Magnetic structure determination from the combined use of representation analysis and magnetic crystallographic symmetry

Vladimir Pomjakushin
Laboratory for Neutron Scattering and Imaging, LNS, Paul Scherrer Institute
Plan

• Introduction to Representational & magnetic symmetry ways of description
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• Morden trends in magnetic structure determination from neutron diffraction ND. Advantages of the combined use of Representation Analysis RA and magnetic subgroups: Shubnikov or 3D+1
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• Morden trends in magnetic structure determination from neutron diffraction ND. Advantages of the combined use of Representation Analysis RA and magnetic subgroups: Shubnikov or 3D+1

• Examples: multiferroic TmMnO$_3$, pyrochlore Tm$_2$Mn$_2$O$_5$
Two ways of description of magnetic structures

Magnetic structure is an axial vector function \( S(r) \) defined on the discreet system of points (atoms), e.g. \( S(r) = s(r_1) \oplus s(r_2) \oplus s(r_3) \oplus s(r_4) \)

Crystal with space group \( G \)

1. How to make \( S(r) \) invariant? Find (new) symmetry elements. \( g_{\text{new}} S(r) = S(r) \) to itself, where \( g_{\text{new}} \in G_{\text{sh}} \) subgroup of PG paramagnetic space group: \( PG=G \otimes 1' \), where \( 1'=\text{spin/time reversal} \), \( G \) (parent space group).
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   or

2. How should $S(r)$ be transformed under elements of $G$?
   $g S(r) = S_{\text{new}g}(r)$ to different functions for each $g \in G$
Two ways of description of magnetic structures

1. Magnetic or Shubnikov groups MSG. Historically the first way of description (Landau, Lifshitz 1951). \( S(r) \) invariant under the Shubnikov subgroup \( G_{sh} \) of \( G \otimes 1' \) (1'=spin/time reversal).

Identifying those symmetry elements that leave \( S(r) \) invariant.

The MSG symbol looks similar to SG one, e.g. \( I4/m' \)

1. How to make \( S(r) \) invariant?
\[ gS(r) = S(r) \] to itself, where \( g \in \text{subgroup of PSG paramagnetic space group: PSG} = SG \otimes 1' \), where 1'=spin/time reversal, SG (parent space group).

2. How should \( S(r) \) be transformed?
\[ gS(r) = S_{\text{new}}(r) \] to different functions for each \( g \in SG \)
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MSG Example:

<table>
<thead>
<tr>
<th>MSG</th>
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<tbody>
<tr>
<td>87.1.733</td>
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<td>$I_p 4/m$</td>
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2. Representation analysis. (Bertaut 1967) $S(r)$ is transformed to $S^i(r)$ under $g \in G$ (parent space group) according to a single irreducible representation $\tau_i$ of $G$.

Identifying/classifying all the functions $S^i(r)$ that appears under all symmetry operators of the same space group $G$

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*each group element $g \rightarrow$ matrix $\tau(g)$
Two ways of description of magnetic structures

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Irrep Example:
\( I4/m \), \( k=0 \) has 8 1D irreps \( \tau_1, \ldots \tau_8 \).

<table>
<thead>
<tr>
<th>( \tau, \psi )</th>
<th>( h_1 )</th>
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<tr>
<td>( \tau_2 )</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>-1</td>
<td>-1</td>
<td>-1</td>
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</tr>
<tr>
<td>( \tau_3 )</td>
<td>1</td>
<td>-1</td>
<td>1</td>
<td>-1</td>
<td>1</td>
<td>i</td>
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</tr>
<tr>
<td>( \tau_4 )</td>
<td>1</td>
<td>-i</td>
<td>1</td>
<td>i</td>
<td>1</td>
<td>-1</td>
<td>-i</td>
<td>1</td>
</tr>
<tr>
<td>( \tau_5 )</td>
<td>1</td>
<td>-1</td>
<td>i</td>
<td>1</td>
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*each group element \( g \) --> matrix \( \tau(g) \)

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<tr>
<td>2</td>
<td>\tau_2 \psi</td>
<td>1</td>
<td>4^+_z</td>
<td>2_z</td>
<td>4^-_z</td>
<td>-1</td>
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Magnetic space groups and representation analysis: competing or friendly concepts?

In 1960th-70th opposed

E. F. Bertaut, CNRS, Grenoble
Representation Analysis (RA)*

W. Opechovski, UBC, Vancouver
Shubnikov magnetic space groups

even until recent times RA was considered to be more powerful in neutron scattering community.*

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Currently > 2010-…

(Representation Analysis) and (Magnetic space groups) are complementary and **must** be used together to fully identify the magnetic symmetry.

