



Incommensurate magnetic structures and magnetic superspace groups (MSSGs)

Branton J. Campbell

Department of Physics & Astronomy

Brigham Young University

2024 International Workshop on Magnetic Crystallography (IWMC2024)

Organized by the Chinese Spallation Neutron Source

Dongguan International Exhibition Hotel, Dongguan City, Guangdong, China

24-30 November 2024



Acknowledgements

Harold T. Stokes (*Brigham Young University, USA*)

Sander van Smaalen (*U. Bayreuth, Germany*)

Manuel Perez-Mato (*U. Basque Country, Bilbao, Spain*)

Václav Petříček (*Institute of Physics CAS, Prague, Czech Republic*)

Andrey Gubkin (*Institute of Metal Physics, Yekaterinburg, Russia*)

S. van Smaalen, *Incommensurate Crystallography* (2007), Oxford University Press.

MAGNDATA (<http://webbdcristal.ehu.es/magndata/>) contains many (3+1)D incommensurate magnetic structures.

ISO-MAG (<http://stokes.byu.edu/iso/magneticspacegroups.php>)

ISO-(3+d)D (<http://stokes.byu.edu/iso/ssg.php>)

[Magnetic superspace groups and symmetry constraints in incommensurate magnetic phases](#)

Perez-Mato *et al.*, *Journal of Physics: Condensed Matter* **24** 163201 (2012).



Modulated magnetic structures

Incommensurate magnetic structures are relevant to a wide range of materials phenomena, including skyrmions, topological magnetic textures and excitations, multi-ferroics, and more.

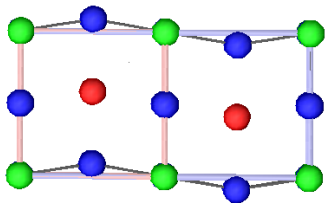
There are already 160 incommensurate magnetic structures in the MAGNDATA database of the Bilbao Crystallographic Server (BCS). Each one possesses symmetries comprising a magnetic superspace-group (MSSG).



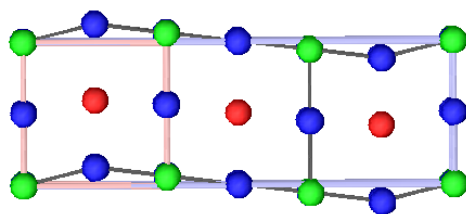
Modulation wavevector

A symmetry-breaking distortion can be decomposed into commensurate and/or incommensurate modulation waves, each with a characteristic wavevector: $A(\mathbf{r}) = \sum_n A_n e^{i(\mathbf{k}_n \cdot \mathbf{r} + \phi_n)}$; position \mathbf{r} and wavevector \mathbf{k} should be interpreted here in unitless lattice coordinates of the parent cell (pink square).

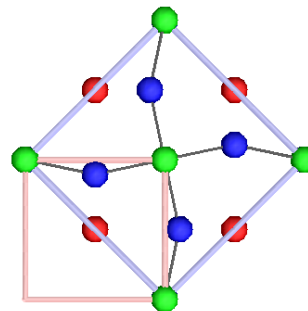
$$\mathbf{k} = (\frac{1}{2}, 0, 0)$$



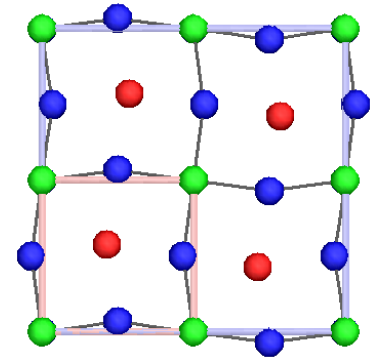
$$\mathbf{k} = (\frac{1}{3}, 0, 0)$$



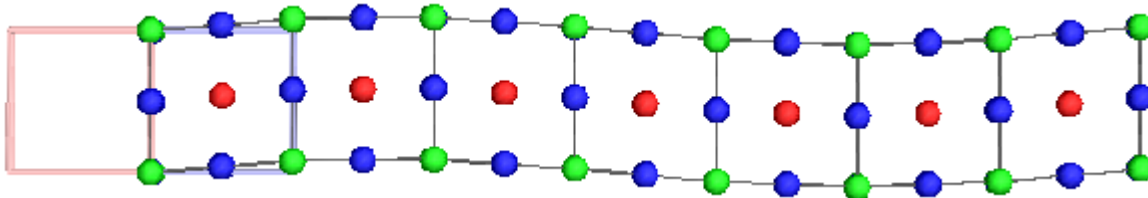
$$\mathbf{k} = (\frac{1}{2}, \frac{1}{2}, 0)$$



