Partial Antiferromagnetic Helical Order in $Fe_3PO_4O_3$

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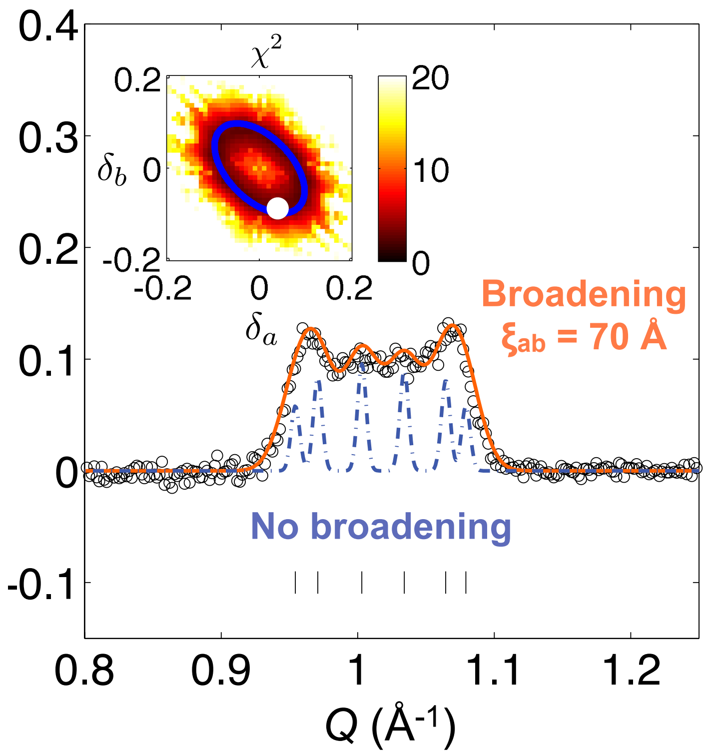
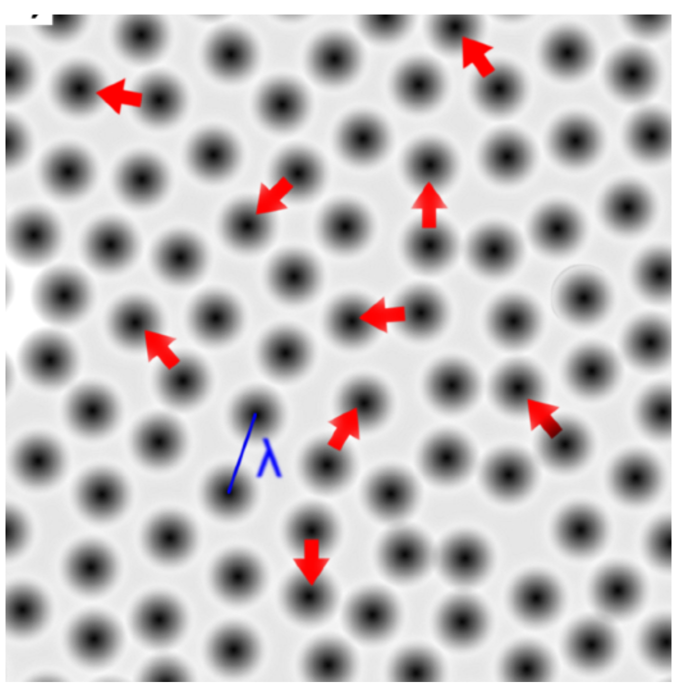
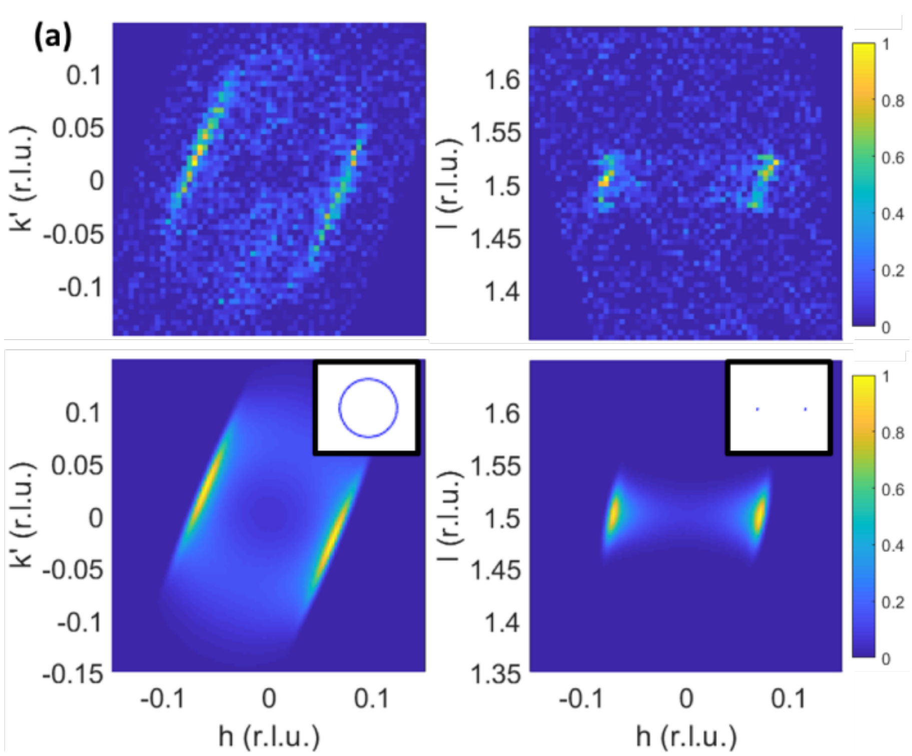
Colorado State University

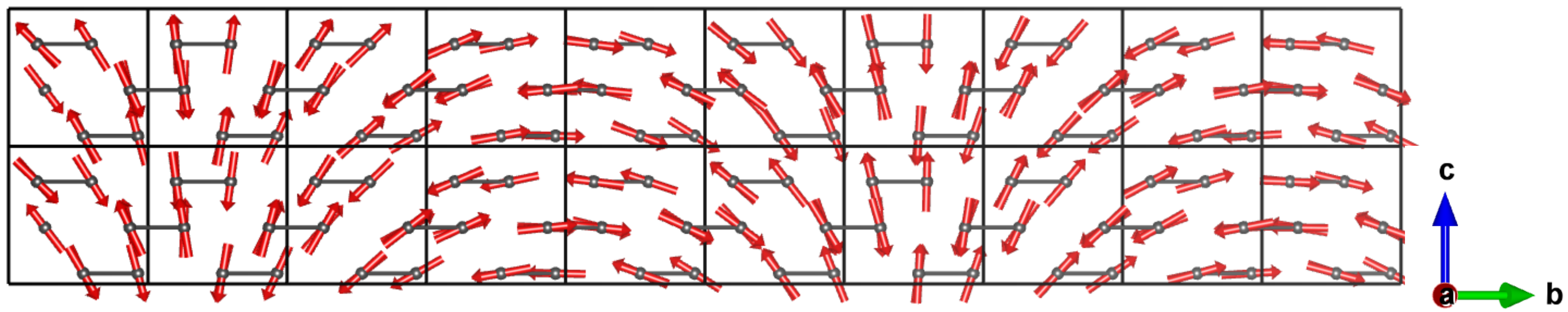
EPIQS-TMS meeting 2019
KITP, Santa Barbara, CA

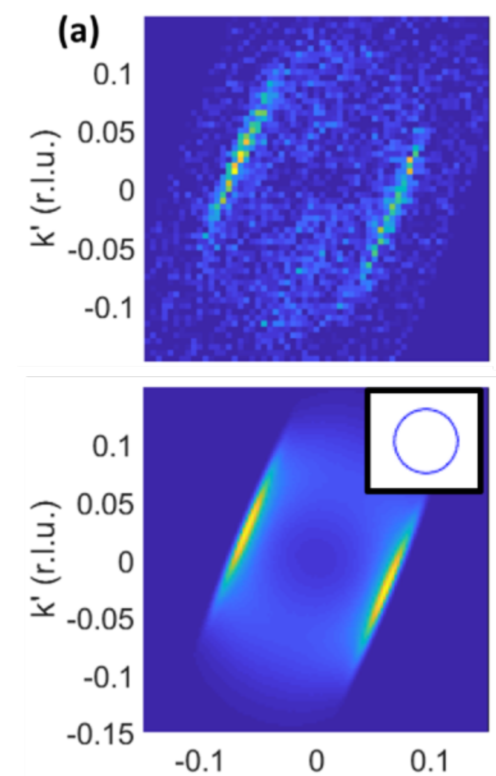




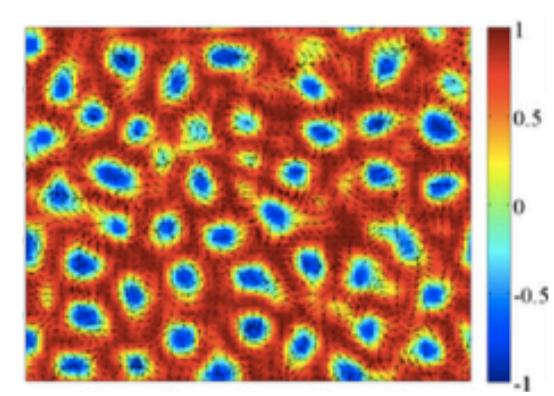


Outline





- "Partial" helical order
- $Fe_3PO_4O_3$: Antiferromagnetic partial helical order with short correlation length
- Evidence for a disordered, antiferromagnetic Skyrmion-like state

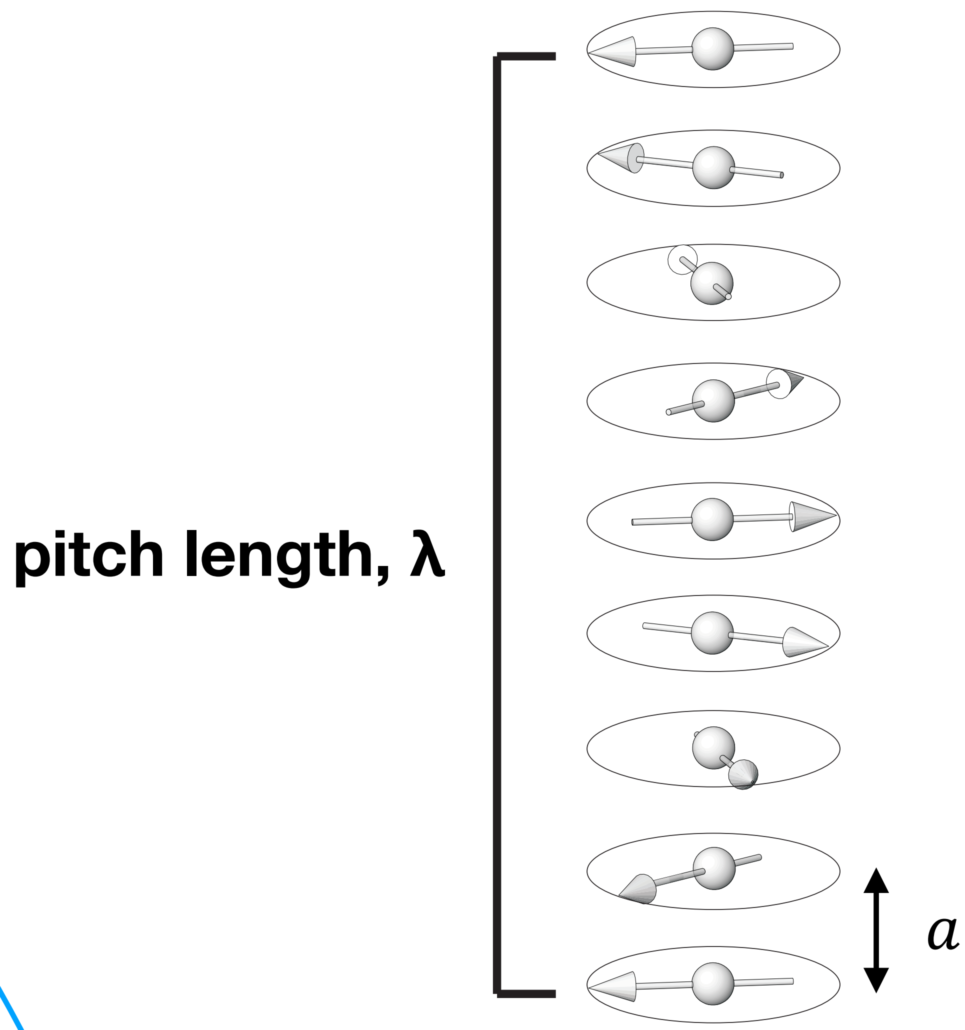


Helical order

Caused by competing interactions e.g.:

1) Ferromagnetism competing with Dzyaloshinskii Moriya (S_i x S_j)

2) Heisenberg AFM 1st and 2nd neighbors

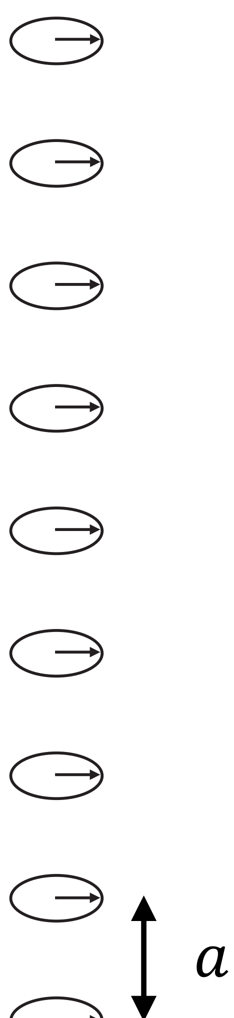


$$H = J_1 \sum_{<i,j>} S_i \cdot S_j + J_2 \sum_{<<i,j>>} S_i \cdot S_j$$

$$H = J \sum_{<i,j>} S_i \cdot S_j + D \sum_{<i,j>} S_i \times S_j$$

Diffraction from helical order: “satellite” peaks

real space



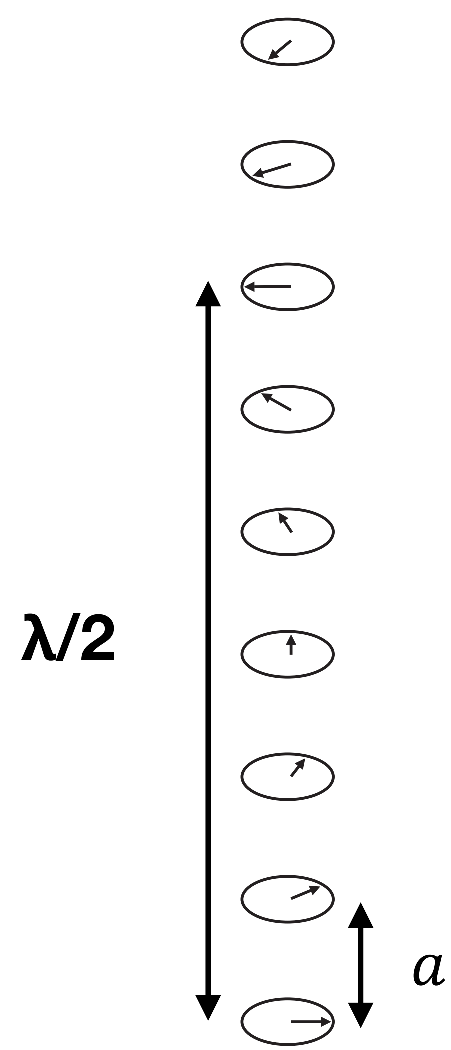
reciprocal space (lattice and magnetic)

$2\pi/a$

0

$-2\pi/a$

real space



reciprocal space (lattice and magnetic)