“Old new” trends in magnetic structure determination from ND. Currently there is solid understanding that both RA and Shubnikov magnetic symmetry should be used together. Big progress in software tools during last 5 years in this way of analysis …

- In some cases this allows one to find a hidden symmetry, which is not evident from the representation analysis alone.
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- In some cases this allows one to find a hidden symmetry, which is not evident from the representation analysis alone.
- **Regular practice for crystal structure transitions:** This approach is routinely used by crystallographers in the analysis of crystal phase transition,
- **Magnetic transitions:** Usually, representation approach with a single arm and general direction of order parameter of propagation vector star. Possible high symmetry Shubnikov subgroups are lost.
IUCr Commission on Magnetic Structures

The Commission on Magnetic Structures (CMS) was established *ad interim* by the Executive Committee in January 2011 and confirmed at the Madrid General Assembly in August 2011. It’s primary purpose is to facilitate research on the discovery and communication of magnetic structures in magnetically ordered materials; and its present focus is to cultivate a community consisting of interested participants from diverse fields of research, who can establish standards for defining and communicating the crystallographic details of magnetic structures. The scope of the Commission’s consideration encompasses a broad range of magnetic structure types, including commensurate magnetic structures, modulated and otherwise aperiodic magnetic structures, low-dimensional magnetic structures, disordered magnetic structures, etc.

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...to establish standards for the description and dissemination of magnetic structures and their underlying symmetries and promote these standards within the IUCr and among other research communities that rely on magnetic structure information.

http://magcryst.org
The use of magnetic space groups and superspace groups, and magnetic irreducible representations, to constrain general magnetic-structure models. New magnetic-symmetry resources and capabilities for solving/refining magnetic structures.

Symmetry heavyweights:
Software tools for detailed symmetry analysis both RA and Shubnikov, superspace 3D+1

http://magcryst.org

ISOTROPY Software Suite
http://stokes.byu.edu/iso/

Bilbao Crystallographic Server
http://www.cryst.ehu.es

University of the Basque Country

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- A. Cornia (Italy)
- D. B. Litvin (USA)
- J.M. Perez-Mato (Spain)
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- A. Pirogov (Russia)
- V. Pomjakushin (Switzerland)
- J. Rodriguez-Carvajal (France)
- T. Sato (Japan)
- W. Sikora (Poland)
- A.S. Wills (UK)

Consultants
- M.I. Aroyo (Spain)
- J. Brown (France)
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IUCr Congress Satellite Workshop (Hamilton, Canada) on the Role of Magnetic Symmetry in the Description & Determination of Magnetic Structures  
http://magcryst.org
http://magcryst.org/meetings/cmsworkshop2014/program/

Downloadable Lecture Presentations on:  
The use of magnetic space groups and superspace groups, and magnetic irreducible representations, to constrain general magnetic-structure models. New magnetic-symmetry resources and capabilities for solving/refining magnetic structures.
Representation* Analysis (RA). Propagation vector $k$ formalism. Magnetic mode $S_0$ is specified in **zeroth block of the cell** == parent cell without centering translations

Magnetic moment below a phase transition

$$S(t_n) = Re \left( C S_0 e^{2\pi i t_n k} \right) \sim \cos(2\pi t_n k + \varphi)$$

0th cell with many atoms in general

*irreducible representation irrep: each group element $g$ --> *matrix* $\tau(g)$ that specifies the spin transformation under element $g$
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\[
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\]

amplitude or mixing coefficients

magnetic mode

magnetic mode $S_0$ for chosen irrep* specifies magnetic configuration of all spins in zeroth cell

\[
S_0 = \begin{pmatrix}
S_{x1} \\
S_{y1} \\
S_{z1} \\
S_{x2} \\
S_{y2} \\
S_{z2} \\
\vdots \\
\vdots \\
\vdots \\
S_{xN} \\
S_{yN} \\
S_{zN}
\end{pmatrix}
\]

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Magnetic moment
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Zeroth cell with many atoms in general

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S_{yN} \\
S_{zN}
\end{pmatrix}
\]

E.g., atom1 \( S_{01} = e_y \)
atom2 \( S_{02} = e_x \)

\[
S_1(t_n) = C e_y \cos(\pi(t_{nx} + t_{ny}))
\]

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S_2(t_n) = C e_x \cos(\pi(t_{nx} + t_{ny}))
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Magnetic moment below a phase transition

0th cell with many atoms in general

E.g., atom1 $S_{01} = e_y$

atom2 $S_{02} = e_x$

$S_0 = \begin{pmatrix} 0 \\ 1 \\ 0 \\ 0 \end{pmatrix}$

$S_1(t_n) = Ce_y \cos(\pi(t_{nx} + t_{ny}))$

$S_2(t_n) = Ce_x \cos(\pi(t_{nx} + t_{ny}))$

$S(t_n) = Re \left( CS_0 e^{2\pi i t_n k} \right) \sim \cos(2\pi t_n k + \varphi)$

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**Representation Analysis (RA).** Propagation vector $k$ formalism. Magnetic mode $S_0$ is specified in **zeroth block of the cell** = parent cell without centering translations.