$$\mathbf{k} = \{(\frac{1}{2}, 0, 0), (0, \frac{1}{2}, 0)\}$$



$$\text{incommensurate } \mathbf{k} = (0.1521, 0, 0)$$

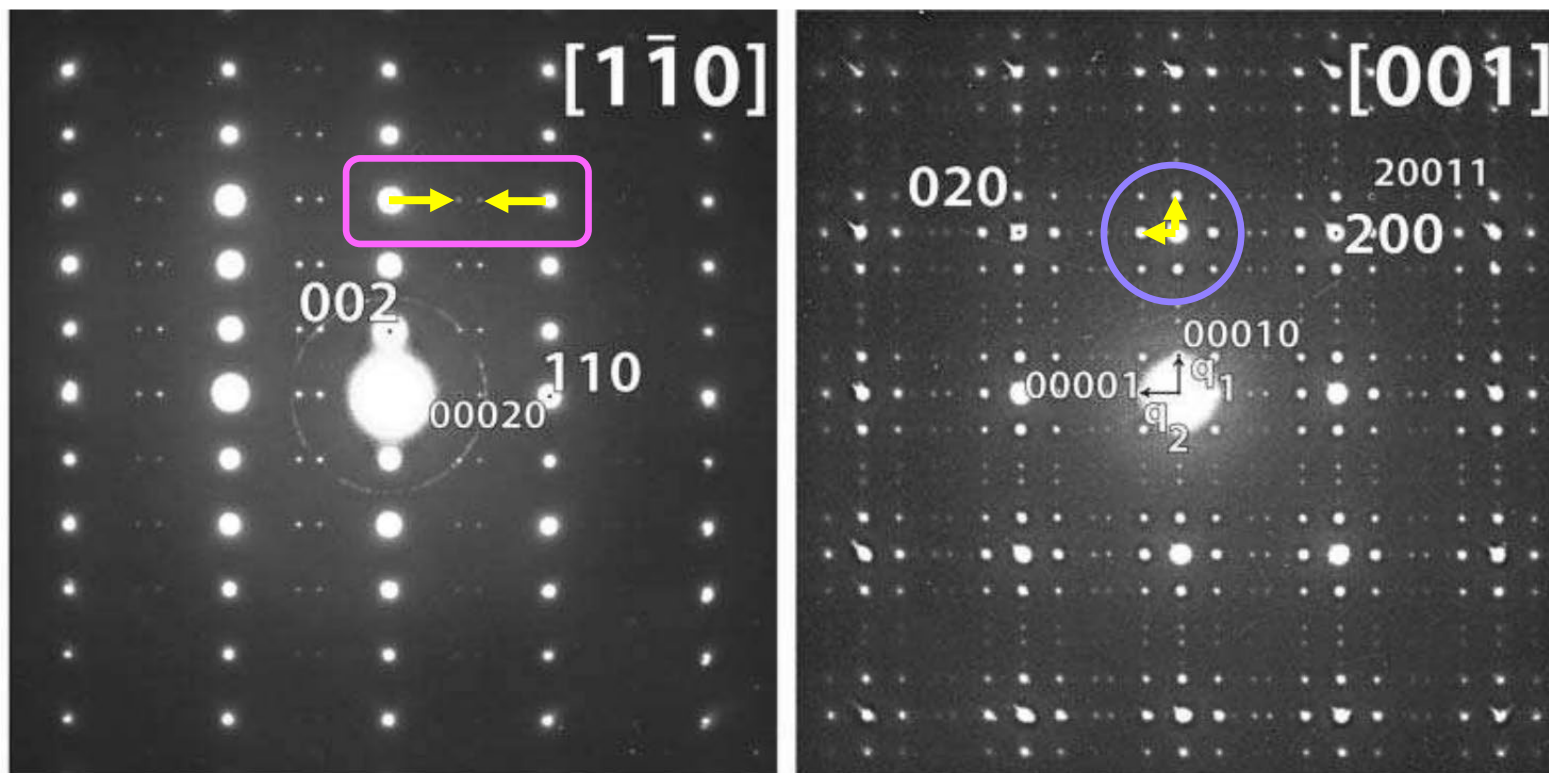


Incommensurate means the wavevector \mathbf{k} has irrational components.



Incommensurate satellites

The (3 + 2)D structure of oxygen deficient $\text{LaSrCuO}_{3.52}$
 Haderman *et al.*, J. Mater. Chem., 2007, 17, 2344–2350



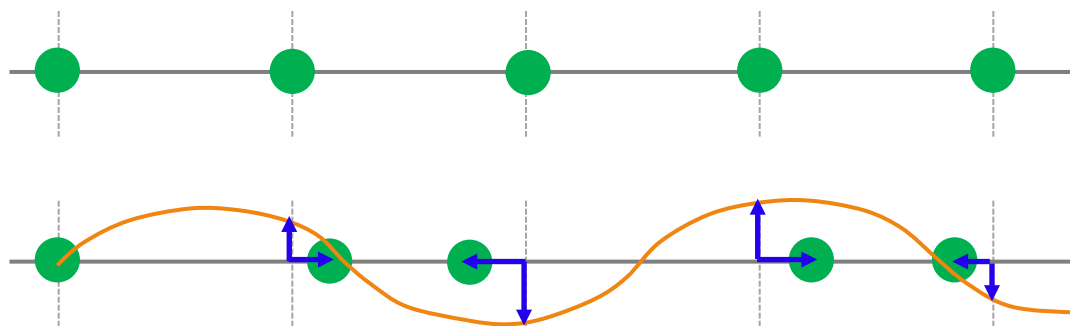
$$\mathbf{k} = (h, k, l, m, n) \cdot (\mathbf{a}^*, \mathbf{b}^*, \mathbf{c}^*, \mathbf{q}_1, \mathbf{q}_2)$$

$$\mathbf{q}_1 = \alpha \mathbf{a}^* + \alpha \mathbf{b}^* \quad \mathbf{q}_2 = \alpha \mathbf{a}^* - \alpha \mathbf{b}^* \quad \alpha = 0.22$$



Goodbye lattice translations?

Incommensurate modulations destroy translational periodicity in 3D space.



The curve drawn shows longitudinal displacement vs position.



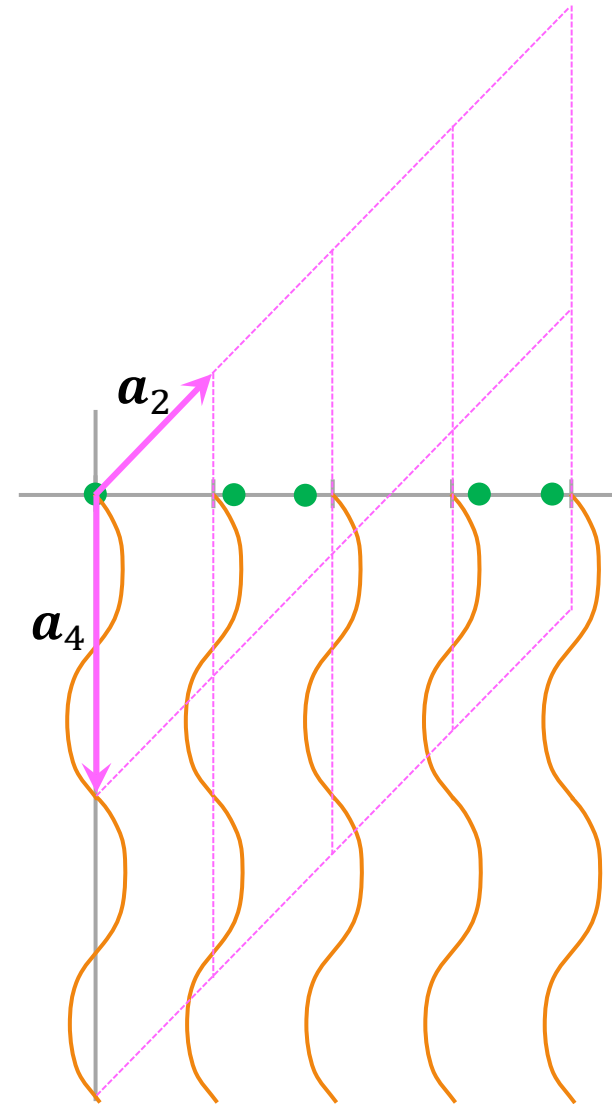
Superspace (1D example)

When we translate right by one lattice vector, the next atom is in the wrong place -- not a symmetry operation.

Treat each atom in the 1D unit cell as a wave along a new “phase” dimension (\mathbf{a}_4 , vertical axis) slide. When translating to a new unit cell, slide the wave phase until the atom is in the right place. The translation along the sloped line (\mathbf{a}_2) is a new (1+1)D symmetry operation.

Translational periodicity (see the repeating unit) is recovered by adding the phase-shift dimension.

In general, we get one extra phase dimension for each of the d independent modulation waves. We call the result $(3 + d)$ -dimensional superspace.