$2\pi/a \pm 2\pi/\lambda$

$0 \pm 2\pi/\lambda$

$-2\pi/a \pm 2\pi/\lambda$

“satellite peaks” near the parent magnetic structure zone centers

Example: B20 Compounds

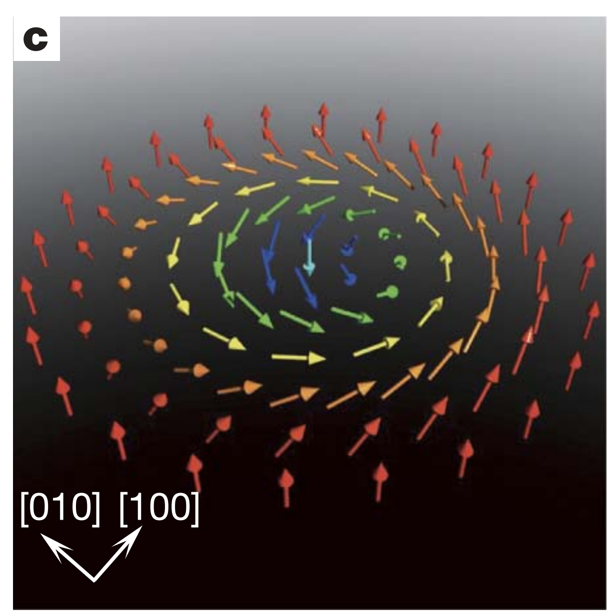
- The B20 compounds with space group $P2_13$, e.g. MnSi, $Fe_{1-x}Co_xSi$ exhibit helical order

- Dominant FM interactions plus DM interaction are responsible ("locally ferromagnetic")

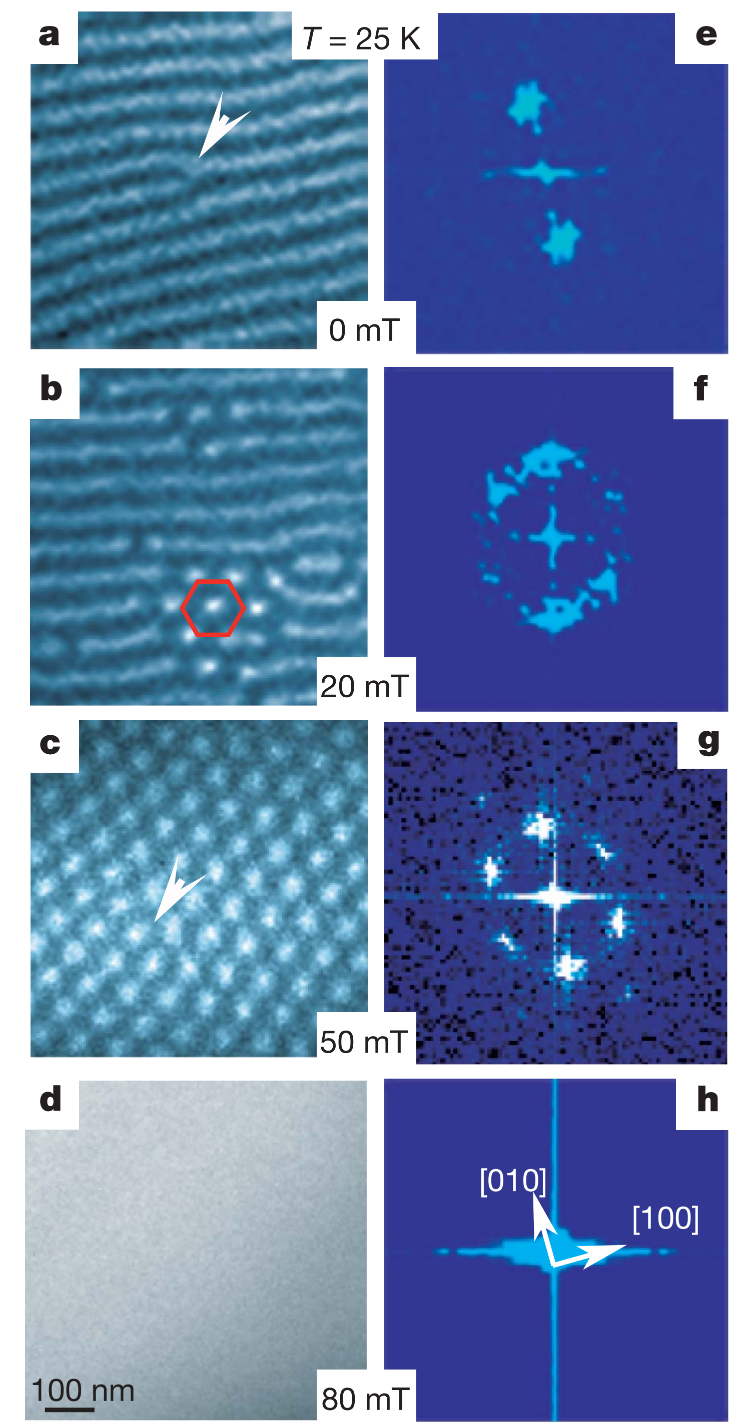
- **Bonus:** under a small applied field, the helical order transforms into a Skyrmion Lattice!

Lorentz TEM: real space images of helical and Skyrmion lattice order in $Fe_{0.5}Co_{0.5}Si$

Yu, Onose, Kanazawa, Park, Han, Matsui, Nagaosa, Tokura, *Nature*, (**2010**), *465*, 901–904.

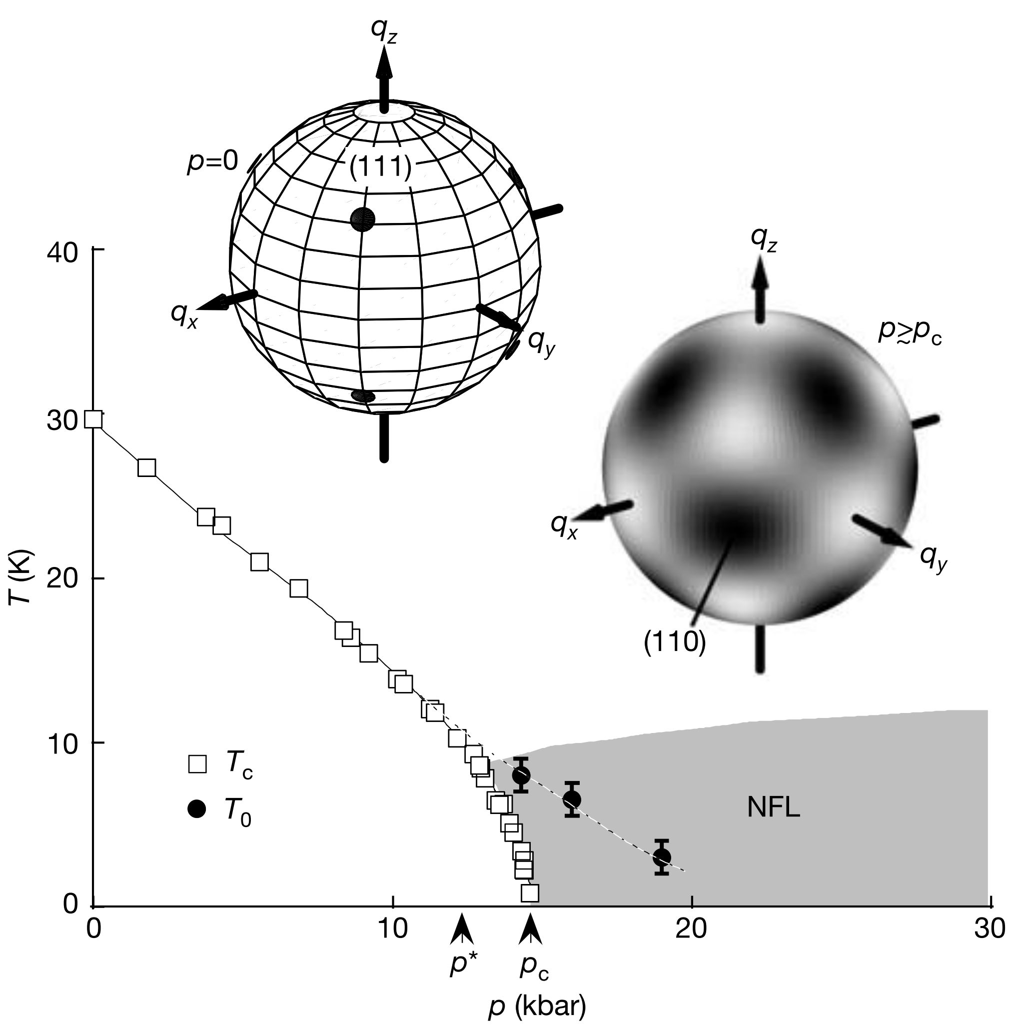


real space **"diffraction" (FFT)**



“Partial” Helical order

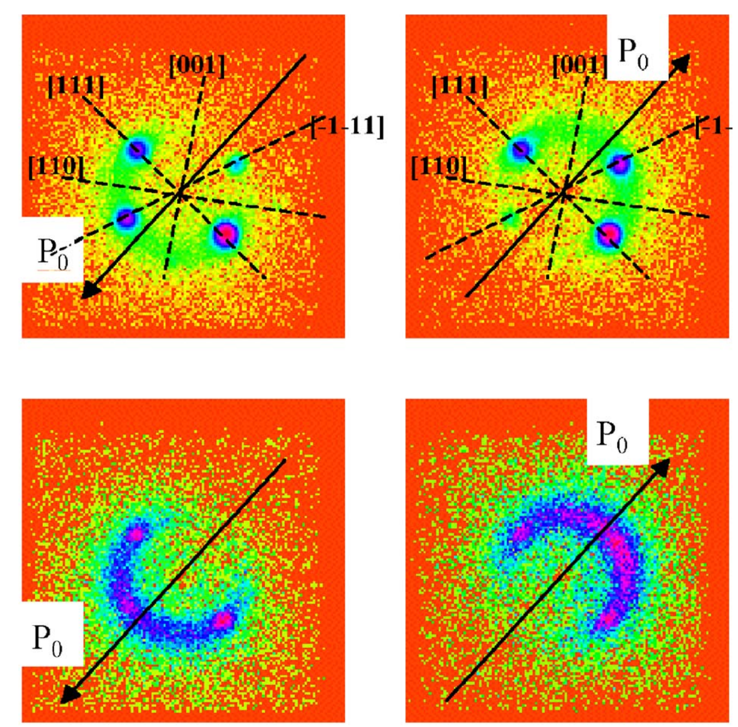
- MnSi: under pressure, and at temperatures $T>T_c$, exhibits a **spherical shell** instead of Bragg spots
- **“Partial order”**: Well-defined pitch length, but not *direction*



C. Pfleiderer et al, Nature **427**, 227-231 (2004)

below T_c (helical)

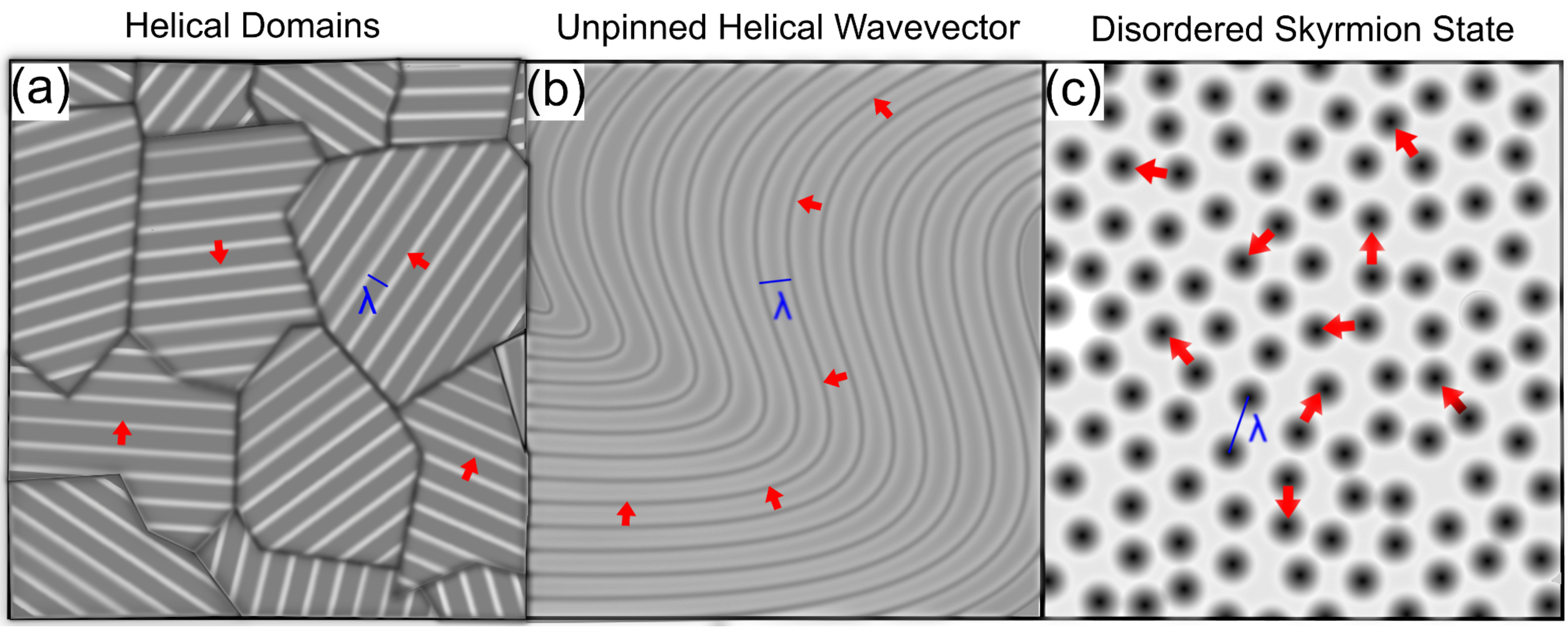
Above T_c (Partial order)



S.V. Grigoriev et al, PRB 72 134420 (2005)

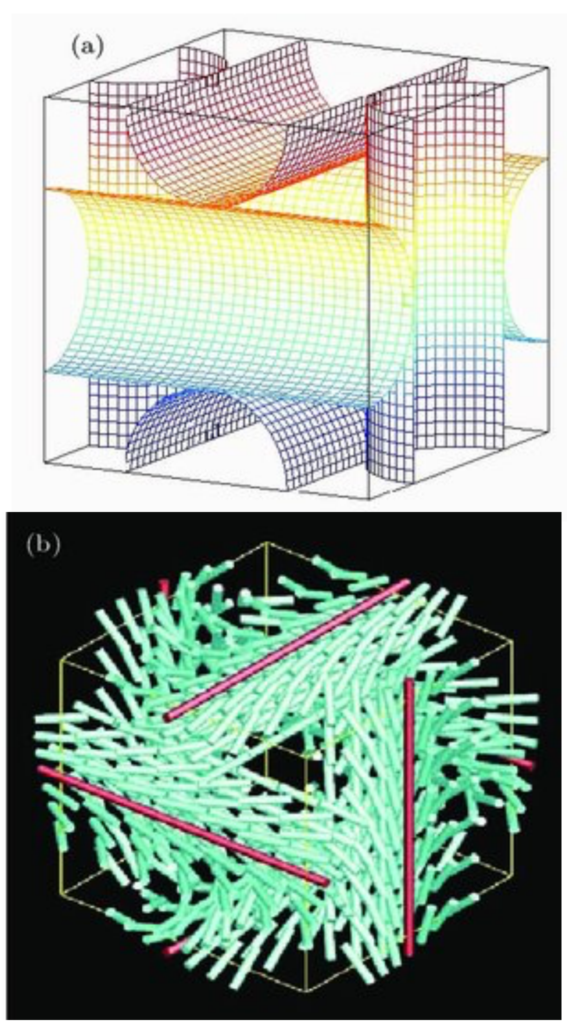
Origin of “Partial” Helical order?