Magnetic moment below a phase transition

**0th cell with many atoms in general**

$$\mathbf{S}(t_n) = \text{Re} \left( C S_0 e^{2\pi i t_n k} \right) \sim \cos(2\pi t_n k + \varphi)$$

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$S_0$ and $C = |C| e^{i\varphi}$ are complex quantities

$$S_0 = \begin{pmatrix} s_{x1} \\ s_{y1} \\ s_{z1} \\ s_{x2} \\ s_{y2} \\ s_{z2} \\ \vdots \\ s_{xN} \\ s_{yN} \\ s_{zN} \end{pmatrix}$$

$$S_{1}(t_n) = C e_y \cos(\pi(t_{nx} + t_{ny}))$$

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E.g., atom1 $S_{01} = e_y$ atom2 $S_{02} = e_x$ $S_0 = \begin{pmatrix} 0 \\ 1 \\ 0 \\ 1 \\ 0 \end{pmatrix}$
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S_{zN}
\end{pmatrix}
$$

$$
S_1(t_n) = C e_y \cos(\pi(t_n x + t_n y))
$$

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E.g., atom 1 $S_{01} = e_y$

atom 2 $S_{02} = e_x$

$S_0 = \begin{pmatrix}
0 \\
1 \\
0 \\
0
\end{pmatrix}$

$|S_{x1}| e^{i\phi_{x1}} e_x$

$|S_{y1}| e^{i\phi_{y1}} e_y$

$|S_{zN}| e^{i\phi_{zN}} e_z$

...
Example of complex magnetic structure

Antiferromagnetic (cycloidal spiral) three sub-lattice ordering in \( \text{Tb}_{14}\text{Au}_{51} \)

P6/m

k-vector=[1/3, 1/3, 0]

Zeroth cell:
only 5 magnetic modes, i.e.
5 mixing coefficients \( C \) to find from experiment.

Zeroth cell contains 14 spins of \( \text{Tb}^{3+} \)
Conventional magnetic unit cell contains
126 spins of \( \text{Tb}^{3+} \)!!
what if several magnetic modes $S_0$ are possible in RA?
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Two commensurate cases when $k$="rational fraction"

1. multi-dimentional (nD) irreducible representation generates $nD$ magnetic modes $S_0^1, S_0^2, S_0^3 ... S_0^{nD}$

$$S(0) \sim \sum_{l=1}^{nD} C_l S_0^l$$
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Two commensurate cases when $k$=“rational fraction”

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$$S(0) \sim \sum_{l=1}^{nD} C_l S_0^l$$

any relations between mixing coefficients $C_l$ ?
TmMnO$_3$

Two magnetic modes $E_1$ and $E_2$ along $x$.

Mn-position

(1) $0,0,\frac{1}{2}$
(2) $\frac{1}{2},\frac{1}{2},0$
(3) $0,\frac{1}{2},\frac{1}{2}$
(4) $\frac{1}{2},0,0$

$S_0^1 = E_1 = +1$

+1  +1  -1  -1

$S_0^2 = E_2 = +1$

-1  -1  +1  +1

$Pnma$ $k=[1/2,0,0]$, $k20$, $X$

irreps: two 2D $\tau_1$, $\tau_2$

Mn m$\Gamma$: $3\tau_1 \oplus 3\tau_2$
Two magnetic modes $E_1$ and $E_2$ along x.

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(1) $0,0,\frac{1}{2}$
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$S_0^1 = E_1 = +1$
$S_0^2 = E_2 = +1$

Any linear combination, in general

$C_1E_1+C_2E_2 = C_1+C_2$
$C_1-C_2$
$-C_1-C_2$
$C_1+C_2$

$(E_1+E_2)/2 = +1$
$(E_1-E_2)/2 = 0$

$Pnma$ $k=[1/2,0,0]$, $k20$, $X$

irreps: two 2D $\tau_1$, $\tau_2$

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Two magnetic modes $E_1$ and $E_2$ along $x$.

Mn-position

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Any linear combination, in general

$C_1E_1 + C_2E_2 = C_1 + C_2$

$(E_1 + E_2)/2 = +1$

$(E_1 - E_2)/2 = 0$
what if several magnetic modes $S_0^1, S_0^2, S_0^3...$ are possible in RA?

Two commensurate cases when $k$="rational fraction"

1. multi-dimentional (nD)
irreducible representation generates
$nD$ magnetic modes $S_0^1, S_0^2, S_0^3...$
$S_0^{nD}$

$S(0) \sim \sum_{l=1}^{nD} C_l S_0^l$

any relations between mixing coefficients $C_l$ ?

2. multi-Arm/multi-$k$ structure
(non-equivalent $k_1, k_2, ... k_m$).
$nA$ magnetic modes $S_0^1, S_0^2,$
$S_0^3... S_0^{nA}$

RA: widespread unfavorable paradigm that one-$k$ is enough...

Example of mutiarm, full star \{k_1,k_2\}: J. Phys.: Condens. Matter 26 496002
what if several magnetic modes $S_{0}^{1}, S_{0}^{2}, S_{0}^{3}...$ are possible in RA?

Two commensurate cases when k="rational fraction"

1. multi-dimentional (nD) irreducible representation generates $nD$ magnetic modes $S_{0}^{l}, S_{0}^{2}, S_{0}^{3}...$

$$S(0) \sim \sum_{l=1}^{nD} C_{l} S_{0}^{l}$$

2. multi-Arm/multi-k structure (non-equivalent $k_{1}, k_{2}, ... k_{m}$).

