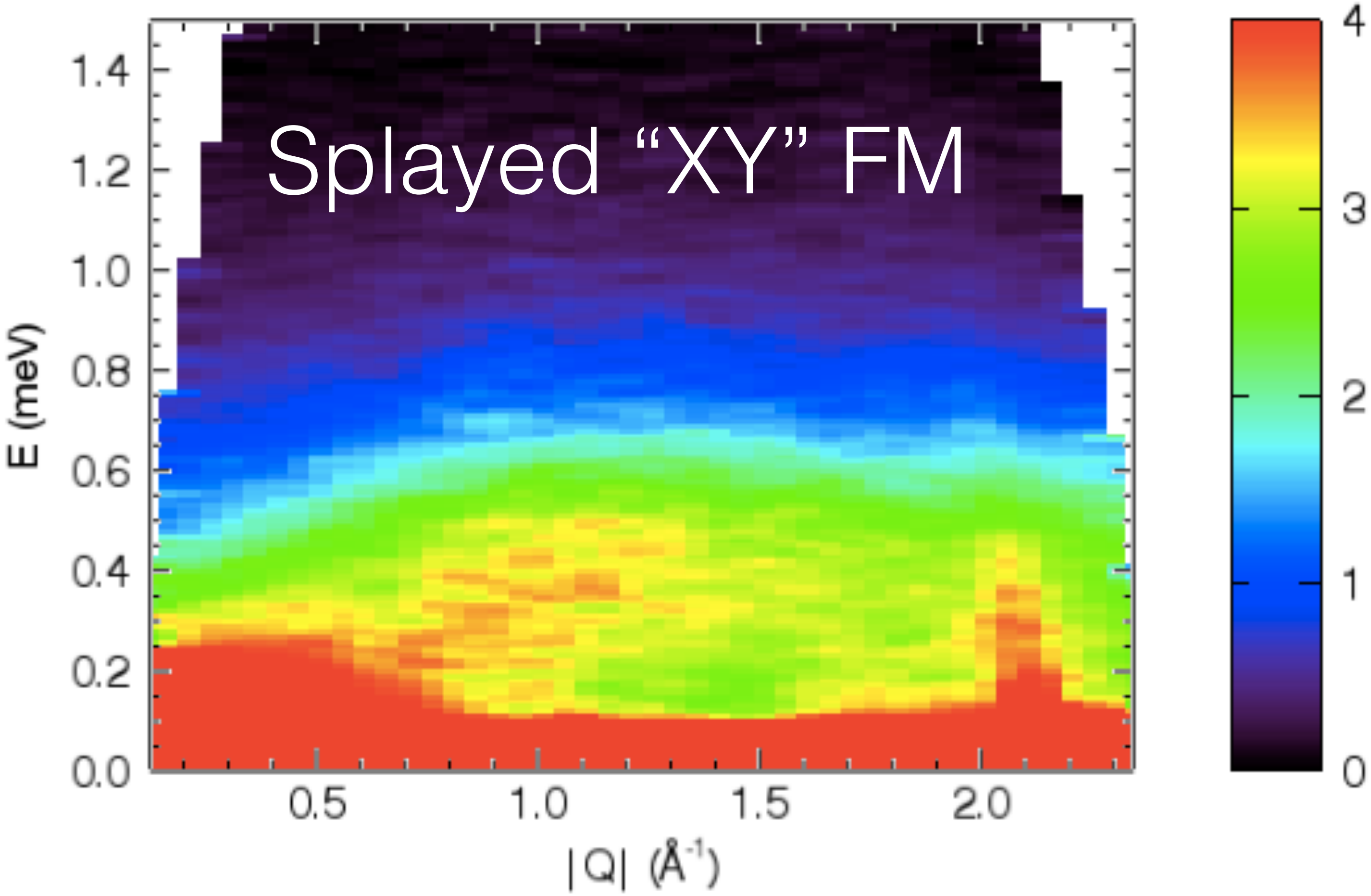




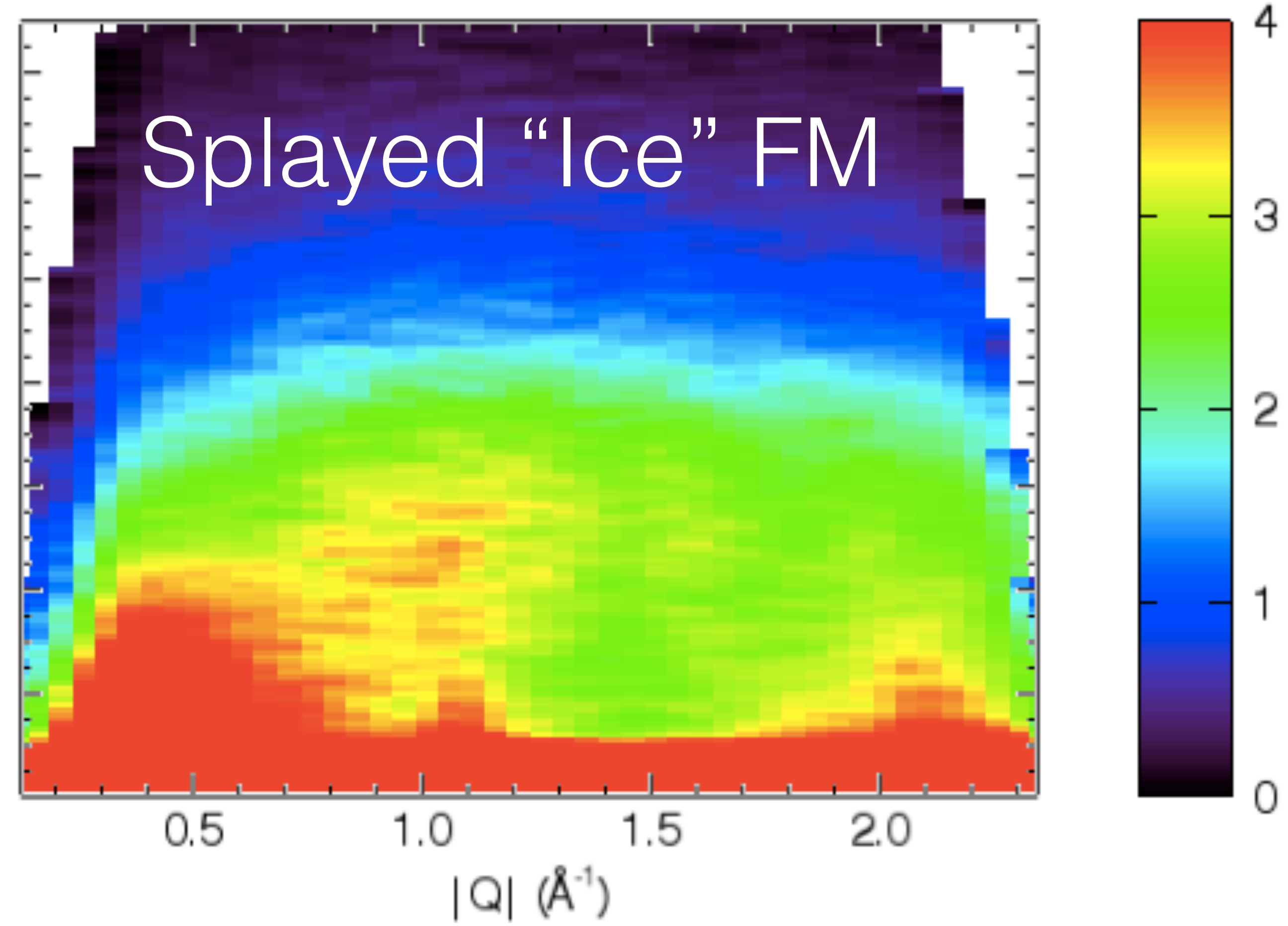
Inelastic Spectrum Compared to $Yb_2Sn_2O_7$