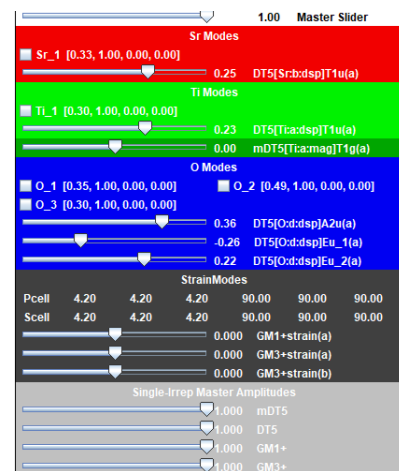
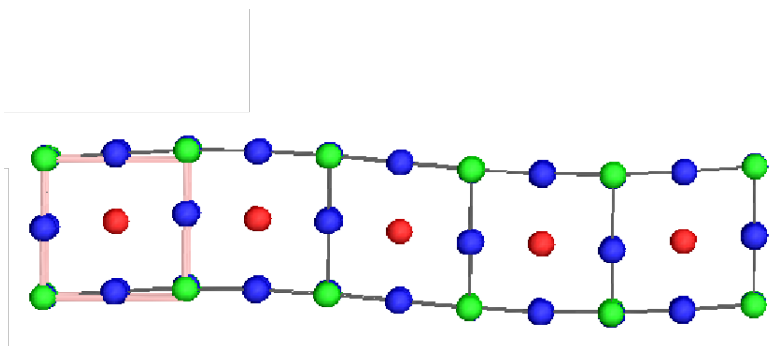


Superspace description

Each atom in the unit cell has local properties such as position, magnetic moment, occupancy, and ADPs. In a modulated structure, each atom also possess extended incommensurate waves, each having a \mathbf{k} vector, amplitude, and phase.

For a modulation with wavevector \mathbf{k} , we let A_n and ϕ_n be the amplitude and phase of the modulation of the n^{th} atom (at location \mathbf{r}_n) in the first unit cell (at the origin) and let \mathbf{t} be a lattice translation. The modulation function at position $\mathbf{r}_n + \mathbf{t}$ is $A_n e^{i(\mathbf{k} \cdot [\mathbf{r}_n + \mathbf{t}] + \phi_n)} = A_n e^{i(\mathbf{k} \cdot \mathbf{r}_n + \phi_n)} e^{i\mathbf{k} \cdot \mathbf{t}}$.

A wave belonging to one atom also belongs to all translationally equivalent atoms in the crystal! We only define waves for atoms in the first unit cell (at the origin).





Presentation of space-group operators

Seitz

xyz

point-shift

augmented
matrix

$$\left(\begin{array}{c|c} R & v \\ \hline 0 & 1 \end{array} \right)$$

$$\{1|000\}$$

$$\{x, y, z\}$$

$$\left\{ \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \begin{pmatrix} 0 \\ 0 \\ 0 \end{pmatrix} \right\}$$

$$\left(\begin{array}{ccc|c} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ \hline 0 & 0 & 0 & 1 \end{array} \right)$$

$$\{2_x|1/2\ 1/2\ 0\}$$

$$\{1/2 + x, 1/2 - y, -z\}$$

$$\left\{ \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & -1 \end{pmatrix}, \begin{pmatrix} 1/2 \\ 1/2 \\ 0 \end{pmatrix} \right\}$$

$$\left(\begin{array}{ccc|c} 1 & 0 & 0 & 1/2 \\ 0 & -1 & 0 & 1/2 \\ 0 & 0 & -1 & 0 \\ \hline 0 & 0 & 0 & 1 \end{array} \right)$$

$$\{2_y|0\ 1/2\ 1/2\}$$

$$\{-x, 1/2 + y, 1/2 - z\}$$

$$\left\{ \begin{pmatrix} -1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{pmatrix}, \begin{pmatrix} 0 \\ 1/2 \\ 1/2 \end{pmatrix} \right\}$$

$$\left(\begin{array}{ccc|c} -1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 1/2 \\ 0 & 0 & -1 & 1/2 \\ \hline 0 & 0 & 0 & 1 \end{array} \right)$$

$$\{2_z|1/2\ 0\ 1/2\}$$

$$\{1/2 - x, -y, 1/2 + z\}$$

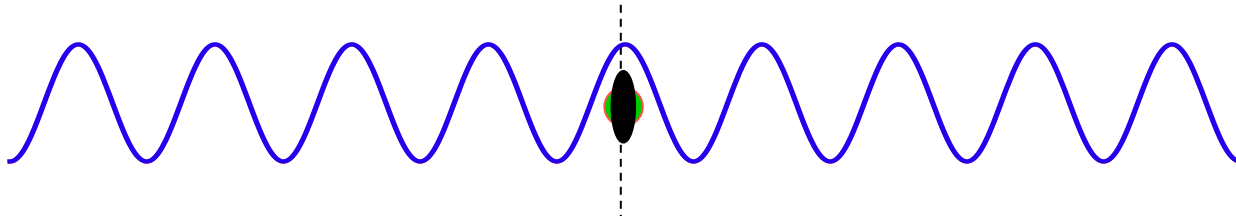
$$\left\{ \begin{pmatrix} -1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \begin{pmatrix} 1/2 \\ 0 \\ 1/2 \end{pmatrix} \right\}$$

$$\left(\begin{array}{ccc|c} -1 & 0 & 0 & 1/2 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & 1 & 1/2 \\ \hline 0 & 0 & 0 & 1 \end{array} \right)$$

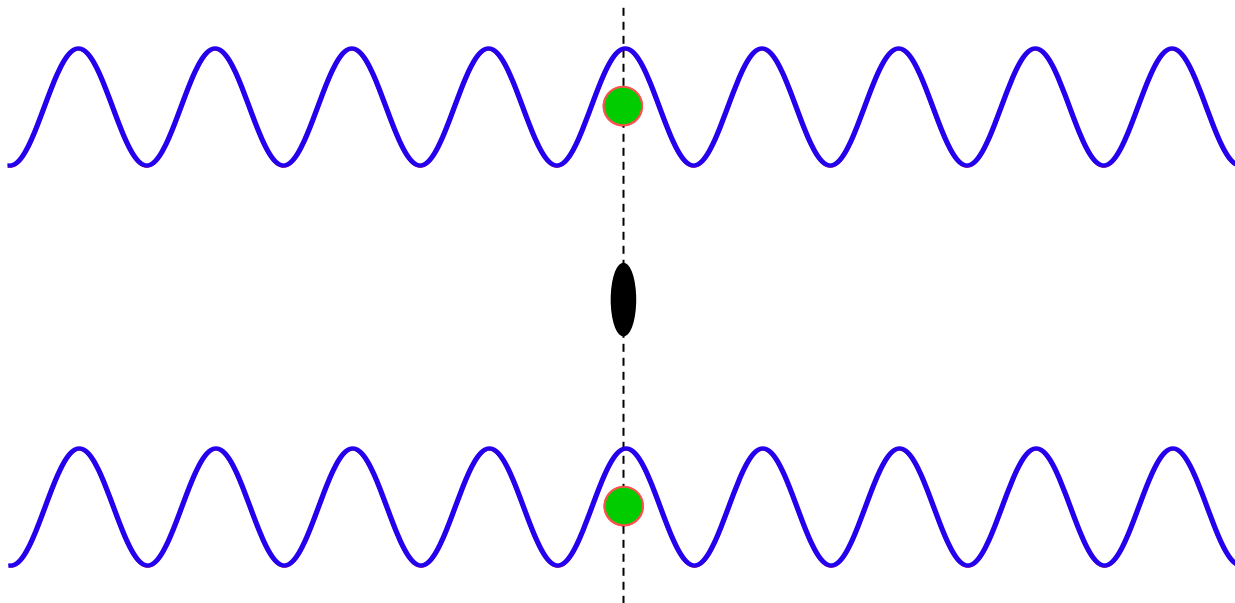
The augmented-matrix form allows translations to be treated via matrix multiplication! Important for theoretical/computational work. Example: space group $P2_12_12_1$ (#19).