$nA$ magnetic modes $S_{0}^{1}, S_{0}^{2}, S_{0}^{3}... S_{0}^{nA}$

$$S(t_{n}) \sim \sum_{m=1}^{nA} C'_{m} S_{0}^{m} \cos(2\pi k_{m} t_{n} + \varphi_{m})$$

RA: widespread unfavorable paradigm that one-k is enough...

any relations between mixing coefficients $C_{l}$ or $C'_{m}$?
what if several magnetic modes $S_0^1$, $S_0^2$, $S_0^3$... are possible in RA?

Two commensurate cases when $k$="rational fraction"

1. multi-dimensional (nD)
irreducible representation generates
$nD$ magnetic modes $S_0^1$, $S_0^2$, $S_0^3$...
$S_0^{nD}$

$$S(0) \sim \sum_{l=1}^{nD} C_l S_0^l$$

2. multi-Arm/multi-$k$ structure
(non-equivalent $k_1$, $k_2$, ... $k_m$).

$nA$ magnetic modes $S_0^1$, $S_0^2$, $S_0^3$... $S_0^{nA}$

$$S(t_n) \sim \sum_{m=1}^{nA} C'_m S_0^m \cos(2\pi k_m t_n + \varphi_m)$$

RA: widespread unfavorable paradigm that one-$k$ is enough...

any relations between mixing coefficients

$C_l$ or $C'_m$?

for incommensurate structures:

any constraints on mixing coefficients?

1. between atoms unrelated by $1$? 2. phases along $x,y,z$?

$$S_0 = |C_x| e^{i\phi_x} e_x + |C_y| e_y, \phi_x = 0, \frac{\pi}{2}, ...?$$

amplitude modulation for $\phi_x = 0$,
cycloid or helix for e.g. $\phi_x = \frac{\pi}{2}, ...$?

$$\{k, -k\}$$
what if several magnetic modes $S_{0}^{1}$, $S_{0}^{2}$, $S_{0}^{3}$... are possible in RA?

Two commensurate cases when $k$="rational fraction"

1. multi-dimentional (nD)
irreducible representation generates $nD$ magnetic modes $S_{0}^{1}$, $S_{0}^{2}$, $S_{0}^{3}$...

$$S(0) \sim \sum_{l=1}^{nD} C_{l} S_{0}^{l}$$

2. multi-Arm/multi-k structure
(non-equivalent $k_{1}$, $k_{2}$, ... $k_{m}$).

$nA$ magnetic modes $S_{0}^{1}$, $S_{0}^{2}$, $S_{0}^{3}$... $S_{0}^{nA}$

$$S(t_{n}) \sim \sum_{m=1}^{nA} C_{m}' S_{0}^{m} \cos(2\pi k_{m} t_{n} + \varphi_{m})$$

RA: widespread unfavorable paradigm that one-\(k\) is enough...

any relations between mixing coefficients

\(C_{l}\) or \(C'_{m}\)?

for incommensurate structures:

No from RA alone...

Yes from magnetic symmetry!

any constraints on mixing coefficients?

1. between atoms unrelated by 1? 2. phases along x,y,z?

$$S_{0} = |C_{x}|e^{i\phi_{x}} e_{x} + |C'_{y}|e_{y}, \phi_{x} = 0, \frac{\pi}{2}, ...?$$

amplitude modulation for \(\phi_{x} = 0\),
cyclod or helix for e.g. \(\phi_{x} = \frac{\pi}{2}, ...?\)  \(\{k, -k\}\)
Antiferromagnetic order in orthorhombic multiferroic TmMnO$_3$

1. one-arm two dimensional irrep $\mathbf{k}=[1/2,0,0]$. Ferro-electric phase polar magnetic group $P_{bmn}2_1$

2. Constraints on basis functions vs. superspace for the incommensurate two arm $\mathbf{k}=[1/2\pm\delta,0,0]$. $\{\mathbf{k}\}=${-k,+k}. Para-electric phase (3D+1) superspace magnetic group $Pmcn1'(00g)000s [Pnma, bca]$

Symmetry analysis using both RA and magnetic subgroups

$Pnma \ k=[1/2,0,0]$, irrep: $2D \ mX1(\tau_1)$

RA with arbitrary mixing coefficients gives different spin sizes for the same type of spins. Symmetry?
Symmetry analysis using both RA and magnetic subgroups

\[ Pnma \ k = [1/2, 0, 0], \text{ irrep: } 2D \ mX1(\tau_1) \]

RA with arbitrary mixing coefficients gives different spin sizes for the same type of spins. Symmetry?
Symmetry analysis using both RA and magnetic subgroups

Symmetry: Pnma, \( k = [1/2, 0, 0] \), irrep: 2D mX1(\( \tau_1 \))

Order parameter direction

Magnetic Shubnikov Space group

RA with arbitrary mixing coefficients gives different spin sizes for the same type of spins. Symmetry?