$Yb_2Sn_2O_7$ 100 mK



$Yb_2Ti_2O_7$ 100 mK

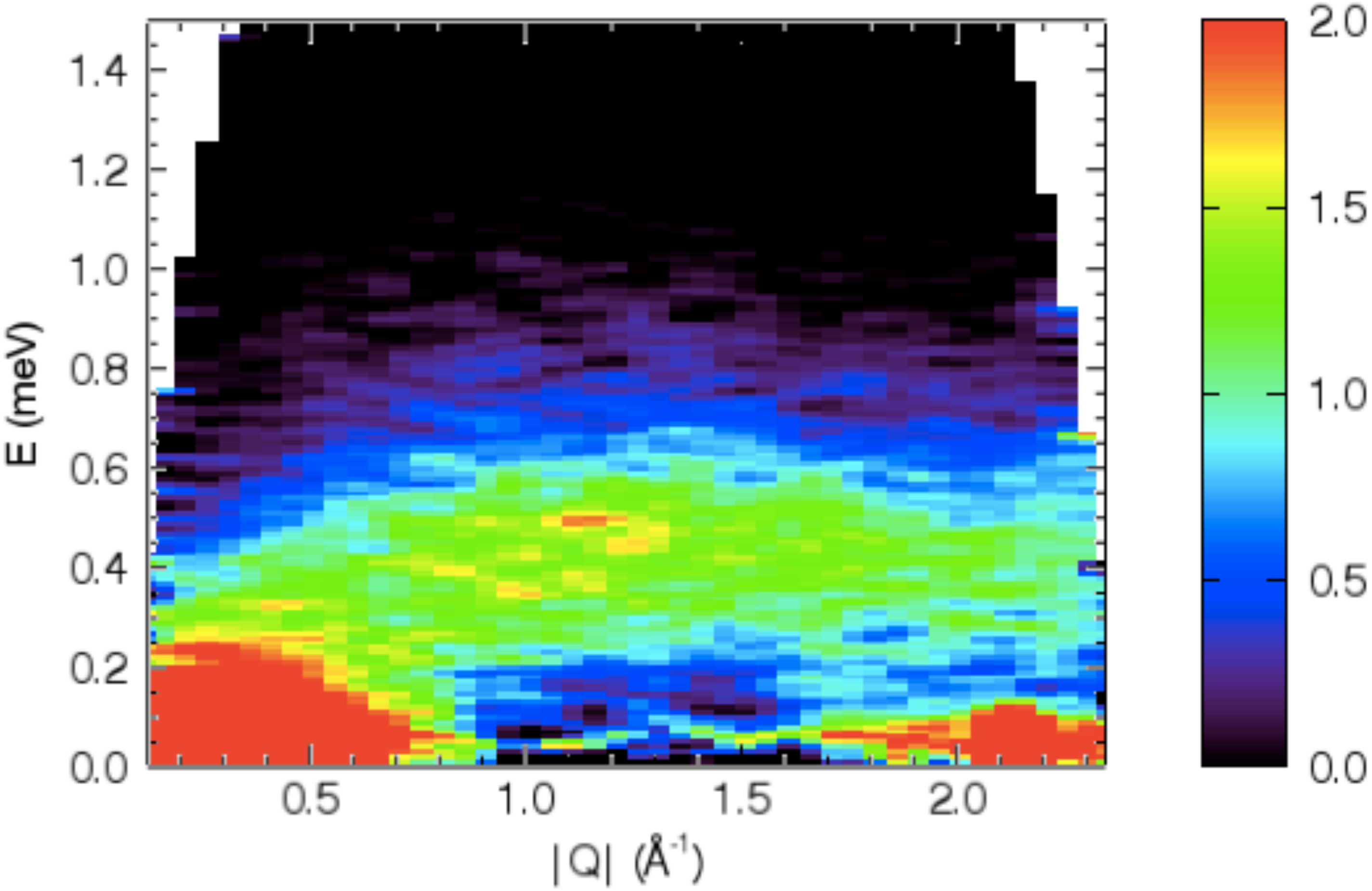


See: Dun et al, PRB **87**, 134408 (2013)