Superspace symmetry operations



Space group elements permute the atoms of the structure and transform their local properties. Superspace group elements do this too, but must also permute/transform the incommensurate waves (\mathbf{k} vectors, amplitudes, and phases) attached to those atoms. **Regular space group operations cannot permute/transform the waves.**





Superspace-group matrix operations

Superspace position 3D atom position

$$x = \begin{pmatrix} x_e \\ x_i \\ 1 \end{pmatrix}, \text{ where } x_e = \begin{pmatrix} x_1 \\ x_2 \\ x_3 \end{pmatrix} \text{ and } x_i = \begin{pmatrix} x_{3+1} \\ \vdots \\ x_{3+d} \end{pmatrix} \leftarrow \text{atom phases}$$

$$\left(\begin{array}{cc|c} R & 0 & v \\ M & \epsilon & \delta \\ \hline 0 & 0 & 1 \end{array} \right) \begin{pmatrix} x_e \\ x_i \\ 1 \end{pmatrix} = \begin{pmatrix} R x_e + v \\ M x_e + \epsilon x_i + \delta \\ 1 \end{pmatrix}$$

transformed atom coordinates
transformed modulation phases

External point operation $R(3 \times 3)$ and translation $v(3 \times 1)$ transform atom coordinates, modulation amplitudes, and physical-property tensor components in normal 3-dimensional space.

Internal point operation $\epsilon(d \times d)$ and translation $\delta(d \times 1)$ act in the d -dimensional phase space to transform the modulation phases.

In awkward superspace settings (with rational wave-vector components), the external-space atom position influences the transformation of phases through $M(d \times 3)$.



Superspace-group matrix operations

3D external-space position

d internal-space phases

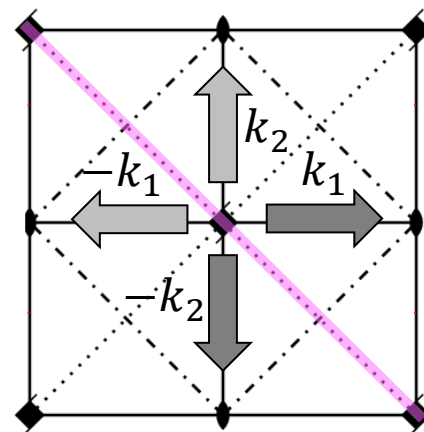
$$\begin{pmatrix} \boxed{R} & 0 & v \\ \boxed{M} & \epsilon & \delta \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} x_e \\ x_i \\ 1 \end{pmatrix} = \begin{pmatrix} \boxed{R x_e + v} \\ \boxed{M x_e + \epsilon x_i + \delta} \\ 1 \end{pmatrix}$$

$P4_2mc(a, 0, 1/2)00q(0, a, 1/2)0sq$

diagonal-plane c glide

$$(-y, -x, z + 1/2, \boxed{z - u + 3/4, z - t + 1/4})$$

$$\begin{pmatrix} \begin{pmatrix} 0 & -1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix} & \begin{pmatrix} 0 & 0 \\ 0 & 0 \\ 0 & 0 \end{pmatrix} & \begin{pmatrix} 0 \\ 0 \\ 1/2 \end{pmatrix} \\ \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 1 \\ 0 & 0 & 0 \end{pmatrix} & \begin{pmatrix} 0 & -1 \\ -1 & 0 \\ 0 & 0 \end{pmatrix} & \begin{pmatrix} -1/4 \\ +1/4 \\ 1 \end{pmatrix} \end{pmatrix} \begin{pmatrix} x \\ y \\ z \\ t \\ u \\ 1 \end{pmatrix}$$



A magnetic superspace-group (MSSG) operation also has a time-reversal component ($\theta = \pm 1$).



Obtain ϵ and M from R and σ

Each row of matrix $\sigma (d \times 3)$ contains the components of a 3D wavevector, which may have irrational (σ^i) and rational (σ^r) parts. The rows of σ^i must be linearly independent!

$$\sigma = \begin{pmatrix} k_{1x} & \cdots & k_{1z} \\ \vdots & \ddots & \vdots \\ k_{dx} & \cdots & k_{dz} \end{pmatrix} = \begin{pmatrix} \alpha & 0 & 1/2 \\ 0 & \alpha & 1/2 \end{pmatrix} = \begin{pmatrix} \alpha & 0 & 0 \\ 0 & \alpha & 0 \end{pmatrix} + \begin{pmatrix} 0 & 0 & 1/2 \\ 0 & 0 & 1/2 \end{pmatrix}$$

R , ϵ , and M contain only integers; these matrices are not independent.

$$\boxed{M = \sigma R - \epsilon \sigma} = (\sigma^i R \overset{0}{\cancel{- \epsilon \sigma^i}}) + (\sigma^r R - \epsilon \sigma^r) \quad \begin{pmatrix} R & 0 & v \\ M & \epsilon & \delta \\ 0 & 0 & 1 \end{pmatrix}$$

$$\Rightarrow \boxed{\sigma^i R = \epsilon \sigma^i \quad \text{and} \quad M = \sigma^r R - \epsilon \sigma^r}$$

ϵ is determined by R and σ^i , and M is determined by R , ϵ , and σ^r . When extending normal operation $\{R, v\}$ to superspace, only δ is not fixed from the start.