| P1 (a,0) 11.55 | P_a2\( \overline{1}/m \), basis\( = \{(2,0,0),(0,1,0),(0,0,1)\} \), origin\( =(1/2,0,0) \), s=2, i=4, k-active\(=(1/2,0,0) \)
| P3 (a,a) 31.129 | P_bmn2\( \_1 \), basis\( = \{(0,1,0),(2,0,0),(0,0,-1)\} \), origin\( =(3/4,1/4,0) \), s=2, i=4, k-active\(=(1/2,0,0) \)
| C1 (a,b) 6.21 | P_am, basis\( = \{(2,0,0),(0,1,0),(0,0,1)\} \), origin\( =(0,1/4,0) \), s=2, i=8, k-active\(=(1/2,0,0) \)

http://stokes.byu.edu/iso/

ISODISTORT
Version 6.1.8, November 2014
Harold T. Stokes, Branton J. Campbell, and Dorian M. Hatch,
Symmetry analysis using both RA and magnetic subgroups

$Pnma \ k=[1/2,0,0], \ irrep: \ 2D \ mX1(\tau_1)$

Order parameter direction

Magnetic Shubnikov

Space group

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http://www.cryst.ehu.es

P1 (a,0) 11.55 $P_{a2_1/m}$,  
basis=\{(2,0,0),(0,1,0),(0,0,1)\}, origin=(1/2,0,0), s=2, i=4, k-active= (1/2,0,0)
P3 (a,a) 31.129 $P_{bmn2_1}$,  
basis=\{(0,1,0),(2,0,0),(0,0,-1)\}, origin=(3/4,1/4,0), s=2, i=4, k-active= (1/2,0,0)
C1 (a,b) 6.21 $P_{am}$,  
basis=\{(2,0,0),(0,1,0),(0,0,1)\}, origin=(0,1/4,0), s=2, i=8, k-active= (1/2,0,0)

RA with arbitrary mixing coefficients gives different spin sizes for the same type of spins. Symmetry?
Case 1: magnetic mode $E_1 \rightarrow$ most symmetric maximal subgroup of $Pnma1'$

Order parameter direction → Magnetic Shubnikov Space group

$P1 (a,0)$ 11.55 \(P_{a2/m}\)

$P3 (a,a)$ 31.129 \(P_{bm21}\)

$C1 (a,b)$ 6.21 \(P_{am}\)

Solution!

TmMnO$_3$

$irrep$: 2D mX1(τ$_1$)

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ISODISTORT
Version 6.1.8, November 2014
Harold T. Stokes. Branton J. Campbell, and Dorian M. Hatch,

Harold T. Stokes, Dorian M. Hatch, and Branton J. Campbell, Department of Physics and Astronomy, Brigham Young University, Provo, Utah 84605, USA.
**Case 1:** magnetic mode $E_1 \rightarrow \text{most symmetric maximal subgroup of } P_{nma1'}$

**Order parameter direction**

P1 (a,0)  $\begin{array}{c} \text{11.55} \\ P_{a2}/m \end{array}$,  

P3 (a,a)  $\begin{array}{c} \text{31.129} \\ P_{bmn21}, \end{array}$  

C1 (a,b)  $\begin{array}{c} \text{6.21} \\ P_{am}, \end{array}$

**Magnetic Shubnikov Space group**

- P1 (a,0)  $\begin{array}{c} \text{11.55} \\ P_{a2}/m, \end{array}$  
  - basis=$\{(2,0,0),(0,1,0),(0,0,1)\}$, origin=$(1/2,0,0)$, $s=2$, $i=4$, k-active= $(1/2,0,0)$

- P3 (a,a)  $\begin{array}{c} \text{31.129} \\ P_{bmn21}, \end{array}$  
  - basis=$\{(0,1,0),(2,0,0),(0,0,-1)\}$, origin=$(3/4,1/4,0)$, $s=2$, $i=4$, k-active= $(1/2,0,0)$

- C1 (a,b)  $\begin{array}{c} \text{6.21} \\ P_{am}, \end{array}$
  - basis=$\{(2,0,0),(0,1,0),(0,0,1)\}$, origin=$(0,1/4,0)$, $s=2$, $i=8$, k-active= $(1/2,0,0)$

**Orthorhombic $P_{mn21}$**

- (1) $1_1$ 0,0,0
- (2) $2(0,0,\frac{1}{2})$  $\frac{1}{4},0,z$
- (3) $2(0,\frac{1}{2},0)$  0,$y,0$
- (4) $2(\frac{1}{2},0,0)$  $x,\frac{1}{2},\frac{1}{2}$
- (5) $1_1$ 0,0,0
- (6) $a,x,y,\frac{1}{4}$
- (7) $m,x,\frac{1}{4},z$
- (8) $n(0,\frac{1}{2},\frac{1}{2})$  $\frac{1}{4},y,z$

**Irrep:** $2D$  $mX_1(\tau_1)$

**Electric polarisation along c allowed**

**TmMnO$_3$**

**Solution!**

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http://www.cryst.ehu.es

Pomjakushin, UFOX 7-8 July 2016 University of Salerno  2016
**Case 2: General solution in RA -> low symmetry non-maximal subgroup**