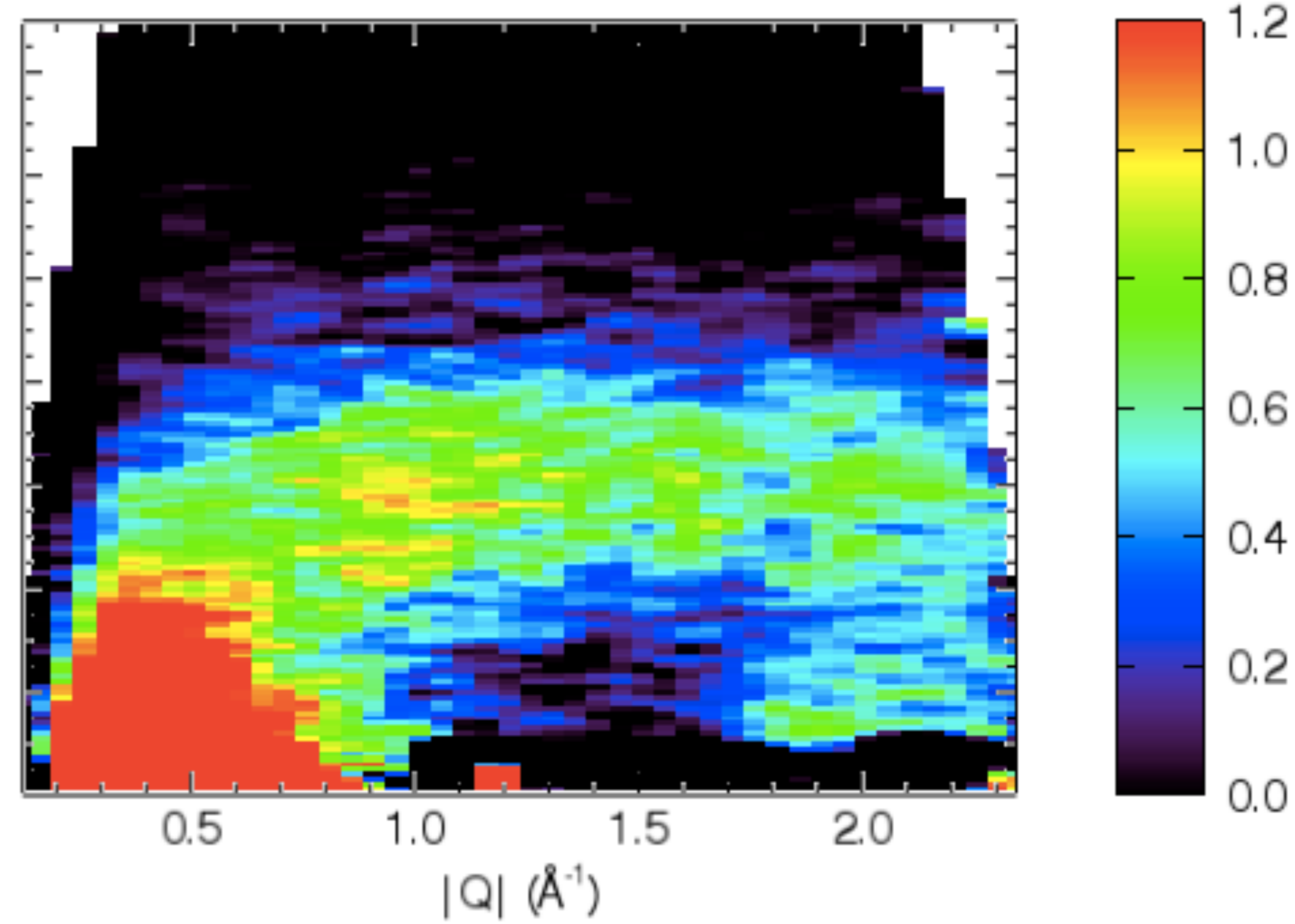
Thanks to C. Wiebe, H. Zhou, Z. Dun

Compare $Yb_2Sn_2O_7$ and $Yb_2Ti_2O_7$

$Yb_2Sn_2O_7$ 100 mK
20K background



$Yb_2Ti_2O_7$ 100 mK
8K background

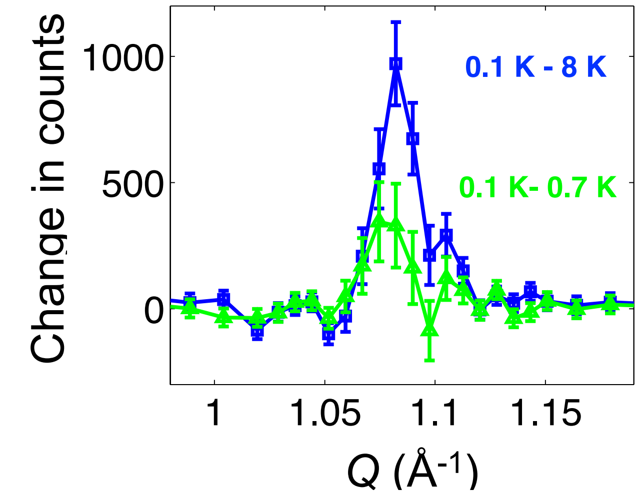


See: Dun et al, PRB **87**, 134408 (2013)