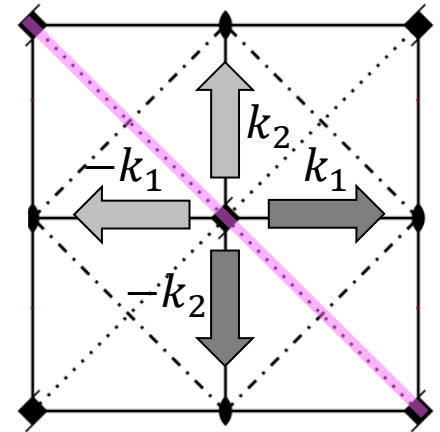


Obtain ϵ and M parts from R and κ

$$\sigma^i R = \epsilon \sigma^i \rightarrow \begin{pmatrix} \alpha & 0 & 0 \\ 0 & \alpha & 0 \end{pmatrix} \begin{pmatrix} 0 & -1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix} = \begin{pmatrix} \epsilon_{11} & \epsilon_{12} \\ \epsilon_{21} & \epsilon_{22} \end{pmatrix} \begin{pmatrix} \alpha & 0 & 0 \\ 0 & \alpha & 0 \end{pmatrix}$$

$$\rightarrow \begin{pmatrix} 0 & -\alpha & 0 \\ -\alpha & 0 & 0 \end{pmatrix} = \begin{pmatrix} \epsilon_{11}\alpha & \epsilon_{12}\alpha & 0 \\ \epsilon_{21}\alpha & \epsilon_{22}\alpha & 0 \end{pmatrix}$$

$$\rightarrow \epsilon = \begin{pmatrix} \epsilon_{11} & \epsilon_{12} \\ \epsilon_{21} & \epsilon_{22} \end{pmatrix} = \begin{pmatrix} 0 & -1 \\ -1 & 0 \end{pmatrix}$$



$$M = \sigma^r R - \epsilon \sigma^r$$

$$= \begin{pmatrix} 0 & 0 & 1/2 \\ 0 & 0 & 1/2 \end{pmatrix} \begin{pmatrix} 0 & -1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix} - \begin{pmatrix} 0 & -1 \\ -1 & 0 \end{pmatrix} \begin{pmatrix} 0 & 0 & 1/2 \\ 0 & 0 & 1/2 \end{pmatrix}$$

$$= \begin{pmatrix} 0 & 0 & 1/2 \\ 0 & 0 & 1/2 \end{pmatrix} - \begin{pmatrix} 0 & 0 & -1/2 \\ 0 & 0 & -1/2 \end{pmatrix} = \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 1 \end{pmatrix}$$



Use superspace centering to zero M

Suppose $\sigma = \begin{pmatrix} k_1 \\ k_2 \end{pmatrix} = \begin{pmatrix} \alpha & 0 & 1/2 \\ 0 & \alpha & 1/2 \end{pmatrix}$. Due to the rational parts, a 90° rotation around $+z$ does not transform k_1 and k_2 into linear combinations of one another.

$$\begin{aligned} M &= \sigma R - \epsilon \sigma \\ &= \begin{pmatrix} \alpha & 0 & 1/2 \\ 0 & \alpha & 1/2 \end{pmatrix} \begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix} - \begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix} \begin{pmatrix} \alpha & 0 & 1/2 \\ 0 & \alpha & 1/2 \end{pmatrix} \\ &= \begin{pmatrix} 0 & -\alpha & 1/2 \\ \alpha & 0 & 1/2 \end{pmatrix} - \begin{pmatrix} 0 & -\alpha & -1/2 \\ \alpha & 0 & 1/2 \end{pmatrix} = \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \end{pmatrix} \end{aligned}$$

In other words, $k_2 \rightarrow k_1$ and $k_1 \rightarrow -k_2 + (0 \ 0 \ 1)$.

We can make this weirdness go away by redefining the third superspace basis vector as $a'_3 = 2a_3 + a_4$. The new cell has a superspace centering vector $\left(00 \frac{1}{2} \frac{1}{2}\right)$, which nicely accounts for the evolution of the wave phase along the z direction. In this new setting, $\sigma = \begin{pmatrix} \alpha & 0 & 0 \\ 0 & \alpha & 0 \end{pmatrix}$ and $M = 0$, so that the phase shifts enacted by the four-fold rotation do **not** depend on atom position. This is a much better way to live!



Use superspace centering to zero M

$Pmna(0\ 1/2\ \gamma)s00 \rightarrow M = \pm(0\ 1\ 0)$ for some operations.

Atom located at $x_e = (0.25\ 0.321\ 0.25)$, so that $Mx_e \neq 0$.

Employ a supercentered cell ($a'_2 = 2a_2 + a_4$) with centering translation $\left(00\ \frac{1}{2}\ \frac{1}{2}\right)$.

Standard setting

```
Symmetry restrictions
structure :

x[Fe1]=0.25
z[Fe1]=0.25
xcos1[Fe1]=-1.5867*xsin1[Fe1]
ycos1[Fe1]=0.63022*ysin1[Fe1]
zcos1[Fe1]=-1.5867*zsin1[Fe1]
xcos2[Fe1]=2.0909*xsin2[Fe1]
ycos2[Fe1]=-0.47826*ysin2[Fe1]
zcos2[Fe1]=2.0909*zsin2[Fe1]
xcos3[Fe1]=0.11677*xsin3[Fe1]
ycos3[Fe1]=-8.5642*ysin3[Fe1]
zcos3[Fe1]=0.11677*zsin3[Fe1]
xcos4[Fe1]=-1.2402*xsin4[Fe1]
ycos4[Fe1]=0.80632*ysin4[Fe1]
zcos4[Fe1]=-1.2402*zsin4[Fe1]
```

Supercentered setting

```
Symmetry restrictions
structure :

x[Fe1]=0.25
z[Fe1]=0.25
ysin1[Fe1]=0
xcos1[Fe1]=0
zcos1[Fe1]=0
ysin2[Fe1]=0
xcos2[Fe1]=0
zcos2[Fe1]=0
ysin3[Fe1]=0
xcos3[Fe1]=0
zcos3[Fe1]=0
ysin4[Fe1]=0
xcos4[Fe1]=0
zcos4[Fe1]=0
```




Magnetic group construct types

A magnetic group M can be constructed by adding time-reversal (indicated as a *prime*') to operations of a related non-magnetic point group, space group, or superspace group called F .

The construction can be implemented in four different ways.

Type 1: $M = F$, no primed operations (colorless).

Type 2: $M = F + F\mathbf{1}'$, primed and unprimed copies of each operation (grey).

Type 3: $M = D + (F - D)\mathbf{1}'$ for each equi-translation index-2 subgroup D of F (bi-colored group with bi-colored MPG).

Type 4: $M = D + (F - D)\mathbf{1}'$ for each equi-class index-2 subgroup D of F (bi-colored group with grey MPG, contains anti-translations).

Each non-magnetic SSG (F) can give us many MSSGs (M)!



MSSGs constructed from SSG = 16.1.9.1

For each non-magnetic SSG, test every group-forming way to add primes to the SSG generators, and isolate unique results as MSSGs.