Order parameter direction → Magnetic Shubnikov Space group

- **P1 (a,0) 11.55**  \( P_{a2\_1/m}, \) basis\(=\{(2,0,0),(0,1,0),(0,0,1)\} \), origin\(=(1/2,0,0)\), s=2, i=4, k-active\(= (1/2,0,0)\)
- **P3 (a,a) 31.129**  \( P_{bmn2\_1}, \) basis\(=\{(0,1,0),(2,0,0),(0,0,-1)\} \), origin\(=(3/4,1/4,0)\), s=2, i=4, k-active\(= (1/2,0,0)\)
- **C1 (a,b) 6.21**  \( P_{am}, \) basis\(=\{(2,0,0),(0,1,0),(0,0,1)\} \), origin\(=(0,1/4,0)\), s=2, i=8, k-active\(= (1/2,0,0)\)

Conventional general solution in RA: lowest symmetry for the given irrep

**monoclinic Pm**

\[
\begin{align*}
(1) & & 1 & & 0,0,0 \\
(2) & & 2(0,0,\frac{1}{2}) & & \frac{1}{4},0,z \\
(3) & & 2(0,\frac{1}{4},0) & & 0,y,0 \\
(4) & & 2(\frac{1}{2},0,0) & & x,\frac{1}{4},\frac{1}{4} \\
(5) & & 0,0,0 & & 0,0,0 \\
(6) & & a,x,y,\frac{1}{4} & & \\
(7) & & m,x,\frac{1}{4},z & & \\
(8) & & n(0,\frac{1}{2},\frac{1}{2}) & & \frac{1}{4},v,z \\
\end{align*}
\]

Irrep: \( \mathbf{2D} mX1(\tau_1) \)

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**TmMnO₃**

Order parameter direction → Magnetic Shubnikov Space group

- **P1 (a,0) 11.55**  \( P_{a2\_1/m}, \) basis\(=\{(2,0,0),(0,1,0),(0,0,1)\} \), origin\(=(1/2,0,0)\), s=2, i=4, k-active\(= (1/2,0,0)\)
- **P3 (a,a) 31.129**  \( P_{bmn2\_1}, \) basis\(=\{(0,1,0),(2,0,0),(0,0,-1)\} \), origin\(=(3/4,1/4,0)\), s=2, i=4, k-active\(= (1/2,0,0)\)
- **C1 (a,b) 6.21**  \( P_{am}, \) basis\(=\{(2,0,0),(0,1,0),(0,0,1)\} \), origin\(=(0,1/4,0)\), s=2, i=8, k-active\(= (1/2,0,0)\)

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(5) & & 0,0,0 & & 0,0,0 \\
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(8) & & n(0,\frac{1}{2},\frac{1}{2}) & & \frac{1}{4},v,z \\
\end{align*}
\]

Irrep: \( \mathbf{2D} mX1(\tau_1) \)

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**TmMnO₃**

Order parameter direction → Magnetic Shubnikov Space group

- **P1 (a,0) 11.55**  \( P_{a2\_1/m}, \) basis\(=\{(2,0,0),(0,1,0),(0,0,1)\} \), origin\(=(1/2,0,0)\), s=2, i=4, k-active\(= (1/2,0,0)\)
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Conventional general solution in RA: lowest symmetry for the given irrep

**monoclinic Pm**

\[
\begin{align*}
(1) & & 1 & & 0,0,0 \\
(2) & & 2(0,0,\frac{1}{2}) & & \frac{1}{4},0,z \\
(3) & & 2(0,\frac{1}{4},0) & & 0,y,0 \\
(4) & & 2(\frac{1}{2},0,0) & & x,\frac{1}{4},\frac{1}{4} \\
(5) & & 0,0,0 & & 0,0,0 \\
(6) & & a,x,y,\frac{1}{4} & & \\
(7) & & m,x,\frac{1}{4},z & & \\
(8) & & n(0,\frac{1}{2},\frac{1}{2}) & & \frac{1}{4},v,z \\
\end{align*}
\]

Irrep: \( \mathbf{2D} mX1(\tau_1) \)
Magnetic structure of Pyrochlore Tm$_2$Mn$_2$O$_7$ at $\Gamma$-point $k=0$

\[ \Gamma_{\text{mag}} = \Gamma_2^+(1D) \oplus \Gamma_3^+(2D) \oplus \Gamma_5^+(3D) \oplus 2\Gamma_4^+(3D) \]

Inorg. Chem. 2015, 54, 9092–9097

HRPT/SINQ Magnetic neutron diffraction
Magnetic structure of Pyrochlore \( \text{Tm}_2\text{Mn}_2\text{O}_7 \) at \( \Gamma \)-point \( k=0 \)

Maximal and non-maximal MG for the parent SG 227 \( (\text{Fd}-3m) \) at gamma point \( k = (0, 0, 0) \)
generated by one irrep for 16d \((1/2,1/2,1/2)\), 16c \((0,0,0)\) position

\[
\Gamma_{\text{mag}} = \Gamma_2^+ (1D) \oplus \Gamma_3^+ (2D) \oplus \Gamma_5^+ (3D) \oplus 2\Gamma_4^+ (3D)
\]

\[\text{Im}'/\text{ma} \quad 10 \]
\[\text{Imm}'/\text{a} \quad 9 \]
\[\text{I}_{4_1}/\text{a}'/\text{md} \quad 8 \]
\[\text{I}_{4_1}/\text{a}'/\text{md}' \quad 7 \]
\[\text{I}_{4_1}/\text{am}'/\text{d} \quad 6 \]
\[\text{I}_{4_1}/\text{am}'/\text{d}' \quad 5 \]
\[\text{Fd}\bar{3}m' \quad 0 \]

\[\text{Solution} \quad \text{Spin Mn, Tm} \neq 0 \]