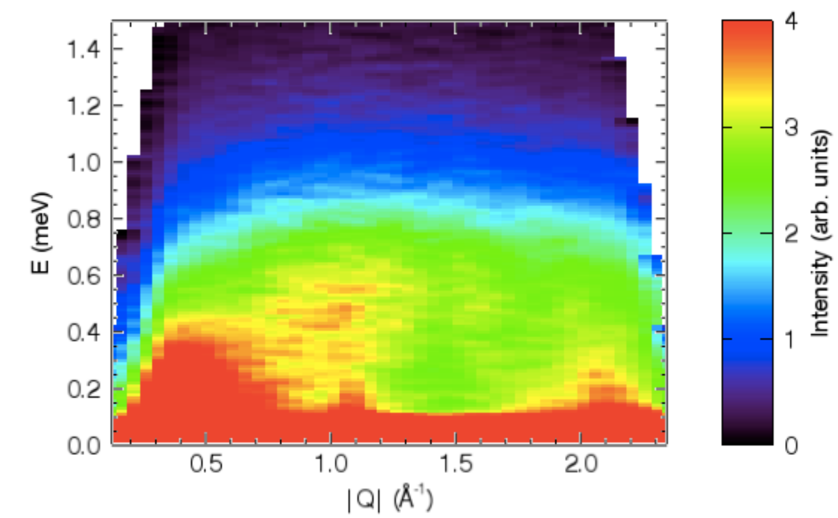
Thanks to C. Weibe, H. Zhou, Z. Dun

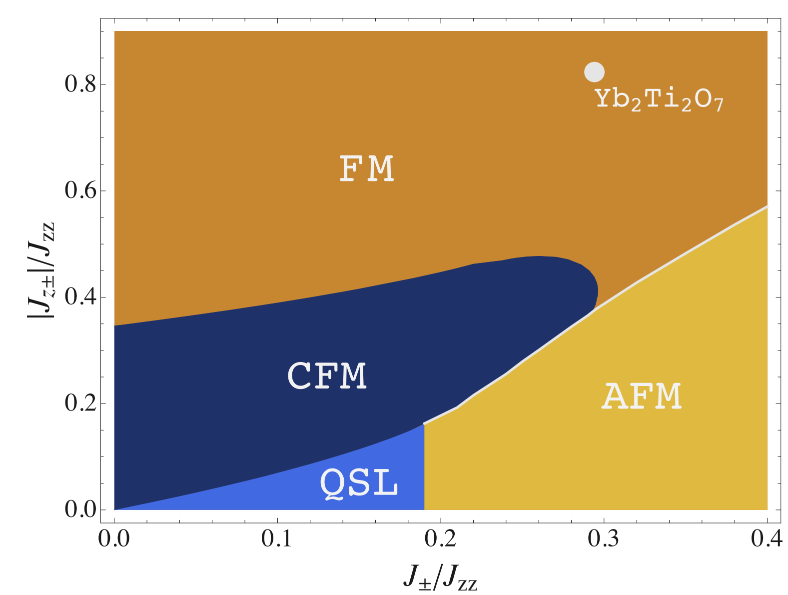
Summary

- $Yb_2Ti_2O_7$'s true ground state has seemingly been obscured by sample dependence issues
- We showed elastic and inelastic neutron scattering studies of a **powder sample known to be stoichiometric**
- **Partially ordered moment of ~0.9 μ_B** (53 % ordered), consistent with splayed "Ice" ferromagnetism, Temp dependence does not correlate with anomaly in specific heat
- **Gapless, continuum-like** spectrum at 100 mK is not conventional magnons, is insensitive to details of transition, and strongly resembles $Yb_2Sn_2O_7$