Type 1:	ssg.m1.1	P222 (0,0,g) 000	2x	2y	2z	
Type 3:	ssg.m3.1	P2'2'2 (0,0,g) 000	2x'	2y'	2z	
	ssg.m3.2	P22'2' (0,0,g) 000	2x	2y'	2z'	
Type 2:	ssg.m2.1	P2221' (0,0,g) 0000	2x	2y	2z	(0,0,0,0)'
	ssg.m2.2	P222.1' (0,0,g) 000s	2x	2y	2z	(0,0,0,1/2)'
	ssg.m4.1	P222.1' _a (0,0,g) 0000	2x	2y	2z	(1/2,0,0,0)'
	ssg.m4.2	P222.1' _c (0,0,g) 0000	2x	2y	2z	(0,0,1/2,0)'
Type 4:	ssg.m4.3	P222.1' _a (0,0,g) 000s	2x	2y	2z	(1/2,0,0,1/2)'
	ssg.m5.1	P222.1' _C (0,0,g) 0000	2x	2y	2z	(1/2,1/2,0,0)'
	ssg.m5.2	P222.1' _B (0,0,g) 0000	2x	2y	2z	(1/2,0,1/2,0)'
	ssg.m5.3	P222.1' _C (0,0,g) 000s	2x	2y	2z	(1/2,1/2,0,1/2)'
	ssg.m6.1	P222.1' _I (0,0,g) 0000	2x	2y	2z	(1/2,1/2,1/2,0)'

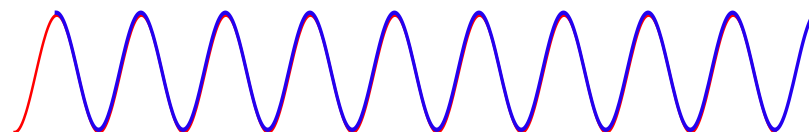




MSSGs constructed from SSG = 16.1.9.1

For each non-magnetic SSG, test every group-forming way to add primes to the SSG generators, and isolate unique results as MSSGs.

Type 1:	ssg.m1.1	P222 (0,0,g) 000	2x	2y	2z	
Type 3:	ssg.m3.1	P2'2'2 (0,0,g) 000	2x'	2y'	2z	
	ssg.m3.2	P22'2' (0,0,g) 000	2x	2y'	2z'	
Type 2:	ssg.m2.1	P2221' (0,0,g) 0000	2x	2y	2z	(0,0,0,0)'
	ssg.m2.2	P222.1' (0,0,g) 000s	2x	2y	2z	(0,0,0,1/2)'
	ssg.m4.1	P222.1' _a (0,0,g) 0000	2x	2y	2z	(1/2,0,0,0)'
	ssg.m4.2	P222.1' _c (0,0,g) 0000	2x	2y	2z	(0,0,1/2,0)'
Type 4:	ssg.m4.3	P222.1' _a (0,0,g) 000s	2x	2y	2z	(1/2,0,0,1/2)'
	ssg.m5.1	P222.1' _C (0,0,g) 0000	2x	2y	2z	(1/2,1/2,0,0)'
	ssg.m5.2	P222.1' _B (0,0,g) 0000	2x	2y	2z	(1/2,0,1/2,0)'
	ssg.m5.3	P222.1' _C (0,0,g) 000s	2x	2y	2z	(1/2,1/2,0,1/2)'
	ssg.m6.1	P222.1' _I (0,0,g) 0000	2x	2y	2z	(1/2,1/2,1/2,0)'





$(3 + d)$ D SSGs for $d = 1, 2, 3$



Acta Cryst. A67, 45-55 (2011).

research papers

Acta Crystallographica Section A
**Foundations of
Crystallography**
ISSN 0108-7673

Generation of $(3 + d)$ -dimensional superspace groups for describing the symmetry of modulated crystalline structures

Received 14 July 2010
Accepted 18 October 2010

Harold T. Stokes,^{a*} Branton J. Campbell^a and Sander van Smaalen^b

^aDepartment of Physics and Astronomy, Brigham Young University, Provo, Utah 84602, USA, and
^bLaboratory of Crystallography, University of Bayreuth, Bayreuth, Germany.
Correspondence e-mail: stokes@byu.edu



Acta Cryst. A69, 75-90 (2013).

research papers

Acta Crystallographica Section A
**Foundations of
Crystallography**
ISSN 0108-7673

Equivalence of superspace groups

Sander van Smaalen,^{a*} Branton J. Campbell^b and Harold T. Stokes^b

Received 2 July 2012
Accepted 5 October 2012

^aLaboratory of Crystallography, University of Bayreuth, Bayreuth, Germany, and ^bDepartment of Physics and Astronomy, Brigham Young University, Provo, Utah 84602, USA. Correspondence e-mail: smash@uni-bayreuth.de

Modulations	$d = 1$	$d = 2$	$d = 3$
SSG count	775	3338	12584



$(3 + d)$ D MSSGs for $d = 1, 2, 3$

research papers



FOUNDATIONS
ADVANCES

ISSN 2053-2733

Enumeration and tabulation of magnetic $(3+d)$ -dimensional superspace groups

Harold T. Stokes and Branton J. Campbell*

Physics and Astronomy, Brigham Young University, Provo, Utah 84602, USA. *Correspondence e-mail: branton_campbell@byu.edu

Received 14 January 2022

Accepted 10 April 2022

A magnetic superspace group (MSSG) simultaneously constrains both the magnetic and non-magnetic (*e.g.* displacive, occupational, rotation and strain)



MSSG enumeration/tabulation

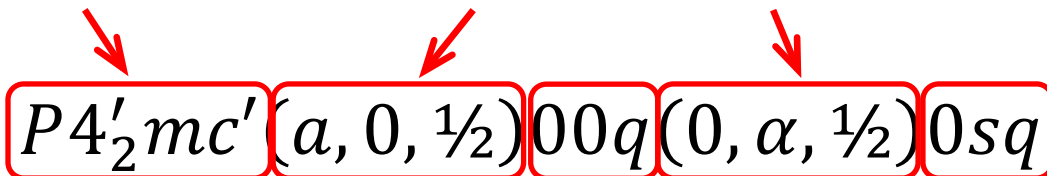
	MPG	MSG (d=0)	MSSG (d=1)	MSSG (d=2)	MSSG (d=3)
Type 1	32	230	775	3338	12584
Type 2	32	230	775	3338	12584
Type 3	58	674	3100	15218	60799
Type 4	--	517	4653	31862	176101
Total	122	1651	9303	53756	262068

Why bother when there are so many? After detecting the MSSG elements of an incommensurate structure (already a challenge) in an arbitrary setting, it's very difficult to subsequently compare its symmetry to that of other known structures. One would need to run an equivalence test on every structure of interest, which is not practical. With an exhaustive table, we need only compare to reference setting of each tabulated MSSG with similar properties (e.g. Bravais class, MBSG).