FM

1 modes

2 modes

3 modes

\[2\Gamma_4^{+}(aa0) \quad \text{FM} \]

\[2\Gamma_5^{+}(aa0) \quad \text{nonFM} \]

\[\Gamma_5^{+}(aa0) \quad \text{nonFM} \]

accidentally or/and

2\( \Gamma_4^{+}(0-a) \) FM

FM

2\( \Gamma_4^{+}(aab), \Gamma_2^{+}(a), \Gamma_3^{+}(0a), \Gamma_5^{+}(0a-a), \)

C2'/m' FM

7 modes

FM

\[2\Gamma_5^{+}(aab), \Gamma_2^{+}(a), \Gamma_4^{+}(0-a) \quad \text{FM,} \]

\[\Gamma_5^{+}(0a-a), \]

C2'/c' FM

6 modes primary \( \Gamma_4^{+}(ab0) \)

\[2\Gamma_4^{+}(ab0), \Gamma_5^{+}(0ab) \]

\[2\Gamma_4^{+}(abc), \Gamma_2^{+}(a), \Gamma_3^{+}(ab), \Gamma_5^{+}(abc), \]

P-1 FM

12 modes

FM

\[\Gamma_5^{+}(abc), \Gamma_2^{+}(a), \Gamma_3^{+}(ab), 2\Gamma_4^{+}(abc) \quad \text{FM.} \]

P-1 FM

12 modes, 3 modes from primary

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Magnetic structure of Pyrochlore Tm$_2$Mn$_2$O$_7$ at $\Gamma$-point $k=0$

Maximal and non-maximal MG for the parent SG 227 ($Fd$-3$m$) at gamma point $k = (0, 0, 0)$ generated by one irrep for 16d ($1/2,1/2,1/2$), 16c (0,0,0) position

$$\Gamma_{mag} = \Gamma_2^+(1D) \oplus \Gamma_3^+(2D) \oplus \Gamma_5^+(3D) \oplus 2\Gamma_4^+(3D)$$

**Solution**

Spin Mn, Tm ≠ 0

**Representation analysis without symmetry considerations for $\Gamma4+$**

Maximal and non-maximal MG for the parent SG 227 ($Fd$-3$m$) at gamma point $k=0$ generated by one irrep for 16d ($1/2,1/2,1/2$), 16c (0,0,0) position
Relation of magnetic Shubnikov/superspace symmetry and representation analysis RA

Paramagnetic crystallographic space group (PSG)  
Propagation vector of magnetic structure $k$

choose one irreducible representation (irrep) of PSG

magnetic symmetry representation
Relation of magnetic Shubnikov/superspace symmetry and representation analysis RA

Paramagnetic crystallographic space group (PSG)

Propagation vector of magnetic structure $\mathbf{k}$

choose one irreducible representation (irrep) of PSG

magnetic symmetry

Construction of basis functions (normal modes)

**Constraints** on the mixing coefficients of basis function for >1D irrep and/or multi-arm star of $\mathbf{k}$, $\{-\mathbf{k}, \mathbf{k}\}$ star for incommensurate.

Magnetic structure made from linear combination of normal modes.
Relation of magnetic Shubnikov/superspace symmetry and representation analysis RA

Paramagnetic crystallographic space group \((PSG)\)

Propagation vector of magnetic structure \(\mathbf{k}\)

choose one irreducible representation \((irrep)\) of \(PSG\)

magnetic symmetry

Construction of basis functions (normal modes)

Constraints 

the mixing coefficients of basis function for >1D \(irrep\) and/or multi-arm star of \(\mathbf{k}\), \([-\mathbf{k},\mathbf{k}]\) star for incommensurate.

Magnetic structure made from linear combination of normal modes.
Relation of magnetic Shubnikov/superspace symmetry and representation analysis RA

Paramagnetic crystallographic space group (PSG)  
Propagation vector of magnetic structure \( k \)

Choose one irreducible representation (irrep) of PSG

Is irrep real and 1D, and single arm \{k\}-star?

Yes

Shubnikov from PSG Symop \( g \) that have \( \text{irrep}(g) = -1 \) are primed in Sh-group

Magnetic structure made from admissible spin directions in Sh-group

Constraints on the mixing coefficients of basis function for >1D irrep and/or multi-arm star of \( k \), \{-k,k\} star for incommensurate.

Magnetic structure made from linear combination of normal modes.
Relation of magnetic Shubnikov/superspace symmetry and representation analysis RA

Paramagnetic crystallographic space group (PSG)

Propagation vector of magnetic structure $k$

choose one irreducible representation (irrep) of PSG

magnetic symmetry

is irrep real and 1D, and single arm $\{k\}$-star? Yes

No

is $k$ commensurate?