$Yb_2Ti_2O_7$ 100 mK





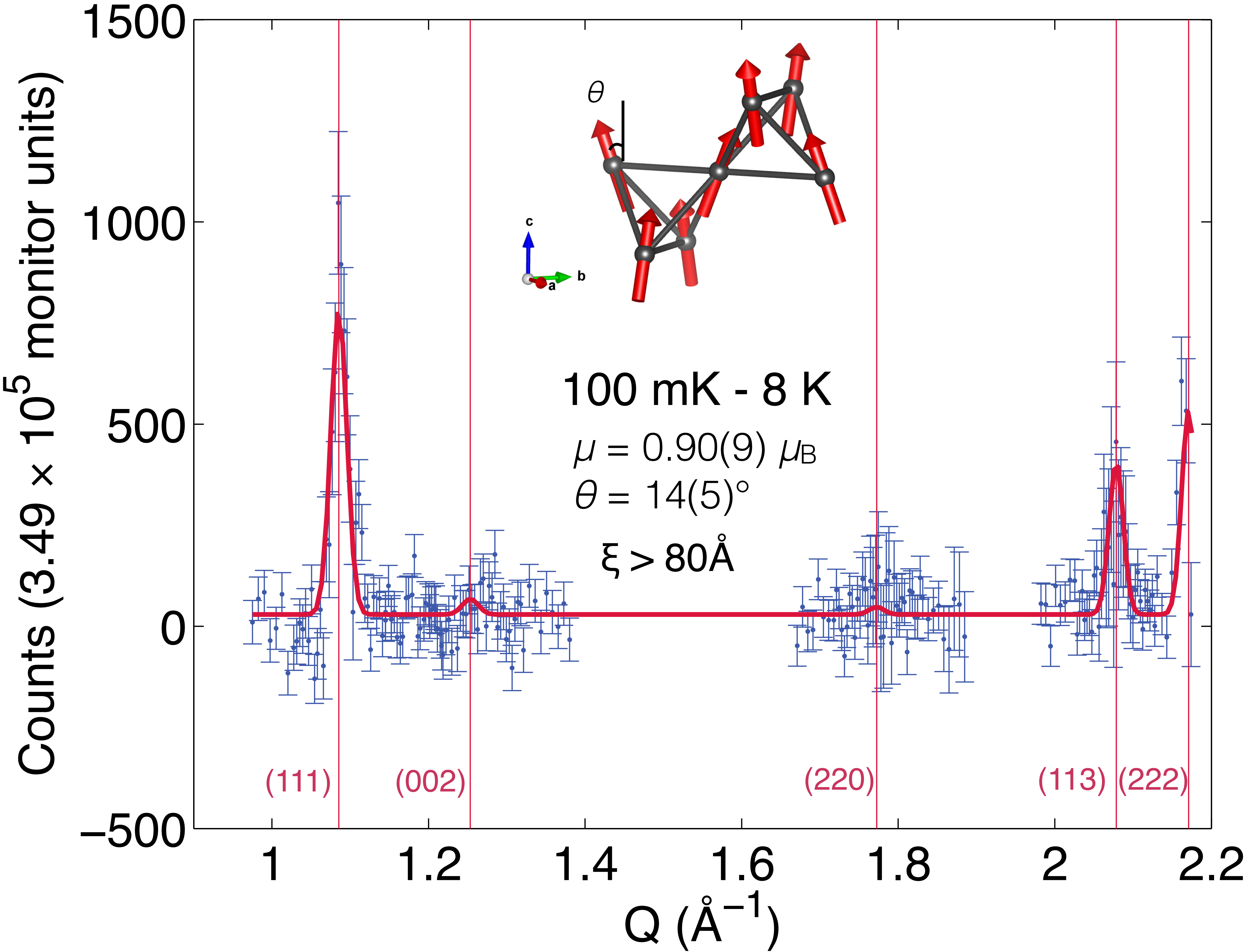
Thanks to…

- Bruce Gaulin
- Edwin Kermarrec
- Jonathan Gaudet

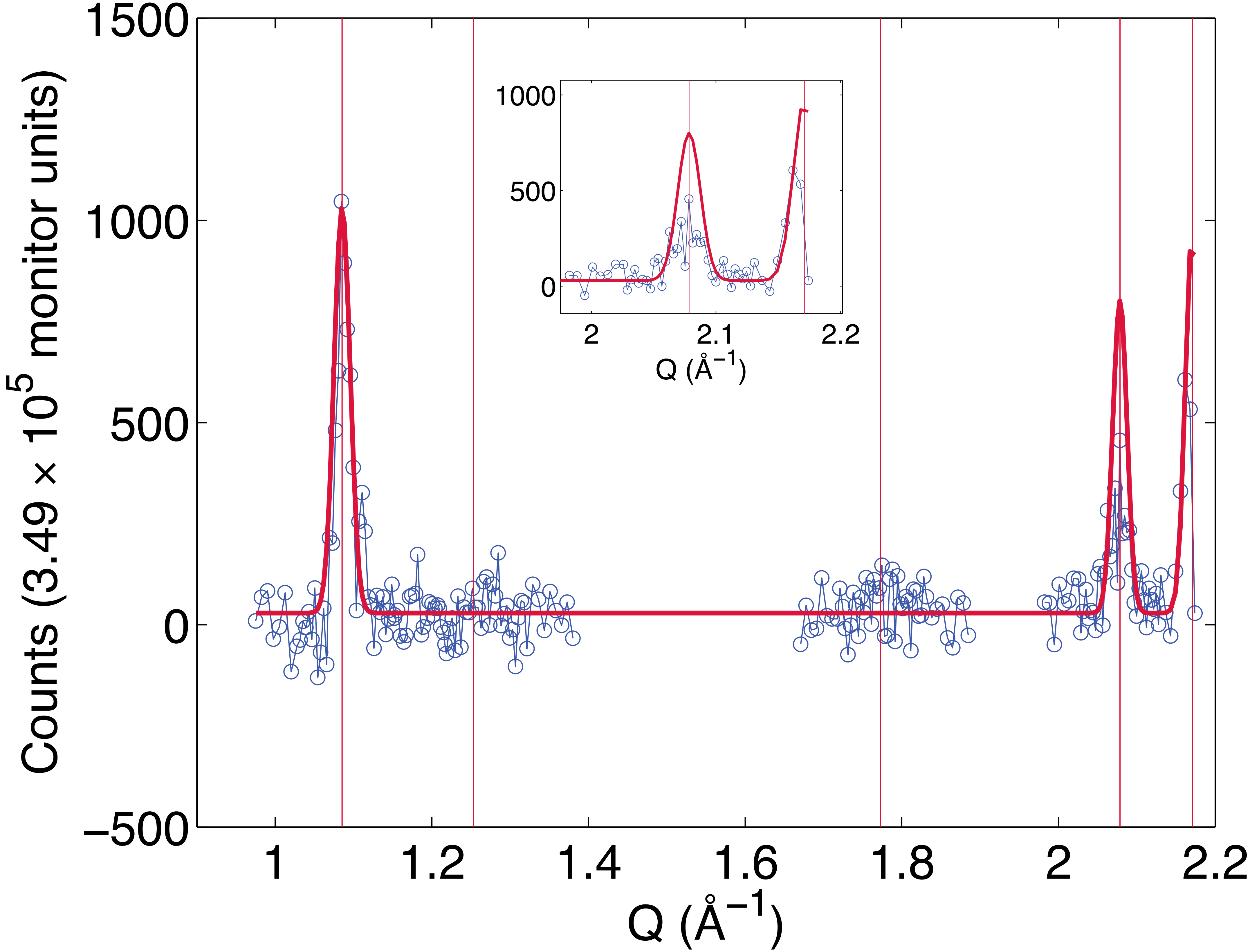
- **Juscelino Leao**
- Jiajia Wen
- Nick Butch
- Leland Harriger
- Jan Kycia
- Jeff Quilliam
- Lucile Savary
- Leon Balents
- Chris Weibe
- Haidong Zhou
- Zhiling Dun