“Partial order”: Well-defined pitch length, but not *direction*

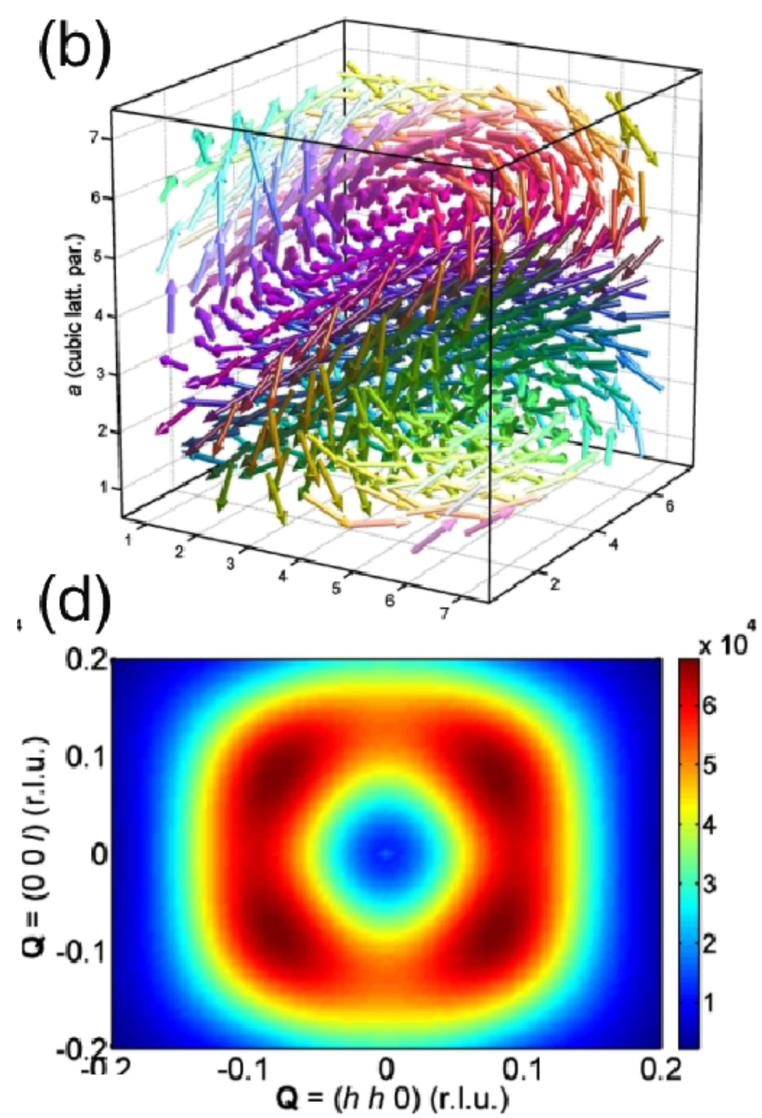


“Blue Phase” description of partial Helical order in MnSi

"Blue Phase" of Liquid Crystals: “Double twist Cylinders” → Same topologies proposed in partial order phase of MnSi. **Very similar to disordered Skyrmions.**



Dou Hu, Ma Hong-Mei, Sun Yu-Bao. Acta Physica Sinica 64 126101 (2015)



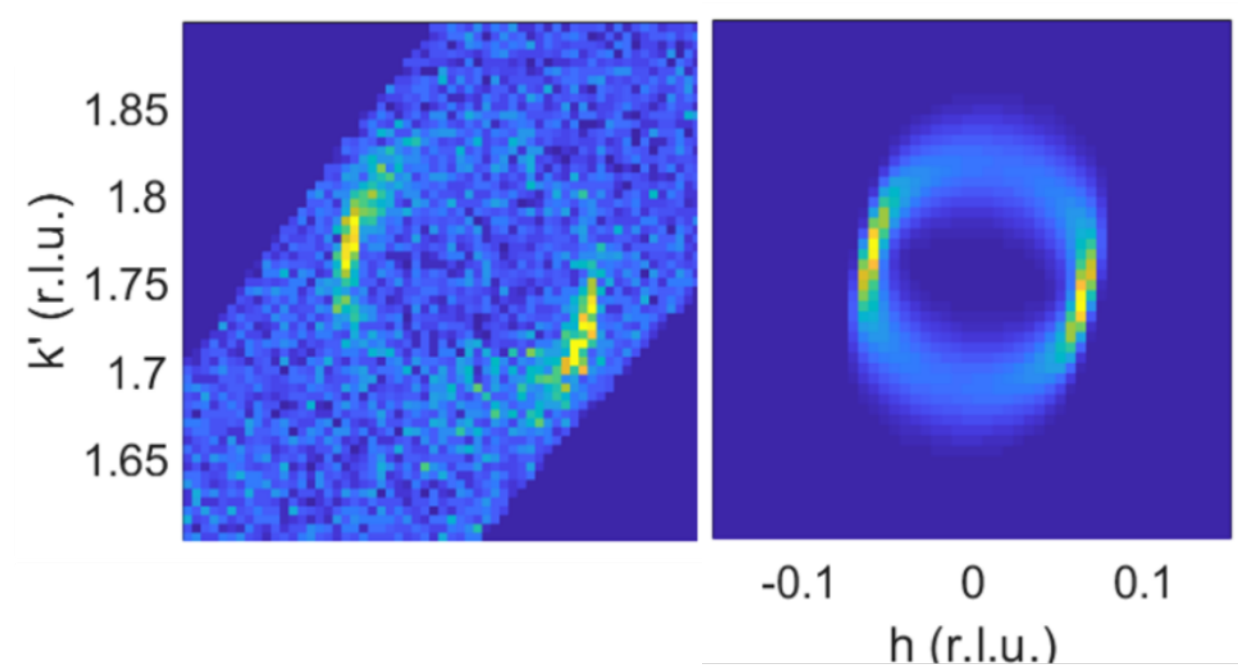
A. Hamann, D. Lamago, Th. Wolf, H. v. Lohneysen, D. Reznik, PRL 107, 037207 (2011)

$Fe_3PO_4O_3$

K. A. Ross, M.M. Bordelon, G. Terho, J. R. Neilson. *Nanosized helical magnetic domains in strongly frustrated $Fe_3PO_4O_3$*. Phys. Rev. B **92**, 134419 [Editor's suggestion] (2015)

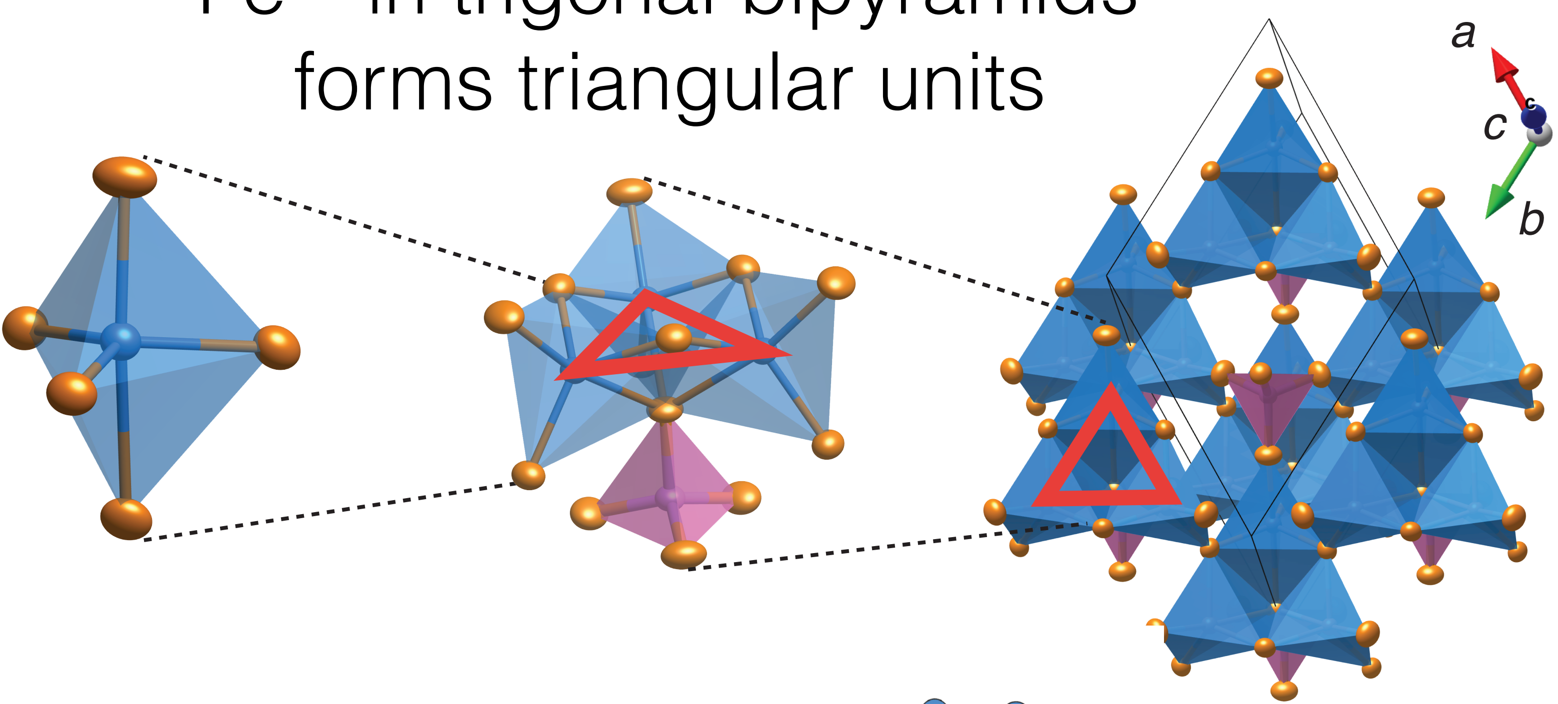
M.J. Tarne, M.M. Bordelon, S. Calder, J.R. Neilson, K.A. Ross. *Tuning the antiferromagnetic helical pitch length and nanoscale domain size in $Fe_3PO_4O_3$ by magnetic dilution.* Phys. Rev. B 96, 214431 (2017)

C. Sarkis, M.J. Tarne, H. Cao, E. Coldren, M. Gelfand, J.R. Neilson, K.A. Ross. *Partial Antiferromagnetic Helical Order in $Fe_3PO_4O_3$.* arXiv:1910.08818 [cond-mat.str-el] (2019)

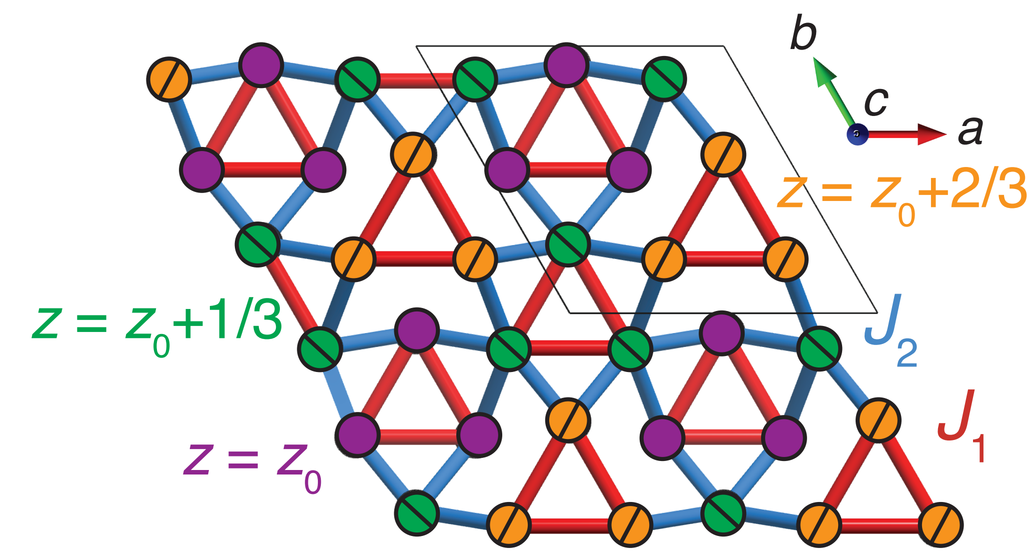


Triangular plaquettes on a rhombohedron

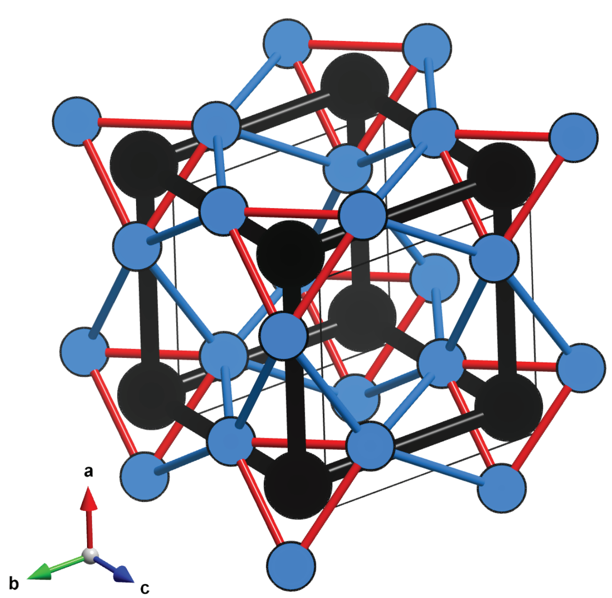
Fe^{3+} in trigonal bipyramids forms triangular units

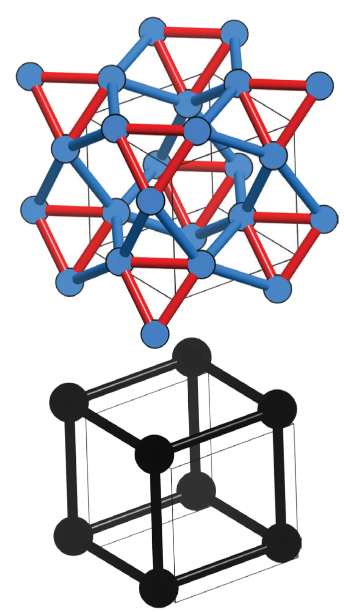


From *c*-axis, looks like "triangular lattice of triangles"



R3m: non-centrosymmetric





Triangles decorate corners of a rhombohedral cell: **fully connected 3D structure**

within a triangle: J_1

between triangles: J_2

$$H = \sum_{n.n.} J_1 \vec{S}_i \cdot \vec{S}_j + \sum_{n.n.n.} J_2 \vec{S}_i \cdot \vec{S}_j$$

Modaressi, Courtois, Gerardin, Malaman, Gleitzer, *J. Sol. State Chem.*, (**1983**) 47, 245–255.

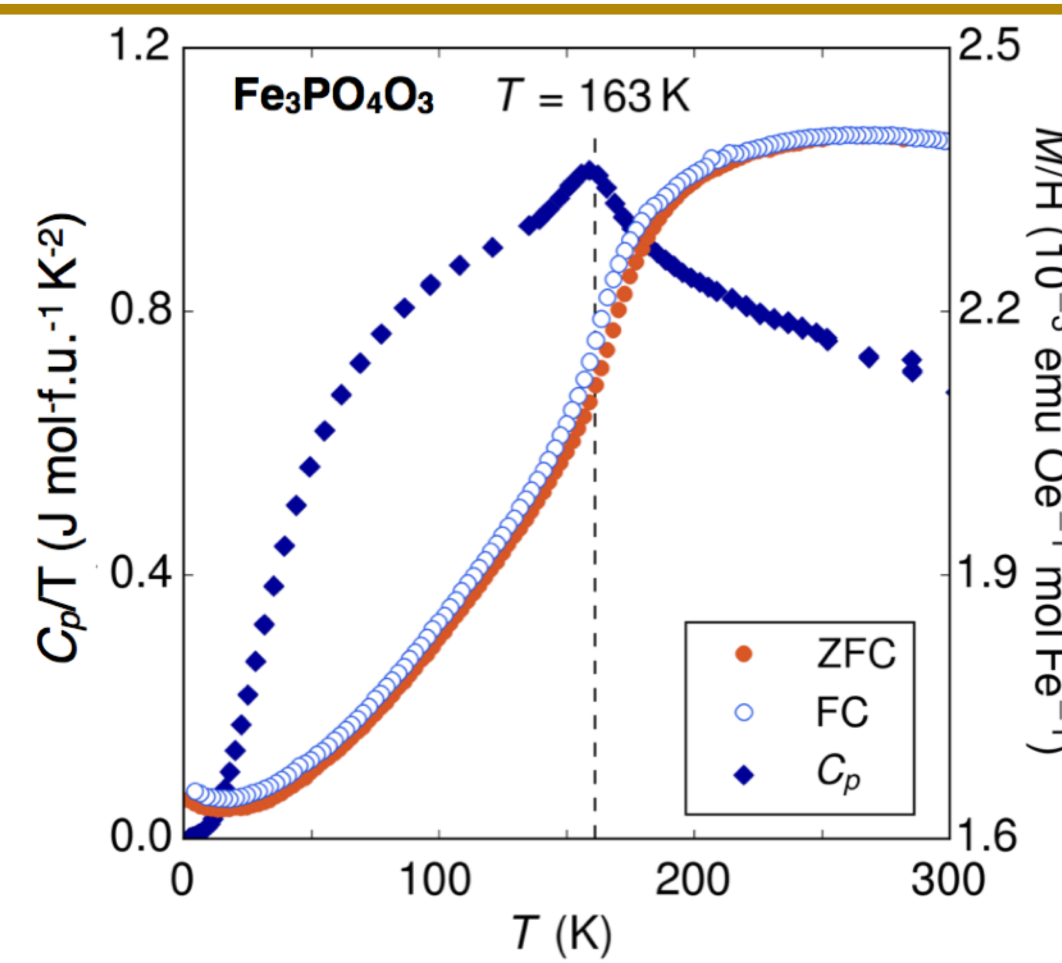
Frustration and Antiferromagnetic Order in $Fe_3PO_4O_3$

Antiferromagnetic order: peak in the specific heat and drop in the susceptibility; T_N ~ 163 K.