Yes

No

Shubnikov from PSG $Symop g$ that have irrep($g$)=-1 are primed in Sh-group

Construction of basis functions (normal modes)

Constraints? the mixing coefficients of basis function for >1D irrep and/or multi-arm star of $k$, $\{-k,k\}$ star for incommensurate.

Magnetic structure made from admissible spin directions in Sh-group

Magnetic structure made from linear combination of normal modes.

choice of direction of order parameter for irrep

combining nD irrep based on full star $\{k\}$ & c.c into real 2nD

equivalent
Relation of magnetic Shubnikov/superspace symmetry and representation analysis RA

Paramagnetic crystallographic space group ($PSG$)

Propagation vector of magnetic structure $k$

choose one irreducible representation ($irrep$) of $PSG$

magnetic symmetry

is $irrep$ real and 1D, and single arm $\{k\}$-star? No is $k$ commensurate?  

Yes

Shubnikov from $PSG$ $Symop g$ that have $irrep(g)=-1$ are primed in Sh-group

No

Construction of basis functions (normal modes)

Constraints?

the mixing coefficients of basis function for $>1D$ $irrep$ and/or multi-arm star of $k$, $\{-k,k\}$ star for incommensurate.

Magnetic structure made from linear combination of normal modes.

Shubnikov from isotropy subgroup of $PSG$.

choice of direction of order parameter for $irrep$

equivalent

Magnetic structure made from admissible spin directions in Sh-group
**Relation of magnetic Shubnikov/superspace symmetry and representation analysis RA**

- **Paramagnetic crystallographic space group (PSG)**
- **Propagation vector of magnetic structure \( \mathbf{k} \)**

choose one irreducible representation \((\text{irrep})\) of PSG

**magnetic symmetry**

**is irrep real and 1D, and single arm \( \{ \mathbf{k} \} \)-star?**
- **Yes**
  - Shubnikov from PSG
  - Symop \( g \) that have \( \text{irrep}(g) = -1 \) are primed in Sh-group

**3D+1 magnetic superspace group for \( \{ \mathbf{k}, -\mathbf{k} \} \) star in general**

**is \( \mathbf{k} \) commensurate?**
- **No**
  - Shubnikov from isotropy subgroup of PSG.
- **Yes**
  - choice of direction of order parameter for irrep

**Combining nD irrep based on full star \( \{ \mathbf{k} \} \) & c.c into real 2nD**

**Construction of basis functions (normal modes)**

**Constraints**
- The mixing coefficients of basis function for \( >1 \)D irrep and/or multi-arm star of \( \mathbf{k}, \{-\mathbf{k}, \mathbf{k}\} \) star for incommensurate.

**Magnetic structure made from linear combination of normal modes.**

**Magnetic structure made from admissible spin directions in Sh-group**
**or in 3D+1 magnetic group**
Relation of magnetic Shubnikov/superspace symmetry and representation analysis RA

Paramagnetic crystallographic space group (PSG)

Propagation vector of magnetic structure \( k \)

The disadvantage of using only RA:

In general: there are no rules to make constraints, (except ones based on physical grounds)

but the constraints appear in a natural way from magnetic group symmetry arguments

Magnetic structure made from admissible spin directions in Sh-group

or in 3D+1 magnetic group
Thank you!
Relation of magnetic Shubnikov/superspace symmetry and representation analysis RA

Paramagnetic crystallographic space group (PSG) → Propagation vector of magnetic structure \( \mathbf{k} \)

1. Choose one irreducible representation (irrep) of PSG

2. Is irrep real and 1D, and single arm \( \{\mathbf{k}\} \)-star?
   - Yes → Shubnikov from PSG
   - No → is \( \mathbf{k} \) commensurate?
     - No → 3D+1 magnetic superspace group for \( \{\mathbf{k}, -\mathbf{k}\} \) star in general
     - Yes → combining nD irrep based on full star \( \{\mathbf{k}\} \) & c.c into real 2nD

3. Shubnikov from isotropy subgroup of PSG.

4. Magnetic structure made from admissible spin directions in Sh-group

5. Choice of direction of order parameter for irrep

6. Constraints on the mixing coefficients of basis function for >1D irrep and/or multi-arm star of \( \mathbf{k} \) \( \{\mathbf{-k}, \mathbf{k}\} \) star for incommensurate.

7. Magnetic structure made from linear combination of normal modes.
Example of HRPT & DMC complementarity

**Magnetic structure TmMnO$_3$**

(a) IC Structure for $T_C < T < T_N$

Para-electric phase
(3D+1) superspace magnetic group $Pmcn 1'(00g)000s$

(b) C Structure ($E_1$) for $T << T_C$

Ferro-electric phase
polar magnetic group $P_{bmn2_1}$

Solved from DMC

$E = 0$

$E$ (polarization)
Example of accuracy on metric: orthorhombic multiferroic TmMnO$_3$

accuracy \( \sim 0.0001\text{Å} = 10 \text{ fm} \)
Example of accuracy on metric:

Orthorhombic multiferroic TmMnO$_3$

Material that have coupled electric, magnetic and structural order

Accuracy $\sim 0.0001\,\text{Å} = 10\,\text{fm}$

Lattice constants

Spin-lattice coupling

Refined from HRPT