- NSERC
- Department of Energy
- Colorado State University

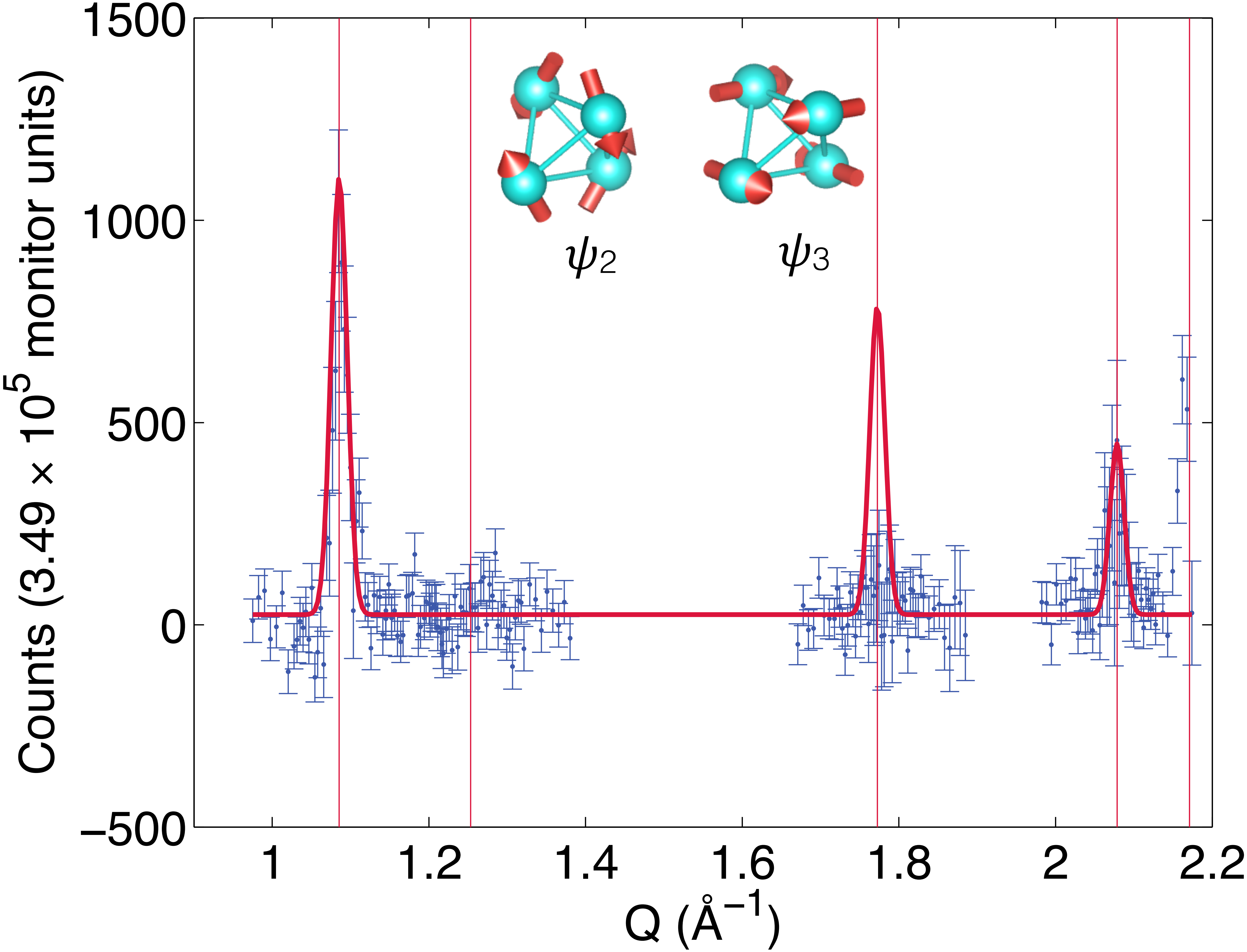
Resolution-Limited Elastic Scattering (100mK - 8K)