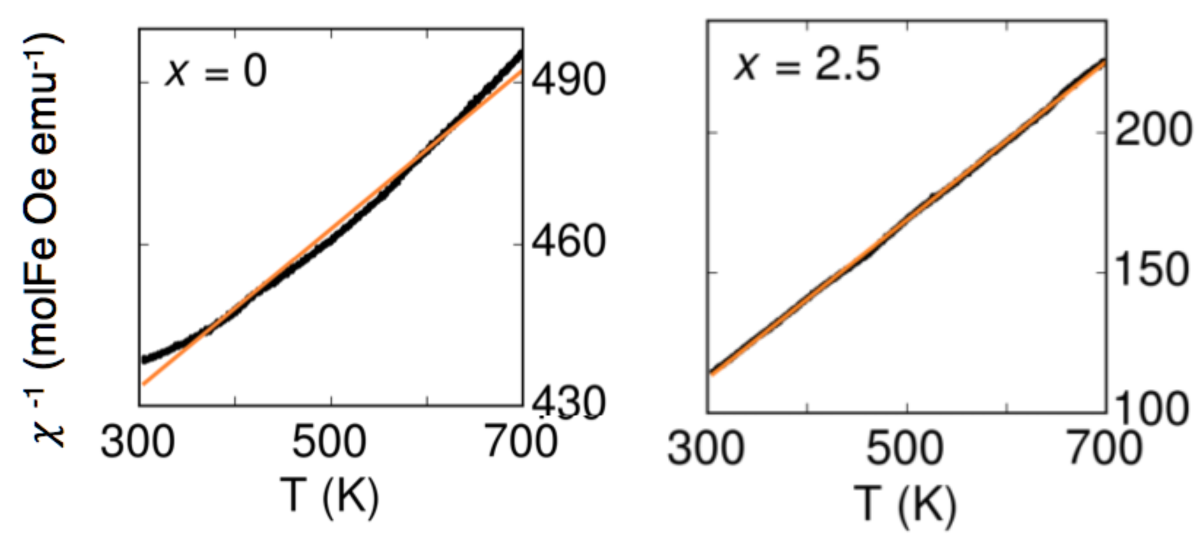
Frustration index:
f > 900 K / 163 K = 5.52

J1 > 319 K

Very strong magnetic interactions and frustration.

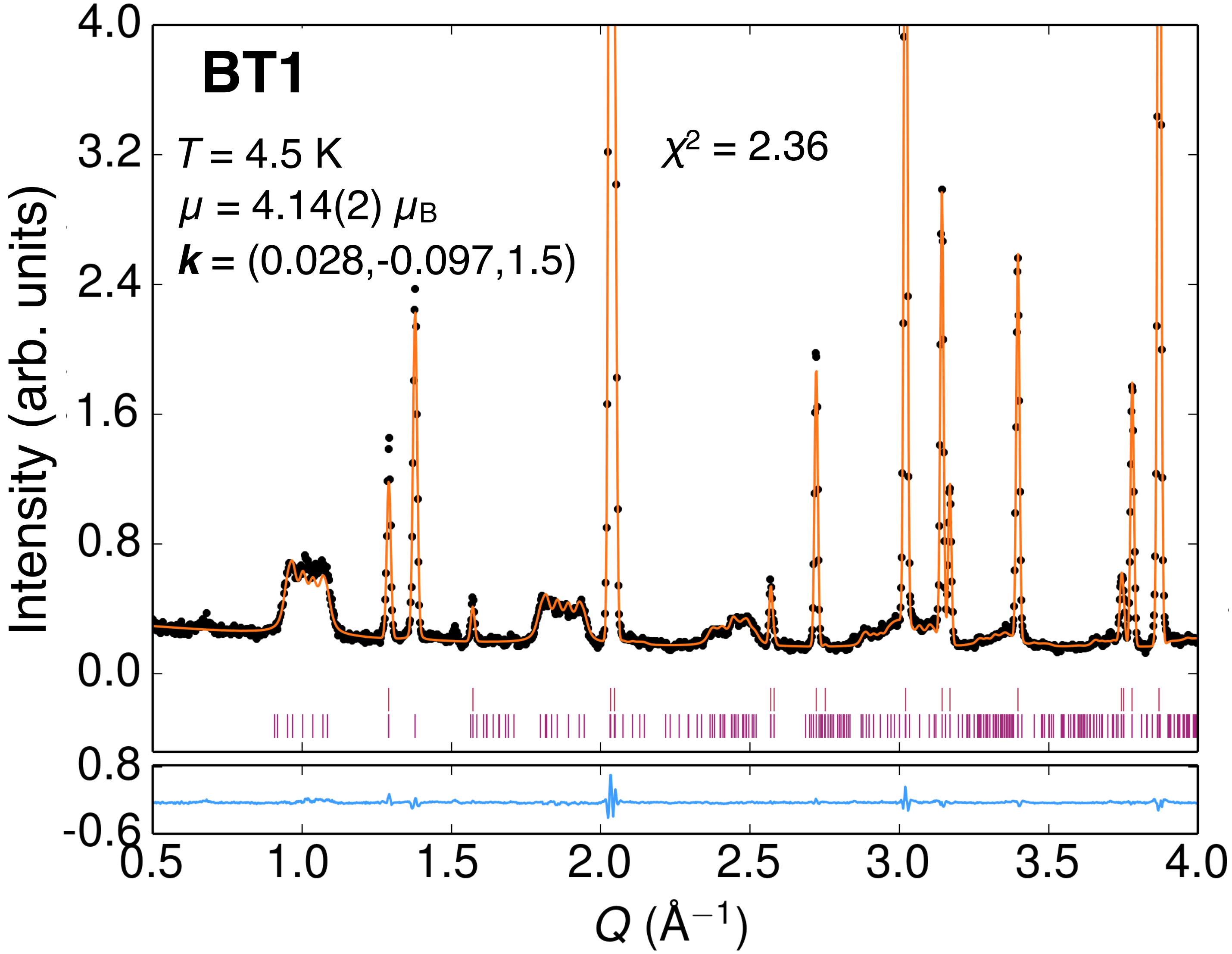


Ross, Bordelon, Terho, Neilson, PRB **92**, 134419 (2015)

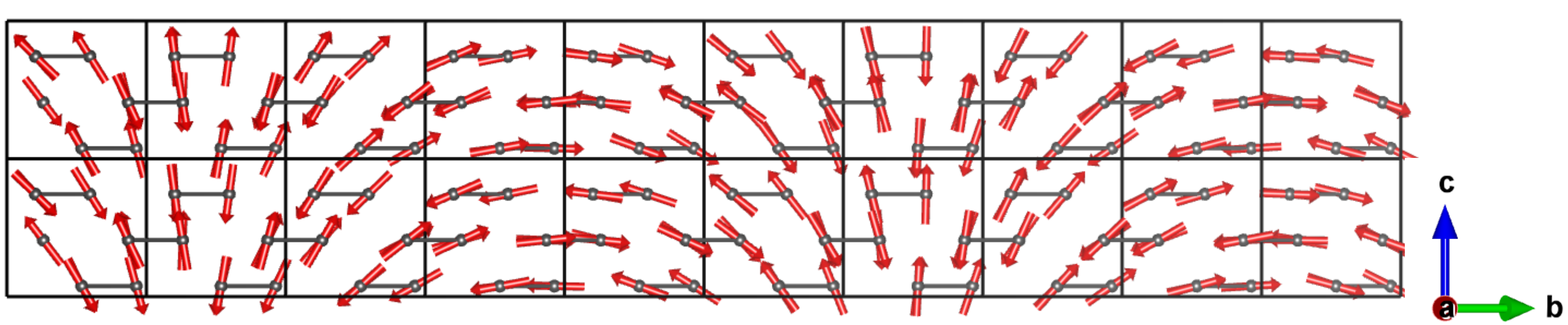


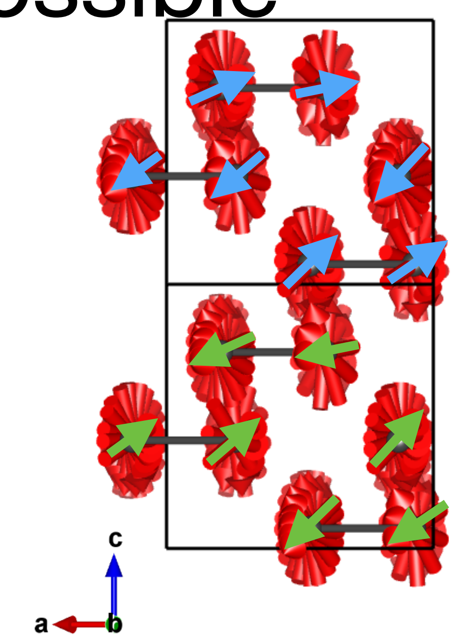
$Fe_{3-x}Ga_xPO_4O_3$

Antiferromagnetic structure…



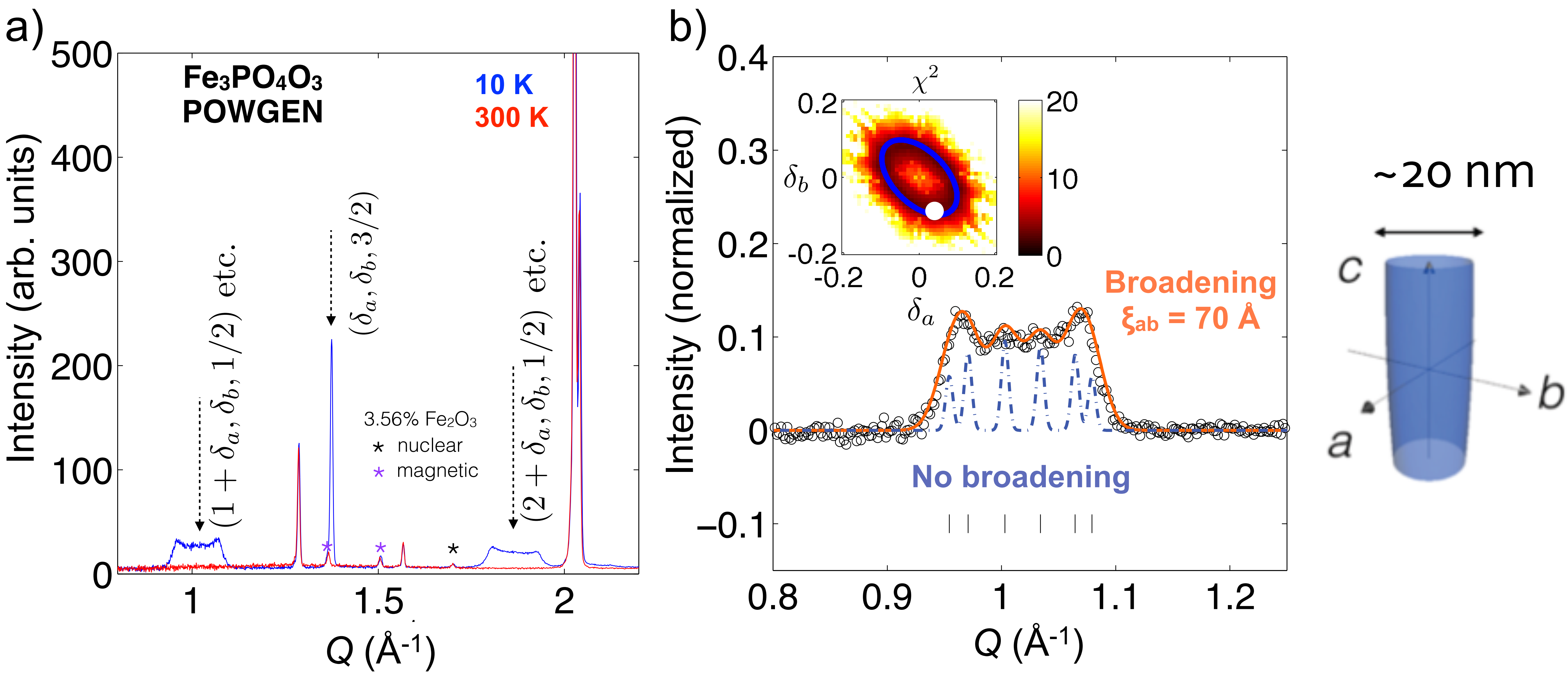
- Helical structure with perfect cancellation of moments along *c*-axis ("AFM Helical")
- λ= 86 Å
- k_{ord} = ($\boldsymbol{\delta}_a$, $\boldsymbol{\delta}_b$, 1.5)
- 6 symmetry-related *k* vectors possible





Ross, Bordelon, Terho, Neilson, PRB **92**, 134419 (2015)

Finite size broadening required in *ab* plane

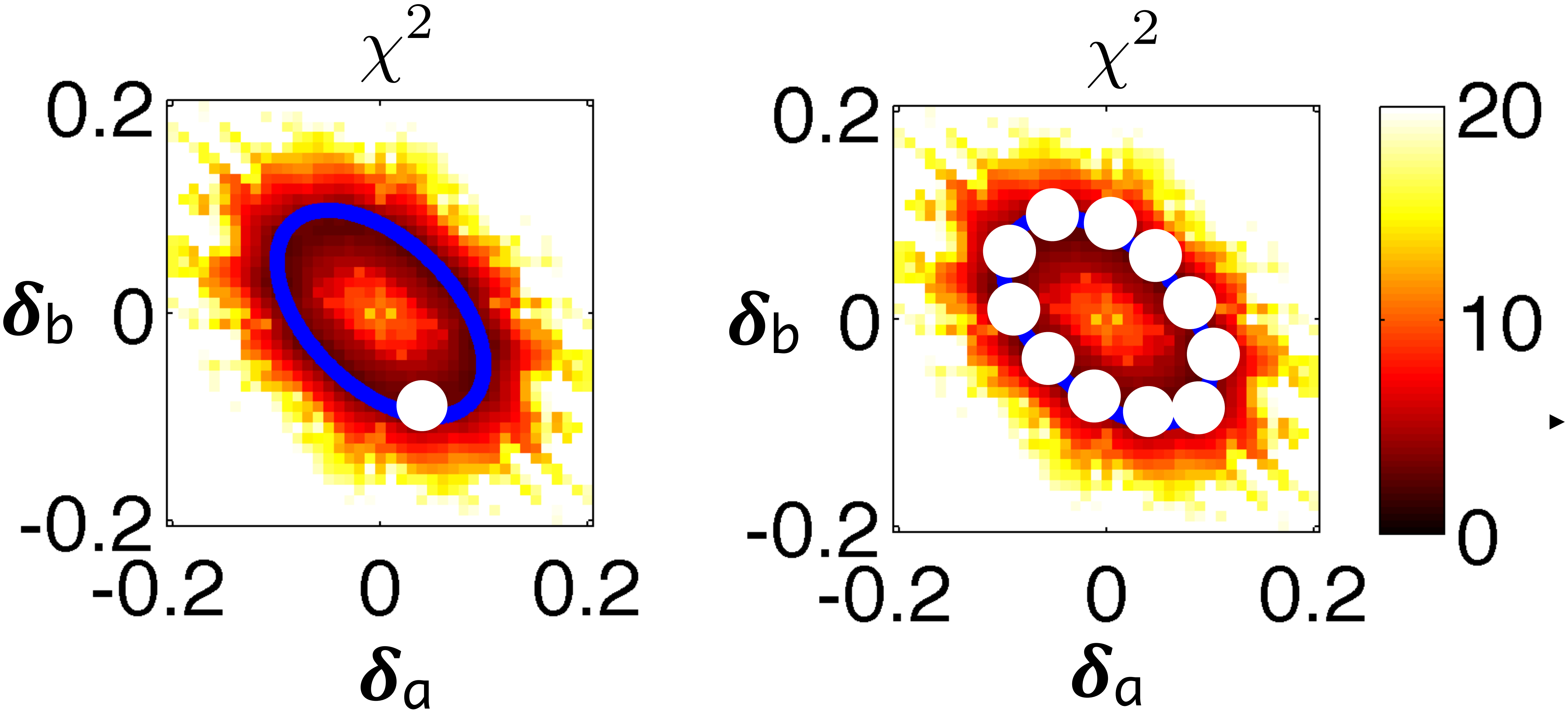


Need finite size broadening in *ab* plane only: needle domains

Ross, Bordelon, Terho, Neilson, PRB **92**, 134419 (2015)

What if it is actually a circular structure factor?