MSSG symbols and numbers

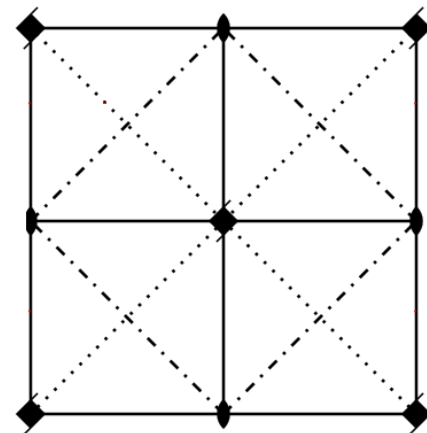
Magnetic basic space group (MBSG) propagation vectors (in standard setting)



$$\begin{pmatrix} R & 0 & v \\ M & \epsilon & \delta \\ 0 & 0 & 1 \end{pmatrix}$$

δ components (phase shifts) for each \mathbf{k} and each PG generator in the supercentered setting

$$\begin{array}{ll} s & 1/2 \\ t, \bar{t} & \pm 1/3 \\ q, \bar{q} & \pm 1/4 \\ h, \bar{h} & \pm 1/6 \end{array}$$



Bravais class = 2.65 $P4/mmm(a, 0, \frac{1}{2})0000(0, \alpha, \frac{1}{2})0000$

FSSG = #105.2.65.9 $P4_2mc(a, 0, \frac{1}{2})00q(0, a, \frac{1}{2})0sq$

MBSG = #105.214 $P4'_2mc'$

MSSG = #105.2.65.9.m214.1 $P4'_2mc'(a, 0, \frac{1}{2})00q(0, a, \frac{1}{2})0sq$

1st and only MSSG with this FSSG and MBSG



DyMn₆Ge₆ double cone

DyMn₆Ge₆ (#1.1.10)

$P62'2'(00\gamma)h00$

177.1.24.

$a = 5.20810,$

Single- k ferrima

have incommens

opposing comm

Type-3 MSSG with

1 $x1, x2, x3, x4, +1$

2 $x1-x2, x1, x3, x4+$

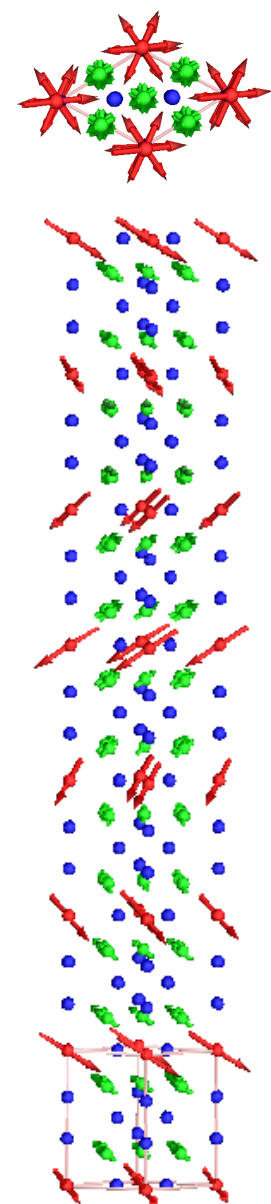
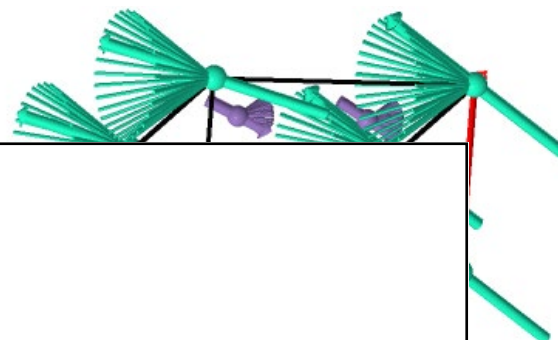
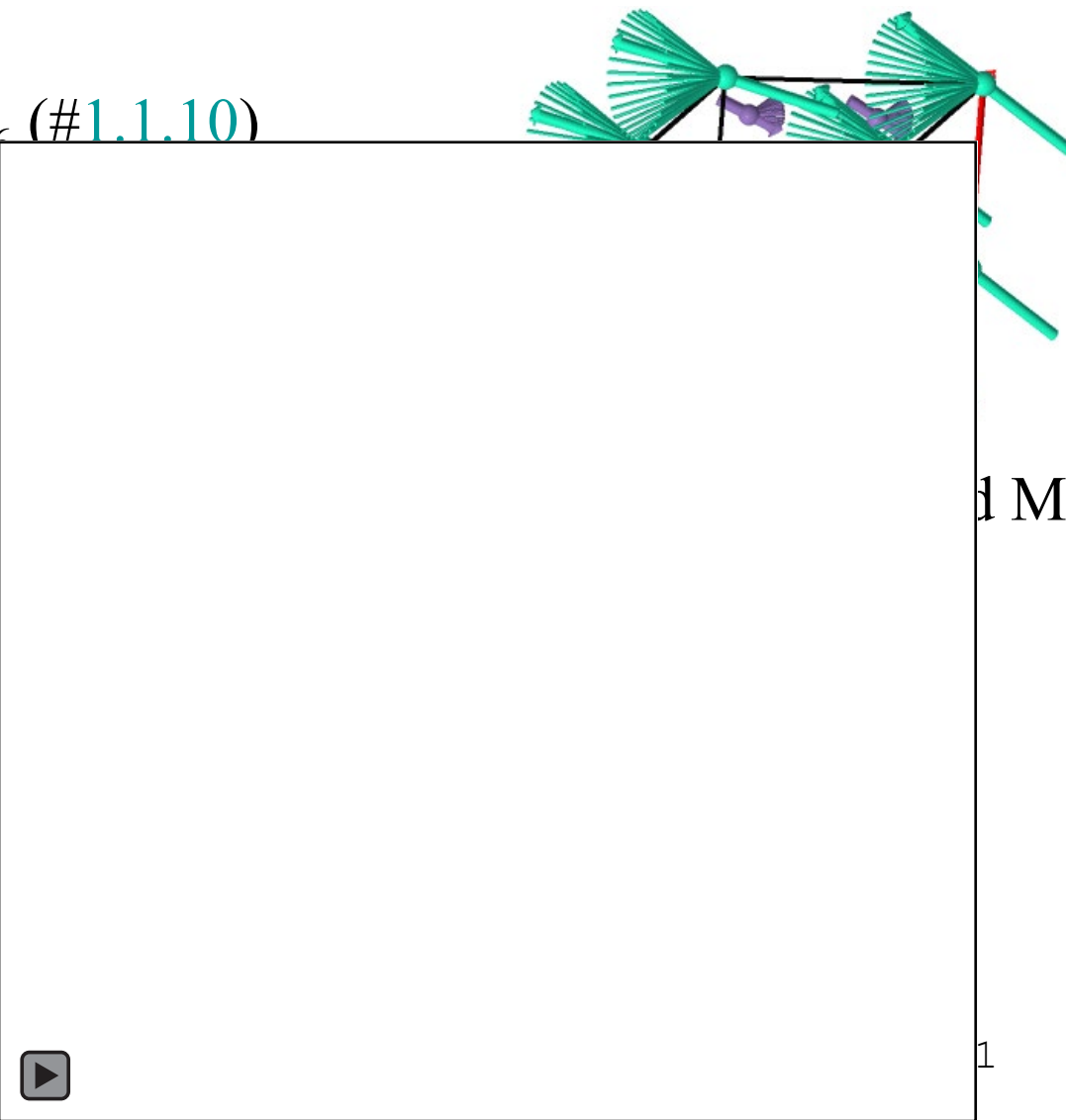
3 $-x2, x1-x2, x3, x4$