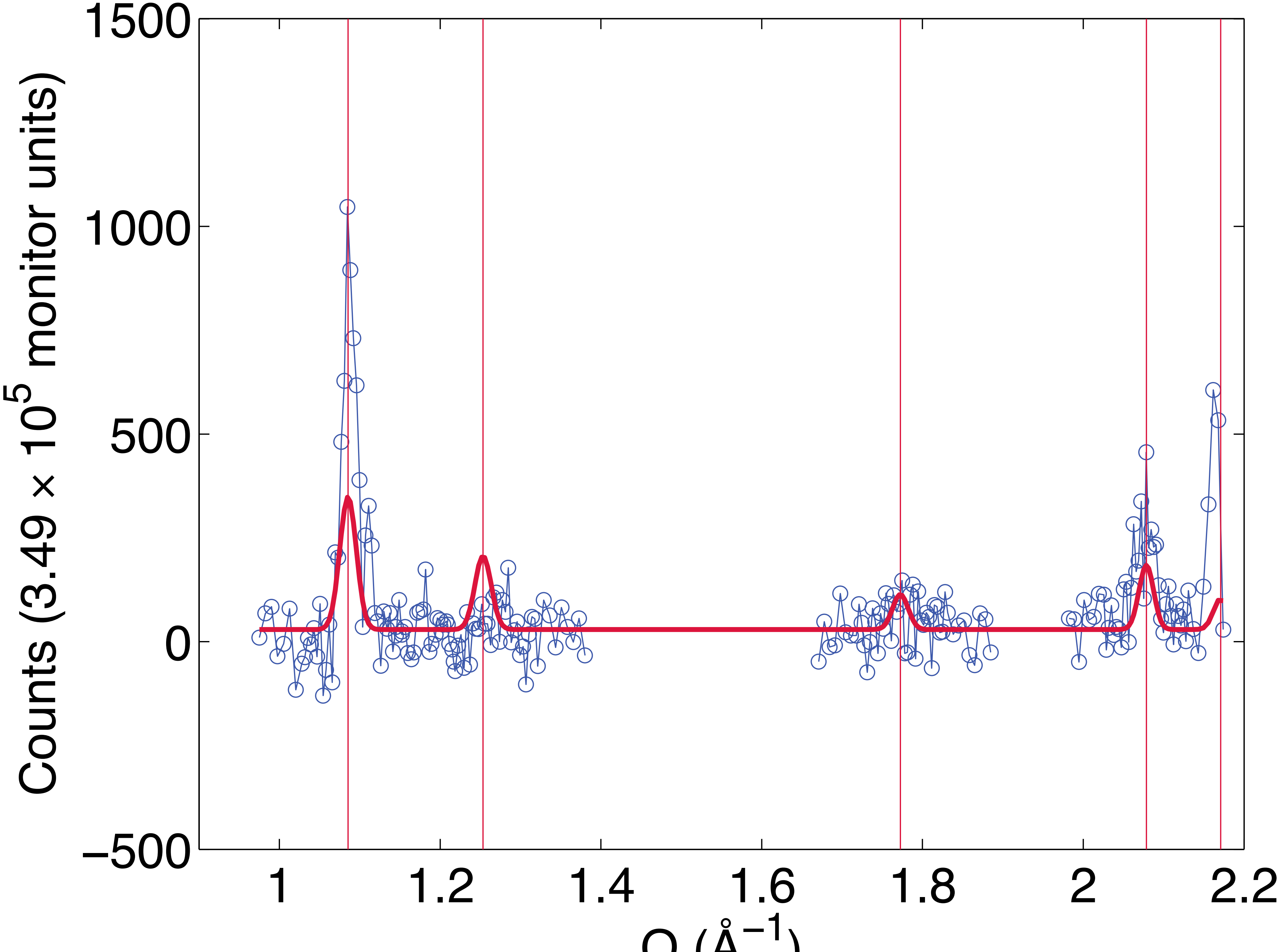
Collinear model



AFM (psi2/psi3) model



Ordered Ice



Counts (3.49 × 10^5 monitor units)
1500
1000
500
0
−500
1
1.2
1.4
1.6
1.8
2
2.2
Q (Å^−1)