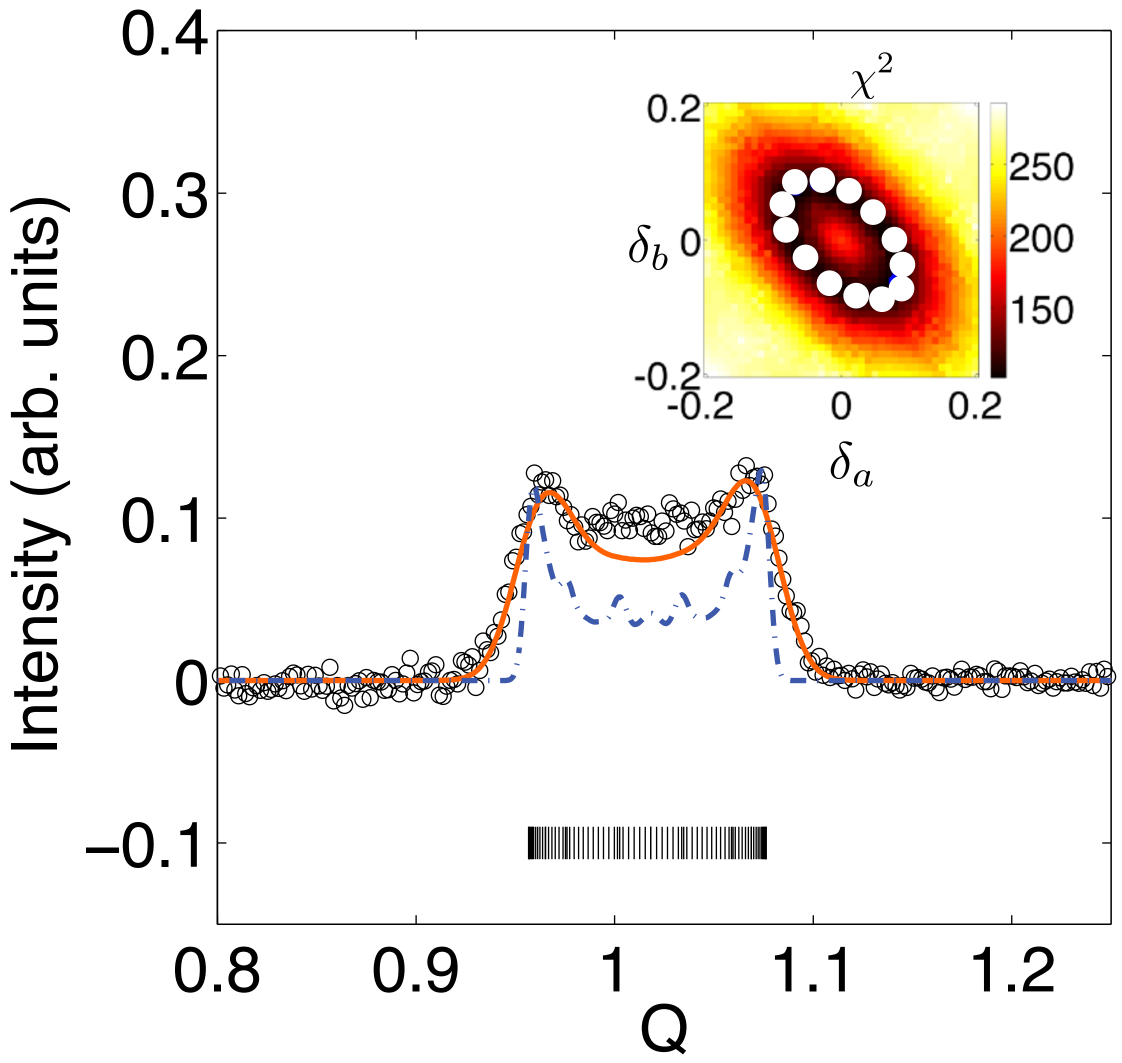
$k_{ord} = (\boldsymbol{\delta}_a, \boldsymbol{\delta}_b, 1.5)$



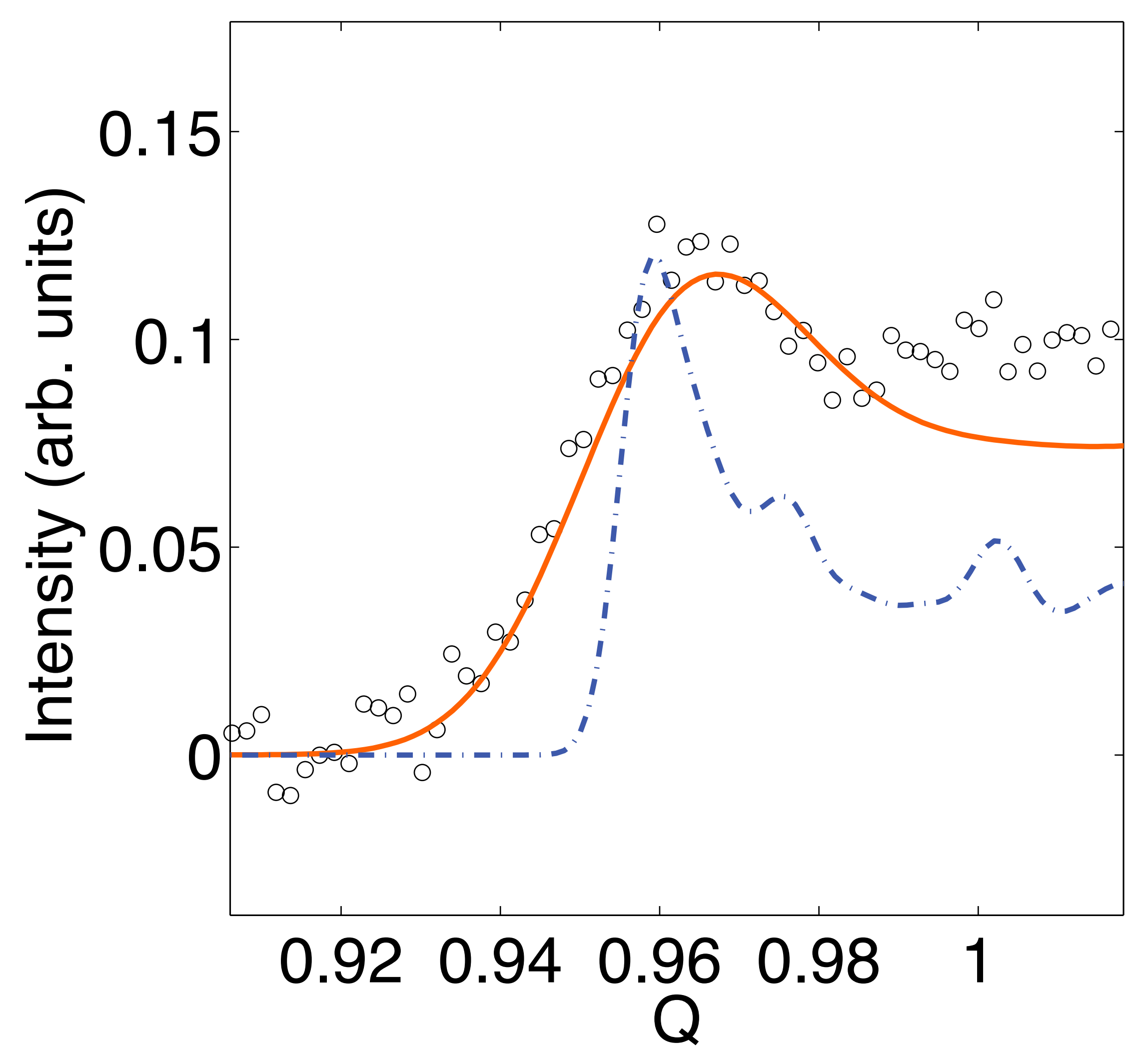
Broadening STILL required in *ab* plane

Assuming an **approximately circular structure factor**, still need broadening in *ab* plane

POWGEN data

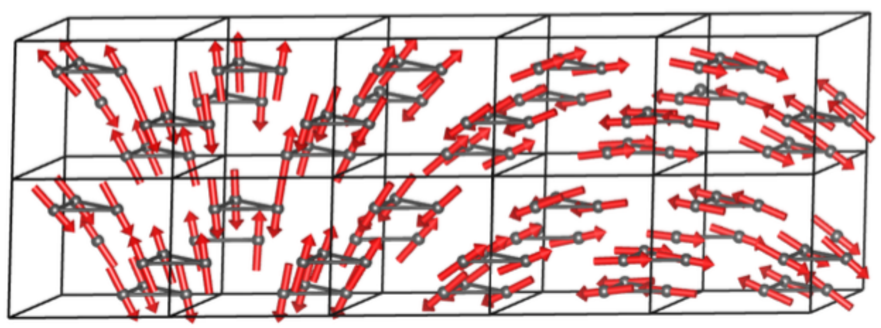


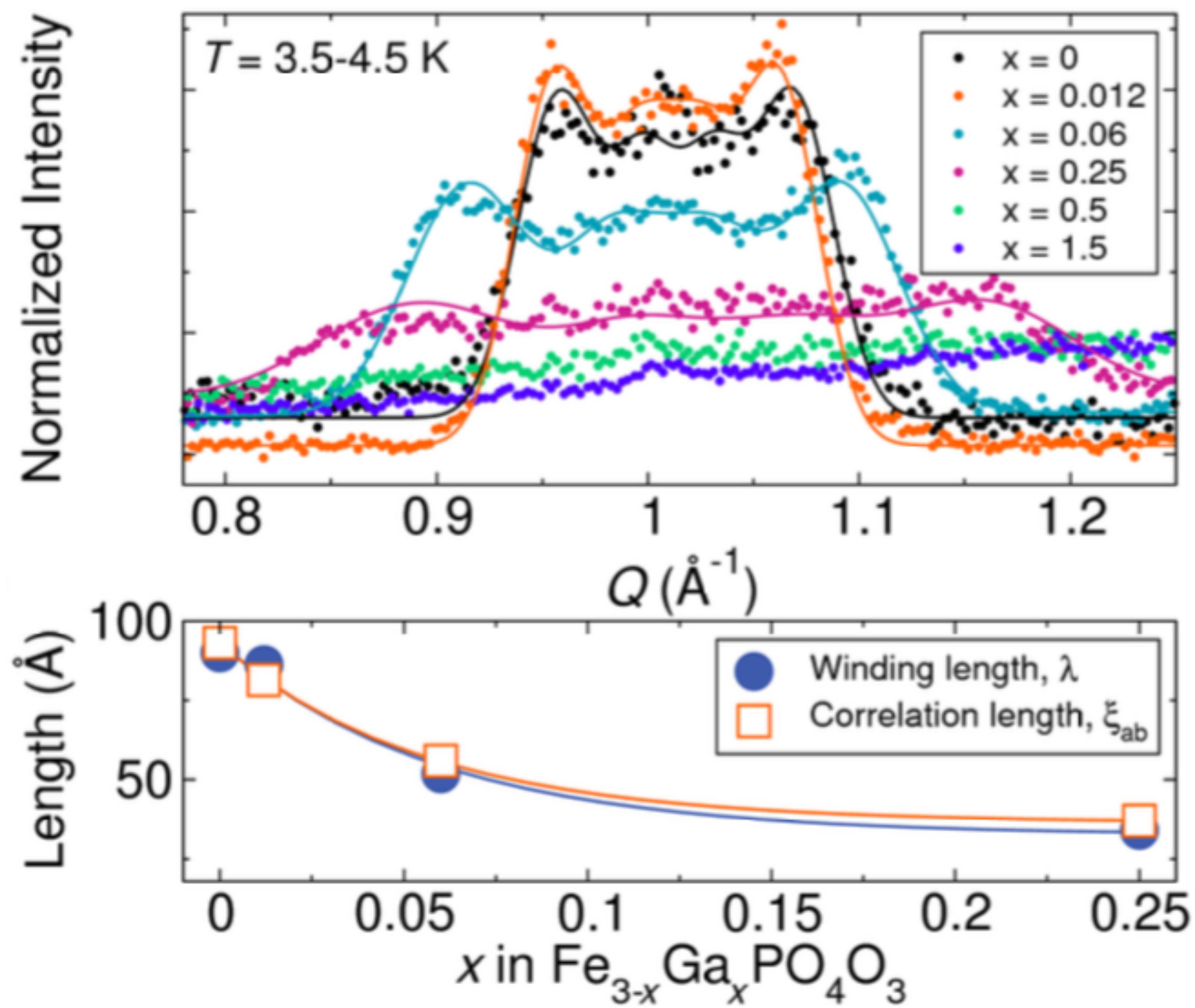
zoom in on edge



Correlation length and pitch length are connected!

Magnetic Dilution: $Fe_{3-x}Ga_xPO_4O_3$



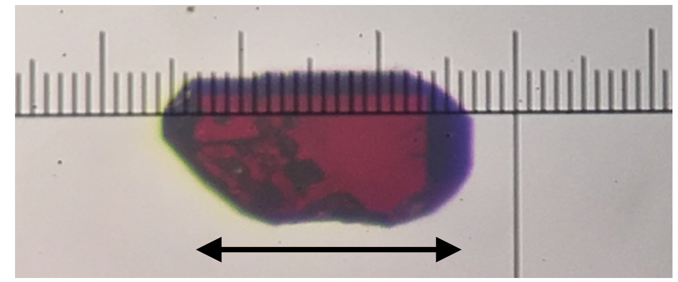


Helical pitch and "domain radius" are tracking each other! What could cause this?

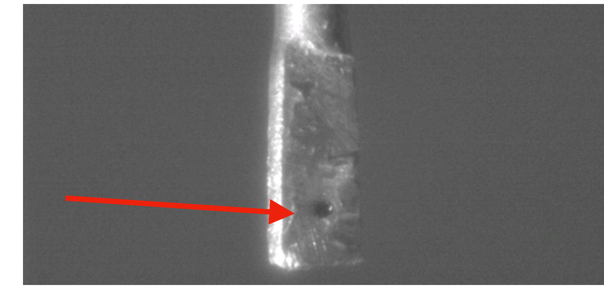
Tarne, Bordelon, Calder, Neilson, Ross, PRB **96** 214431 (2017)

Neutron Diffraction: single crystals

- Crystals grown by chemical vapor transport by Neilson group at CSU
- But they are *tiny*: need huge neutron flux, 2D area detector, and a focusing monochromator



0.33 mm

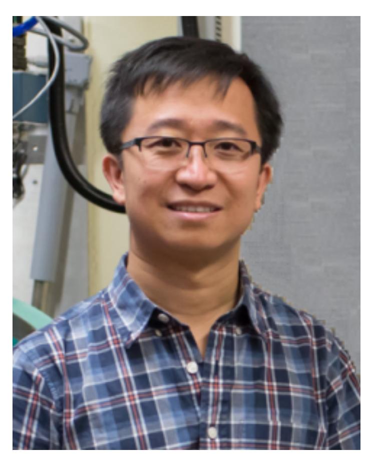


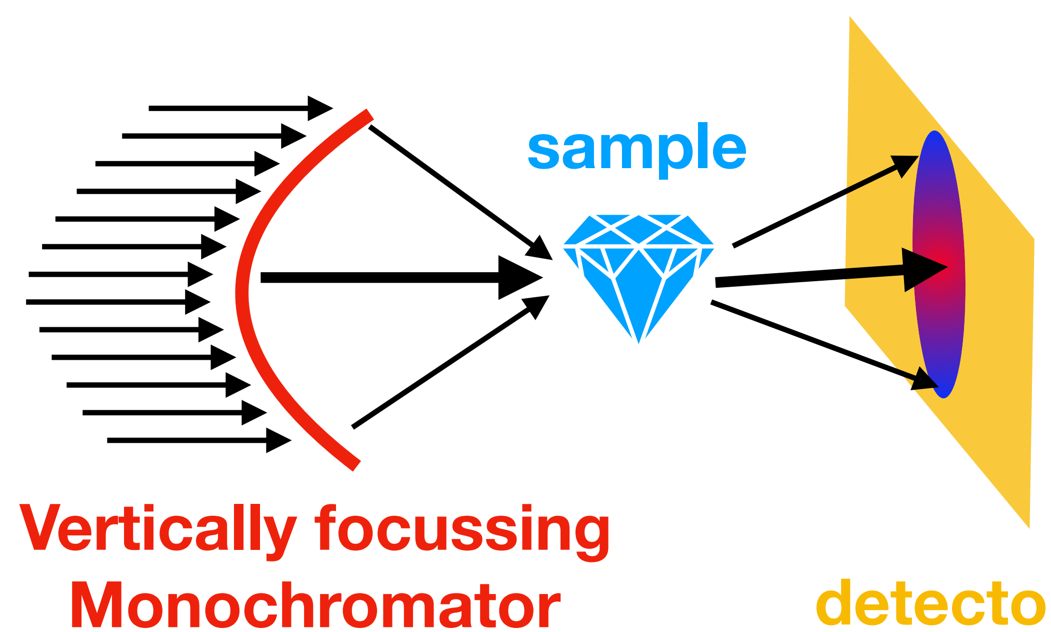
Jamie Neilson

Michael Tarne

HB-3A at High Flux Isotope Reactor (HFIR)

Huibo Cao

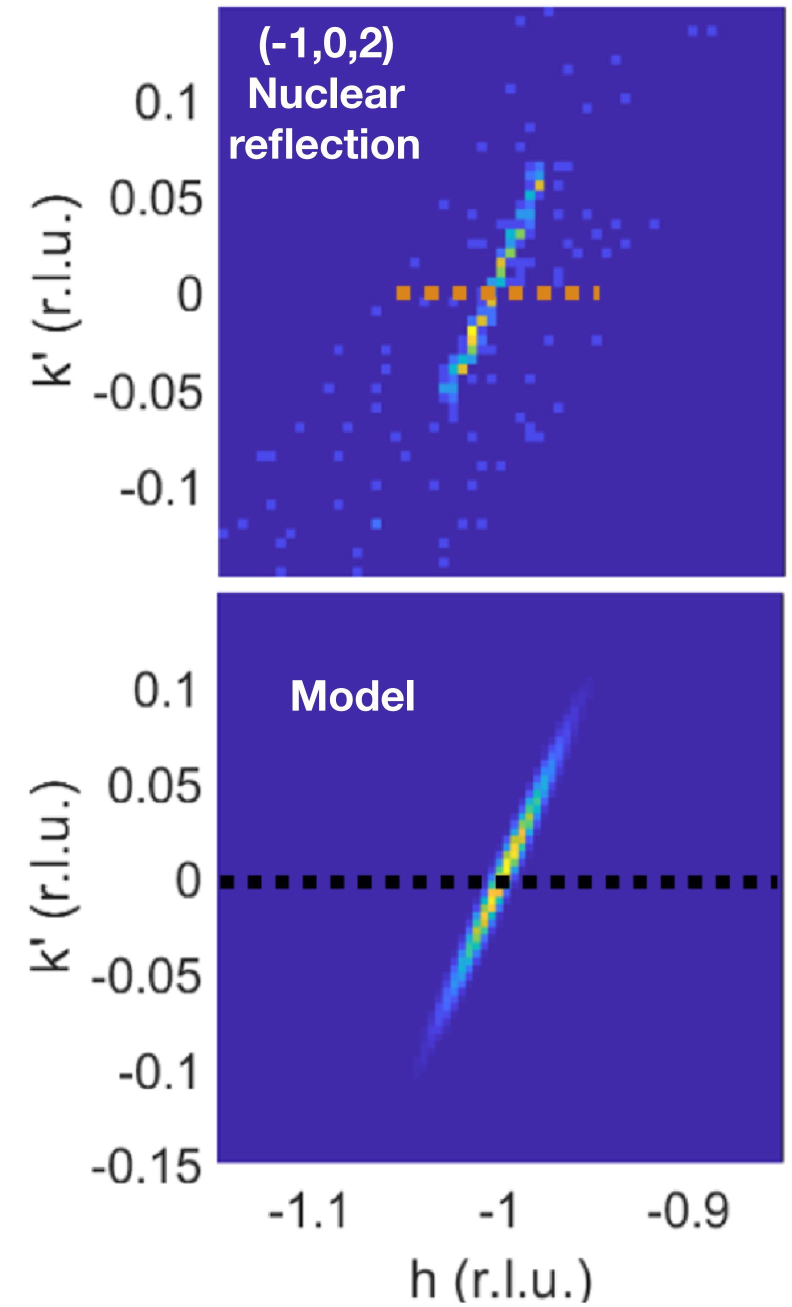


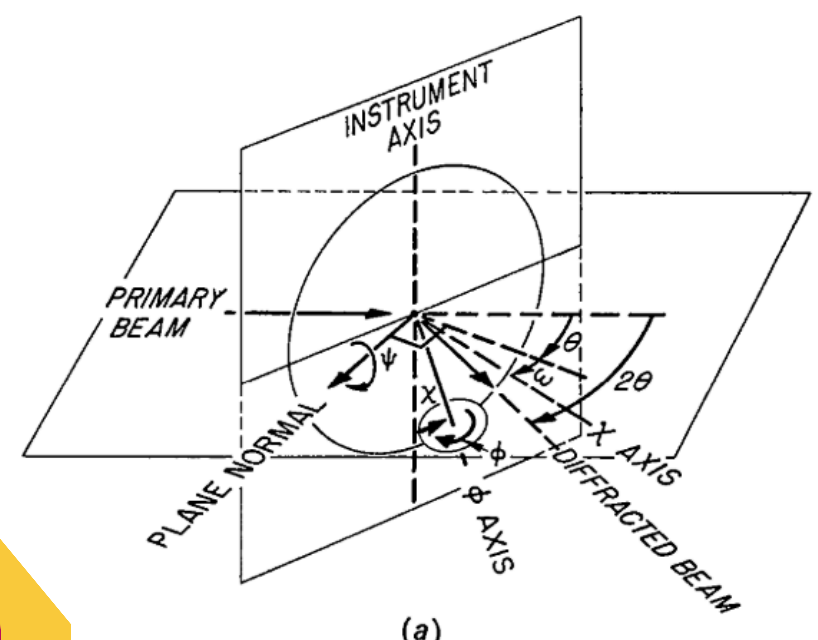


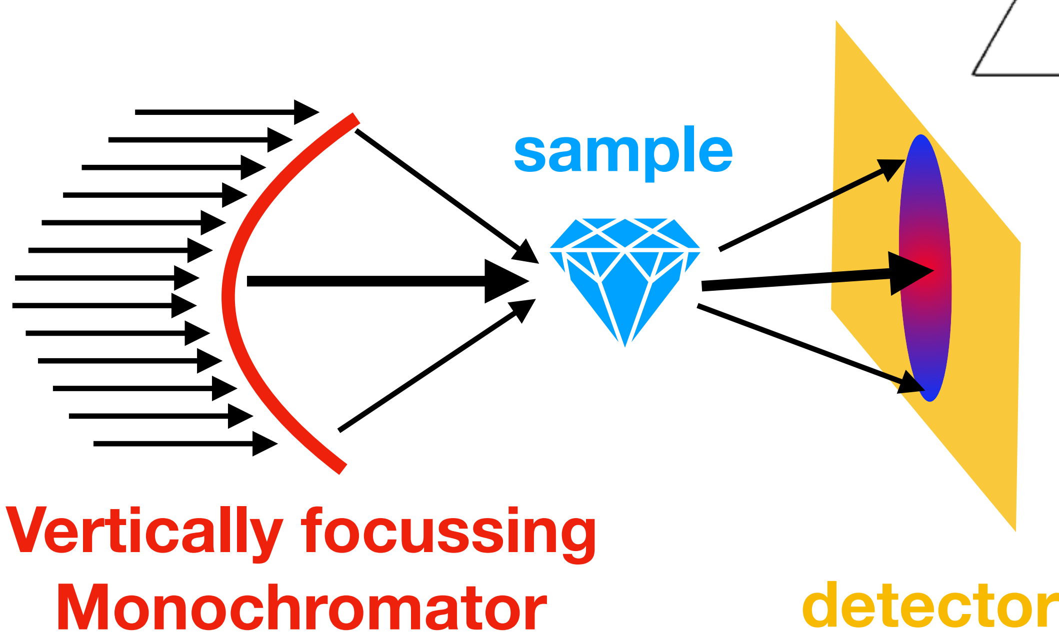
Modeling resolution function: key to success!

- Vertical focusing/divergence creates a very elongated ellipsoid for resolution function
- Resolution function rotates into hkl differently depending on specific measurement conditions

Colin Sarkis







Acta Cryst. (1967). **22**, 457

Angle Calculations for 3- and 4- Circle X-ray and Neutron Diffractometers*

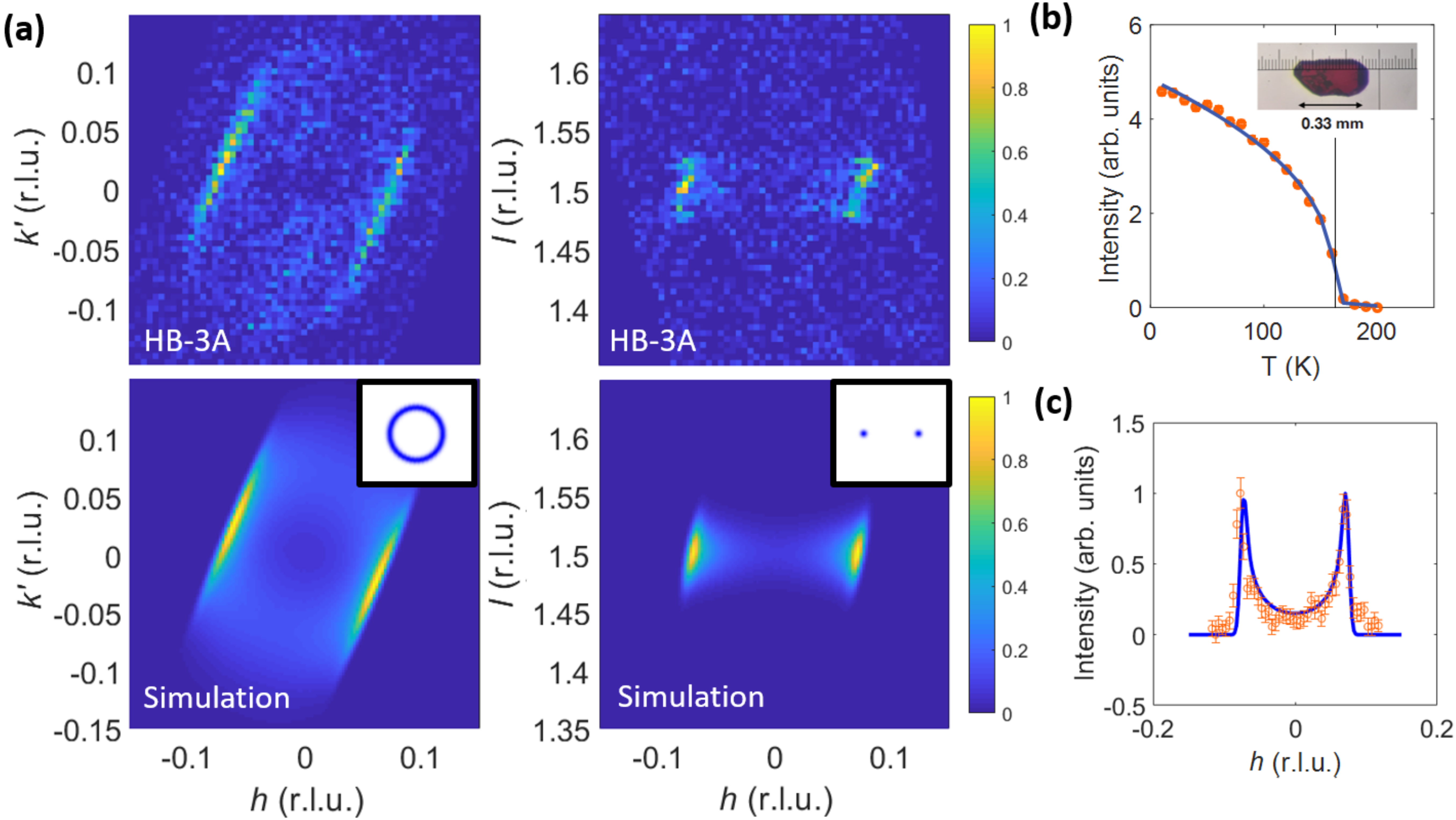
By William R. Busing and Henri A. Levy

Chemistry Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830, U.S.A.

(*Received* 13 *June* 1966)

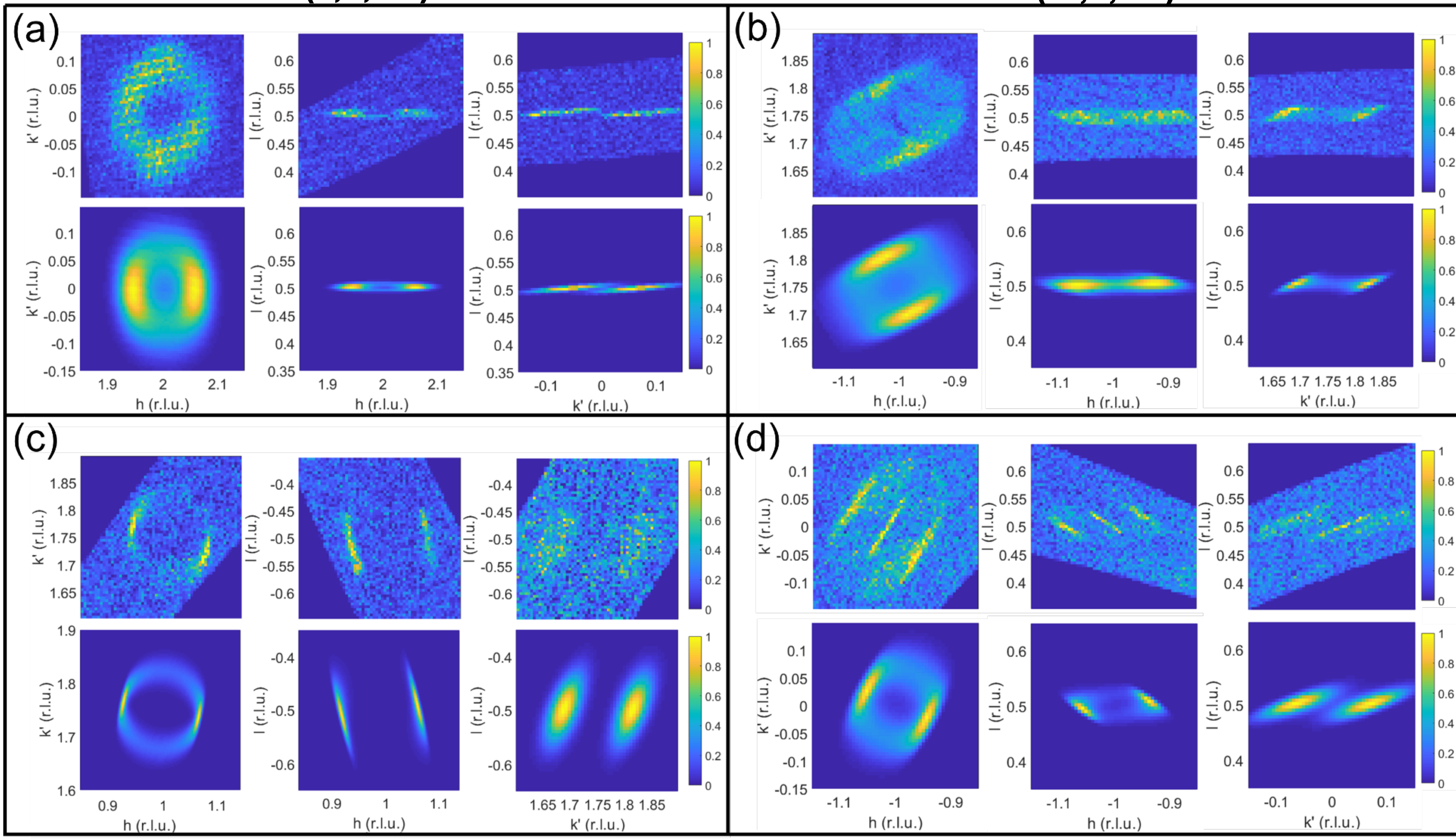
"Incommensurate Rings" in Structure Factor: Helical modulation with no preferred direction

Incommensurate ordering wave vector ring around (0,0,1.5): λ = 98 ± 12 Å



"Incommensurate Rings” in Structure Factor: Helical modulation with no preferred direction

(2,0,0.5) (-2,2,0.5)



(0,2,-0.5) (-2,2,0.5)

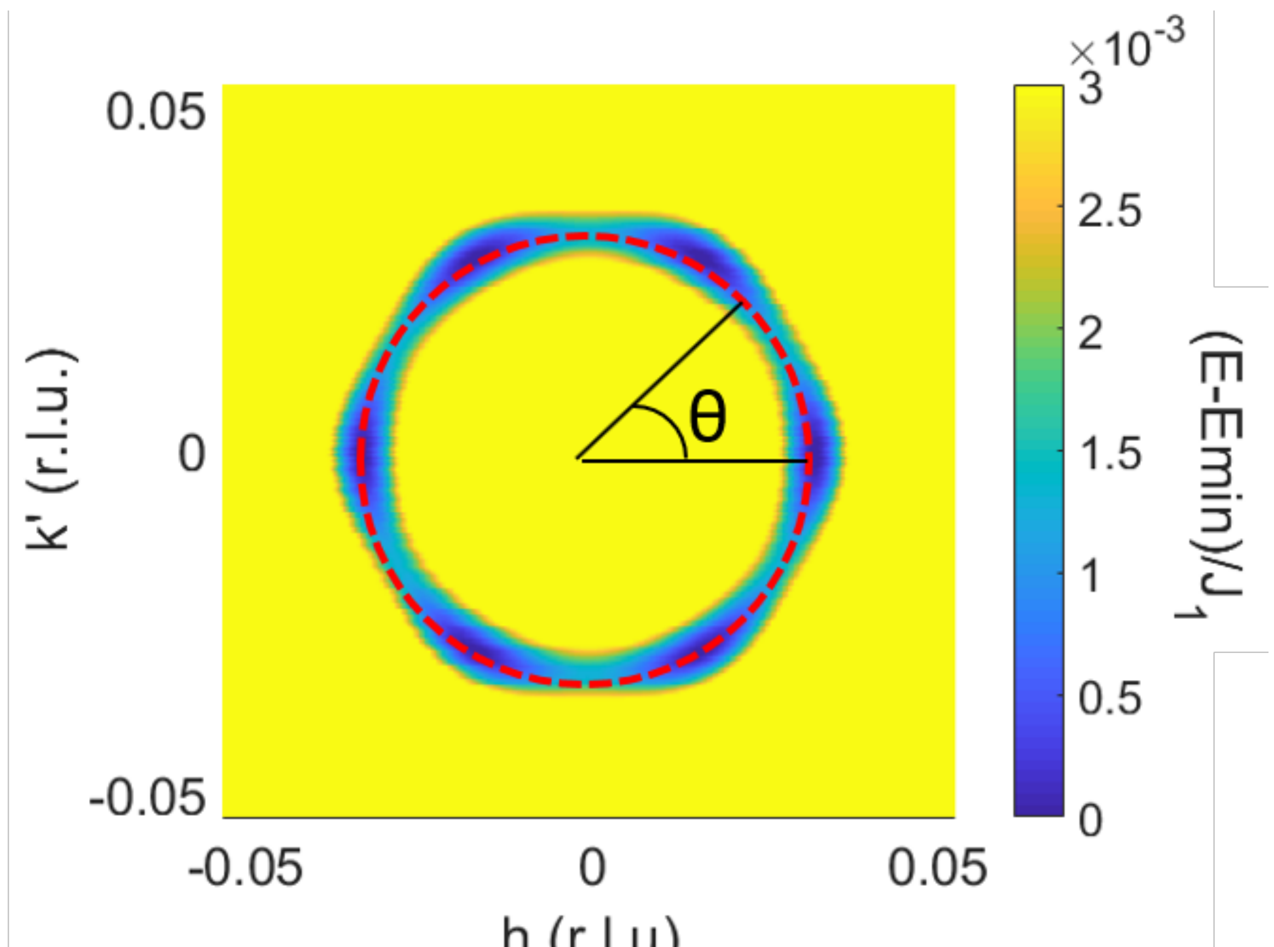
Quasi-degenerate ring of *k*-vectors in the frustrated Heisenberg model

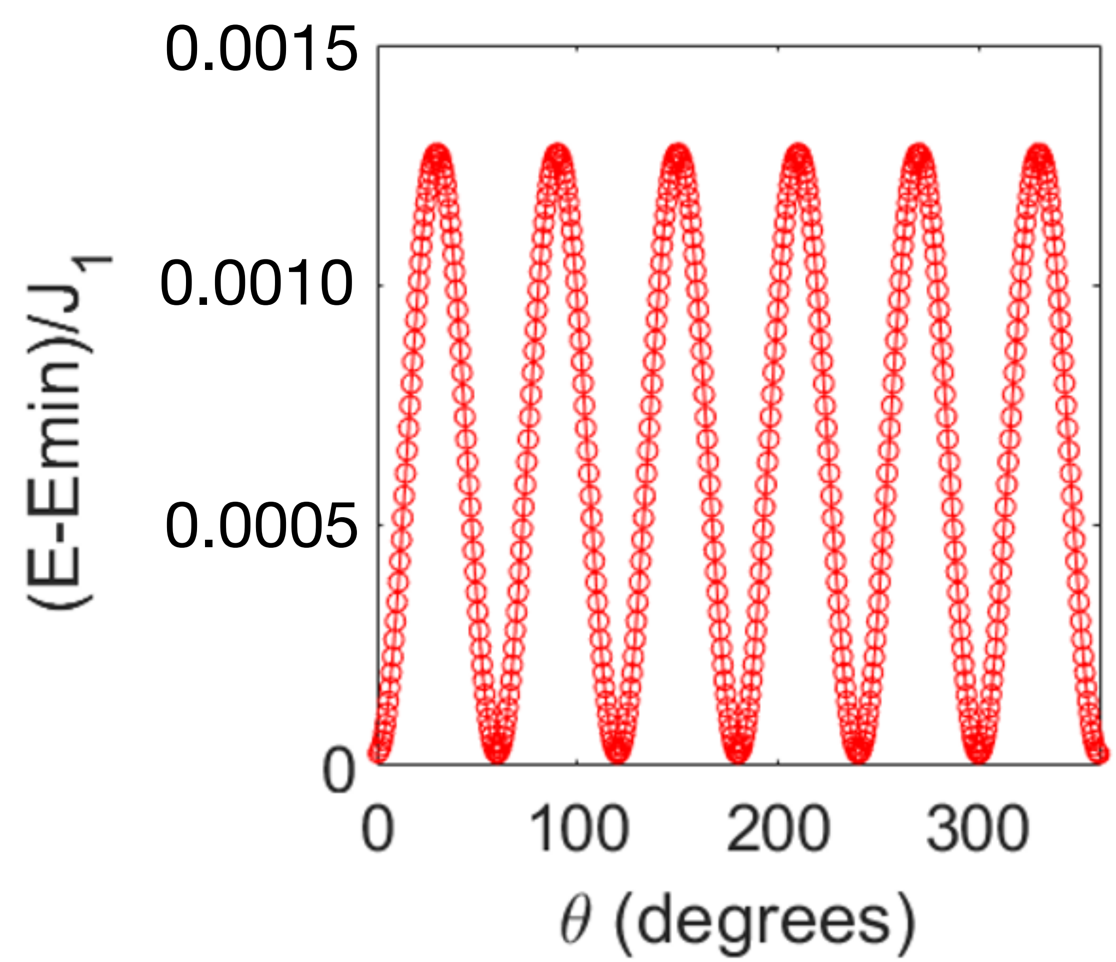
Ethan Coldren

$$H = \sum_{n.n.} J_1 \vec{S}_i \cdot \vec{S}_j + \sum_{n.n.n.} J_2 \vec{S}_i \cdot \vec{S}_j$$

Marty Gelfand

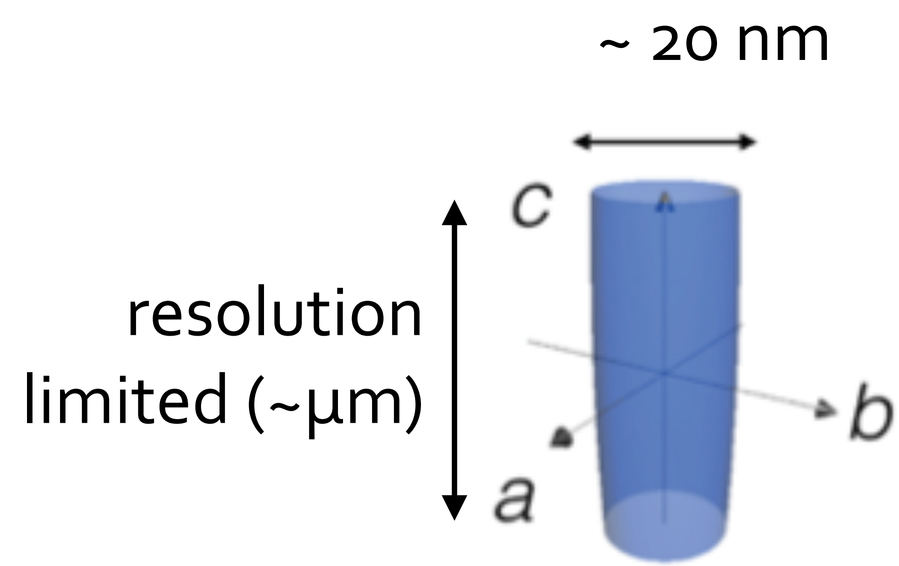
Luttinger-Tisza shows that energetic variation around the ring is ~400 mK (aka TINY!) for J_1 ~ 319 K and J_2/J_1 ~ 1.9, appropriate for for $Fe_3PO_4O_3$

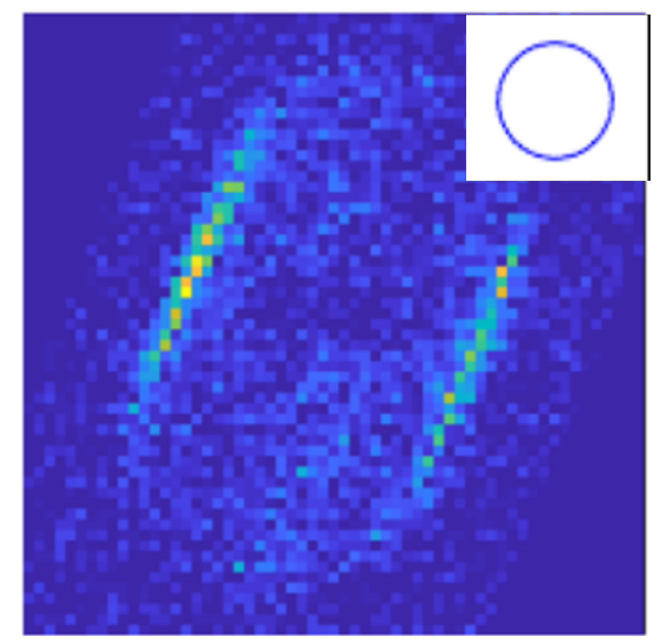


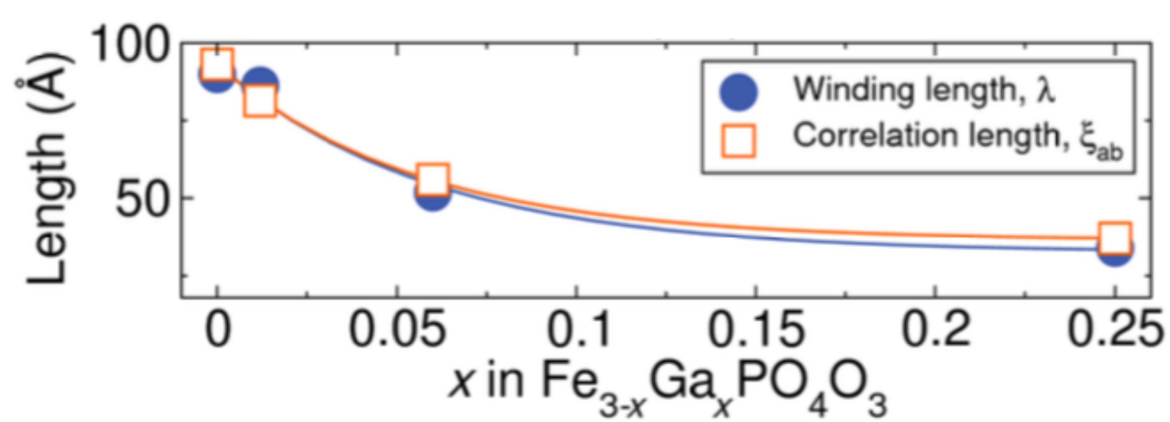


So what do we have?

- $Fe_3PO_4O_3$ has **needle-like domains of antiferromagnetic helical (partial) order**
- **The helical order does not have a preferred modulation direction:** (quasi-)degenerate ring of ordering wavevectors is observed, and is consistent with J_1 - J_2 frustrated Heisenberg model
- **Correlation length in ab plane *tracks* helical pitch length**







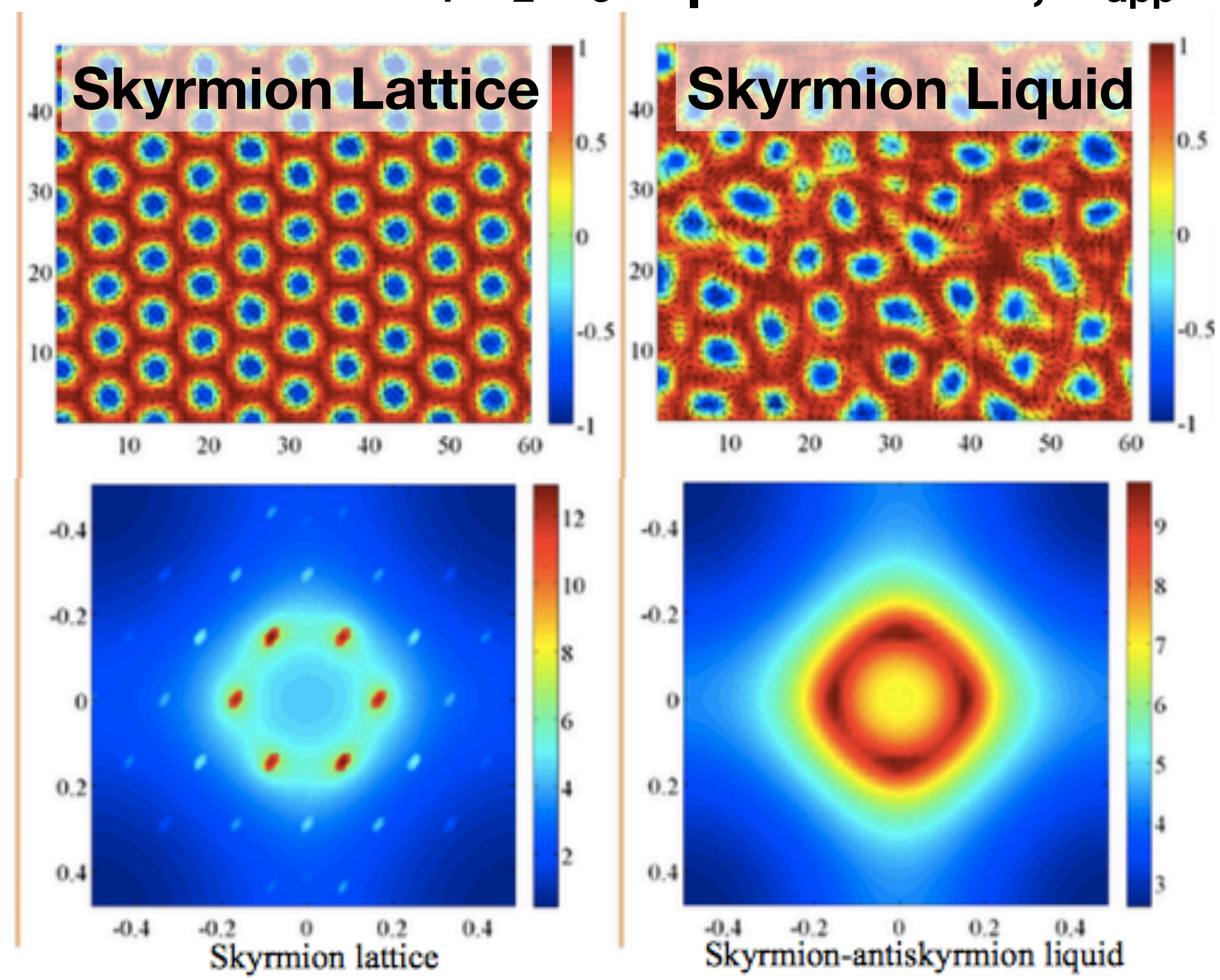
Skyrmion-like liquid / glass in $Fe_3PO_4O_3$?

- "Skyrmion liquids" can be seen above ordering transitions
- **Skyrmion-Skyrmion distance** is the same as in the Skyrmion lattice phase, i.e. **set by helical pitch length**

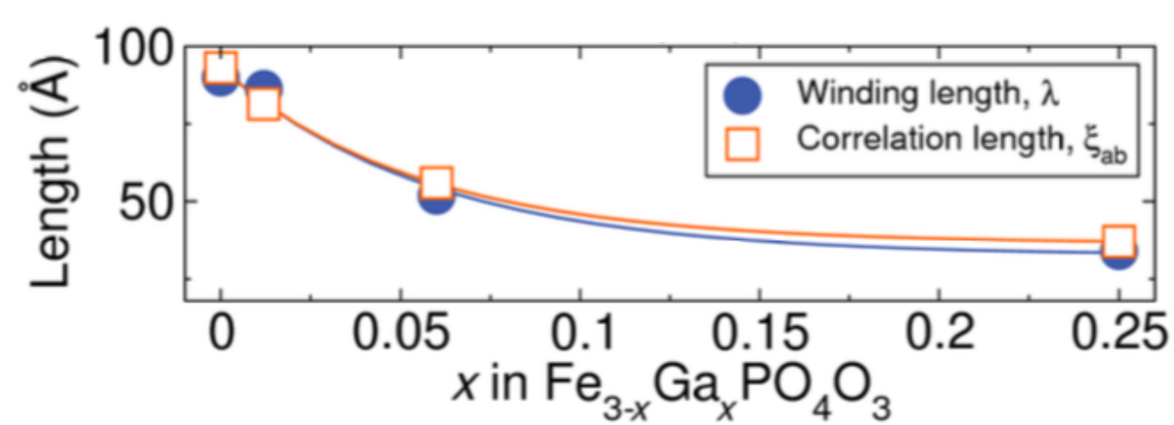
Monte Carlo: J_1-J_2-J_3 Square Lattice, H_{app}

Spin configuration

Spin Structure factor

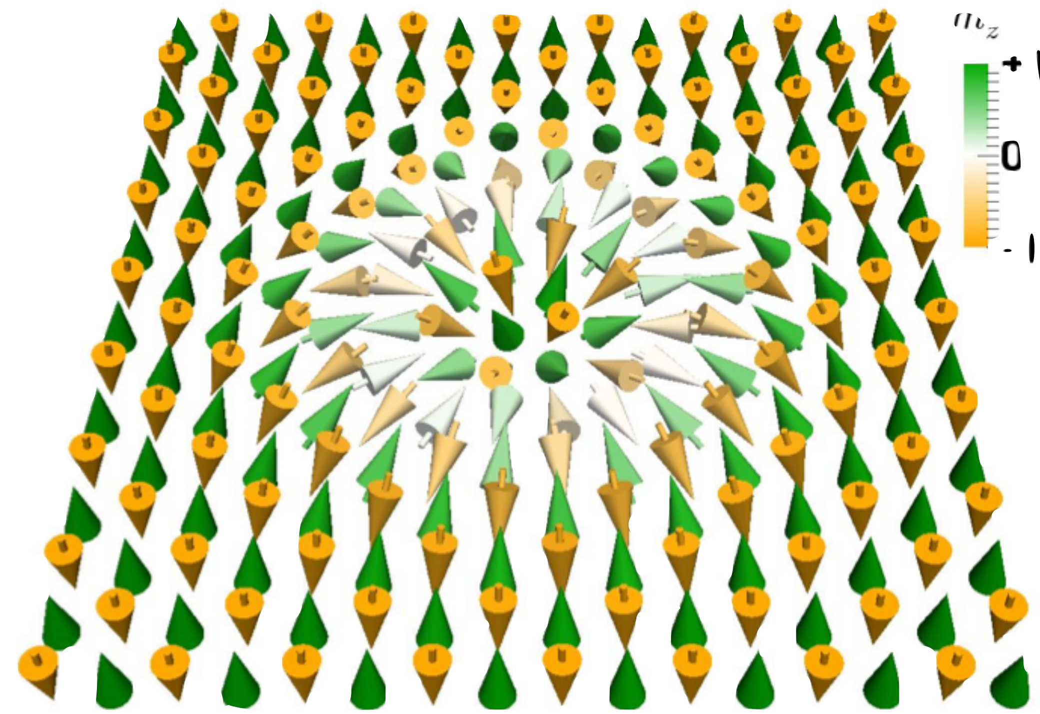


S.Z. Lin, S. Hayami, PRB 93 064430 (2016)

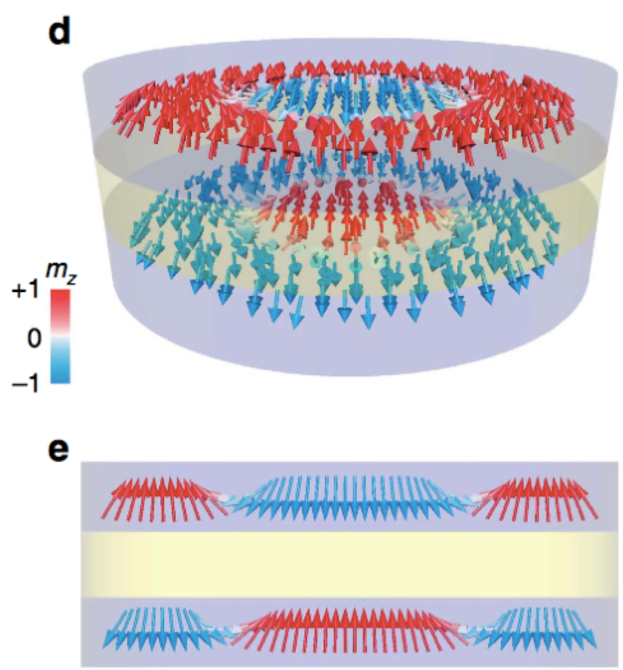


Locally Antiferromagnetic Skyrmions

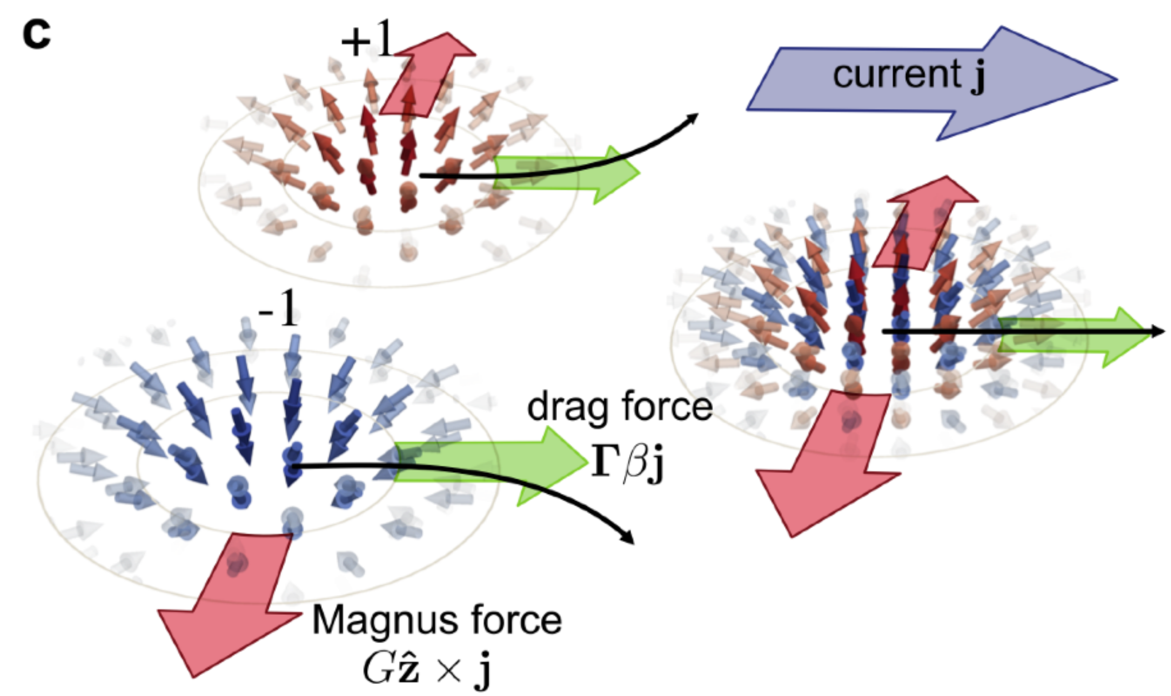
- locally compensated moments in a single layer or as “bilayers”
- Stable against stray fields (Magnus force), smaller — better for information processing
- Recently made in a synthetic antiferromagnet: Legrand et al, Nat. Mater. 2019



Zhang, Zhou, Ezawa, *Sci. Rep.* **6**, 24795 (2016)



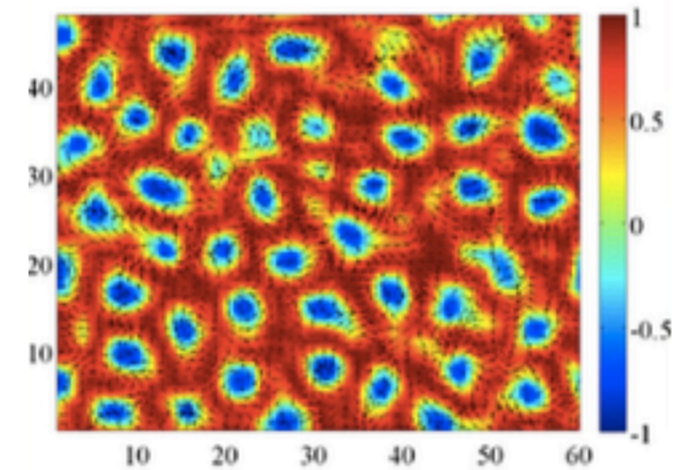
X. Zhang et al, Nat.Comm, **7**,102932016 (2016)



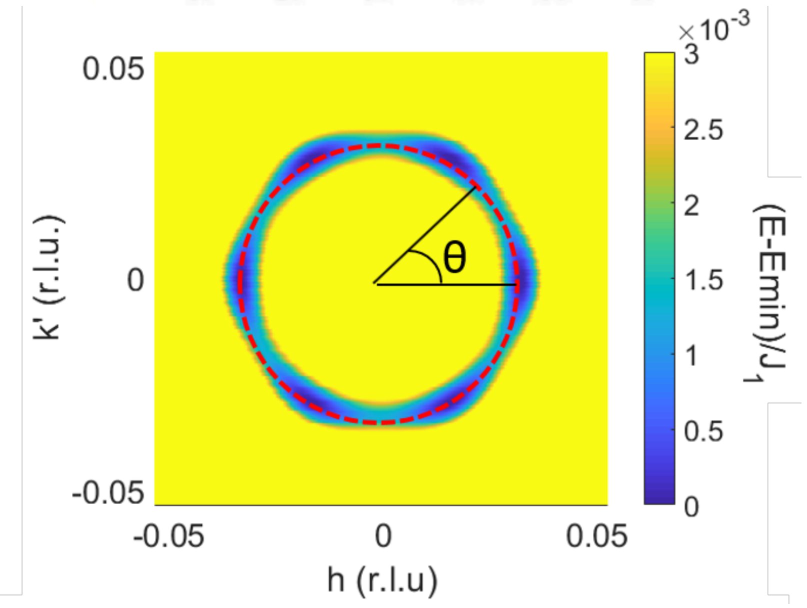
Barker, Tretiakov, PRL 116, 147203 (2016)

Open questions in $Fe_3PO_4O_3$

- If $Fe_3PO_4O_3$ displays a (frozen?) AFM Skyrmion-like phase (or something like Blue phase), how are the spin textures / defects stabilized?



- What role does the quasi-degeneracy of the frustrated J_1-J_2 model play in generating these textures? What role does the DM interaction play?



- **How to see the spin structure in real space?** NOTE: compensated moments, no net field!

- What are the *dynamics* of this weird magnetic phase? Can it be manipulated with e.g. thermal gradients, electric fields?

Acknowledgements

K. A. Ross, et al. Phys. Rev. B **92**, 134419 (2015)

M. J. Tarne, et al. Phys. Rev. B 96, 214431 (2017)

C. Sarkis, et al. arXiv:1910.08818 [cond-mat.str-el] (2019)

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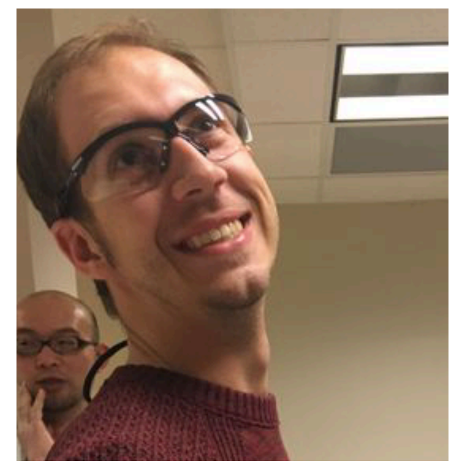

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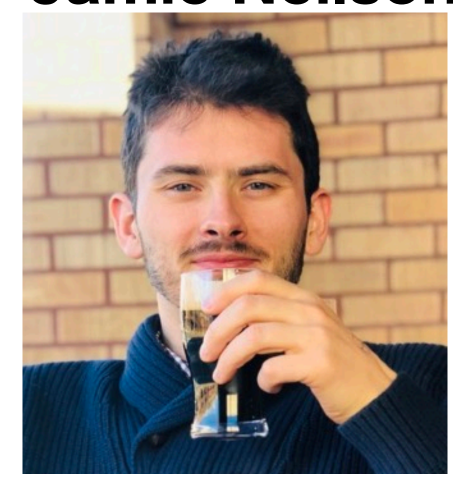

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