4 $-x1, -x2, x3, x4+1$

5 $-x1+x2, -x1, x3, x$

6 $x2, -x1+x2, x3, x4+5/6, +1$

12 $-x2, -x1, -x3, -x4+1/6, -1$



d Mn

1

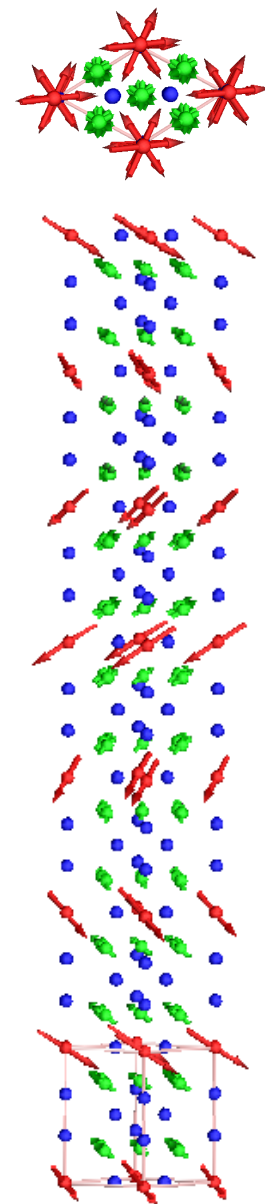


DyMn₆Ge₆ double cone

DyMn₆Ge₆ 177.1.24.2.m153.1 $P6_2'2'(00\gamma)h00$ with $\gamma = 0.165$

atom	position and moment			symmform
Dy1 1a	[0.00000	0.00000	0.00000]	0, 0, 0
	[0.00000	0.00000	-3.93200]	0, 0, Mz
Mn1 6i	[0.50000	0.00000	0.74900]	0, 0, Dz
	[0.00000	0.00000	1.14900]	0, 0, Mz
Ge1 2c	[0.33330	0.66667	0.00000]	0, 0, 0
Ge2 2d	[0.33330	0.66667	0.50000]	0, 0, 0
Ge3 2e	[0.00000	0.00000	0.65530]	0, 0, Dz

param	A_cos	A_sin	symmform	
Dy1_Mx	[7.15400	0.00000]	Mxc1	0
Dy1_My	[3.57700	6.19555]	(Mxc1) / 2	Mxc1*sqrt(3/4)
Dy1_Mz	[0.00000	0.00000]	0	0
Mn1_Mx	[-2.04846	0.20586]	Mxc1	Mxs1
Mn1_My	[-1.20287	-1.67142]	Myc1	Mys1
Mn1_Mz	[0.00000	0.00000]	0	0





DyMn₆Ge₆ double cone

Dysprosium:

$$|M_x| = 7.15400 \mu_B$$

$$|M_y| = \sqrt{3.57700^2 + 6.19555^2} = 7.15400 \mu_B$$

$$\phi(M_x) = \arctan(0/7.15400) = 0^\circ$$

$$\phi(M_y) = \arctan(+6.19555/+3.57700) = +60^\circ$$

$$\phi(M_y) - \phi(M_x) = +60^\circ$$

Manganese:

$$|M_x| = \sqrt{2.04846^2 + 0.20586^2} = 2.05878 \mu_B$$

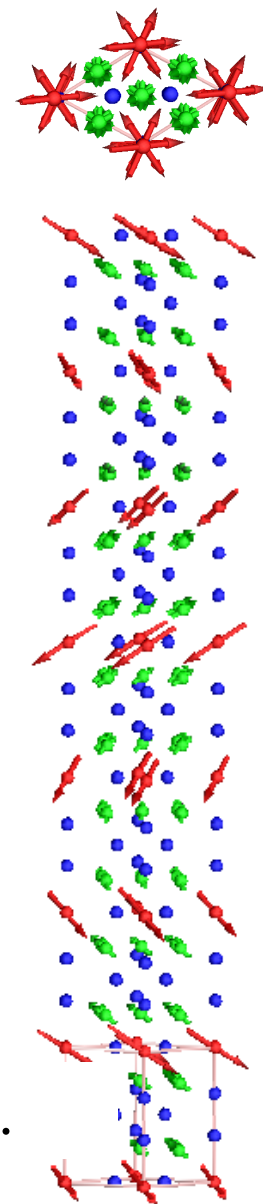
$$|M_y| = \sqrt{1.20287^2 + 1.67142^2} = 2.05878 \mu_B$$

$$\phi(M_x) : \arctan(+0.20586/-2.04846) = +174.26133^\circ$$

$$\phi(M_y) : \arctan(-1.67142/-1.20287) = +234.26133^\circ$$

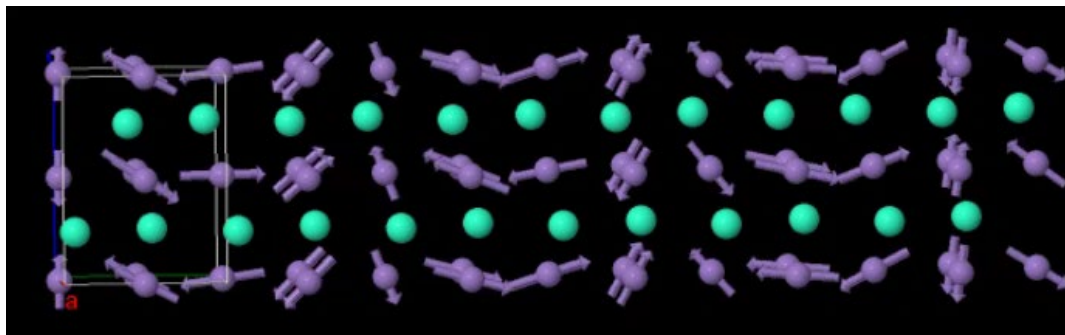
$$\phi(M_y) - \phi(M_x) = +60^\circ$$

Circular Dy spiral runs nearly 180° ahead of circular Mn spiral.





Multi-ferroic TbMnO_3

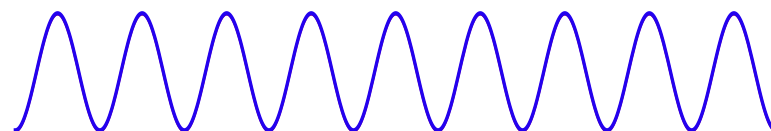


TbMnO_3 (#[1.1.7](#)) $Pbn2_1.1'(0\beta0)000s$ 33.1.9.5.m145.2

- 1 $x1, x2, x3, x4, +1$
- 2 $-x1, -x2, x3+1/2, -x4, +1$
- 3 $x1+1/2, -x2+1/2, x3+1/2, -x4, +1$
- 4 $-x1+1/2, x2+1/2, x3, x4, +1$

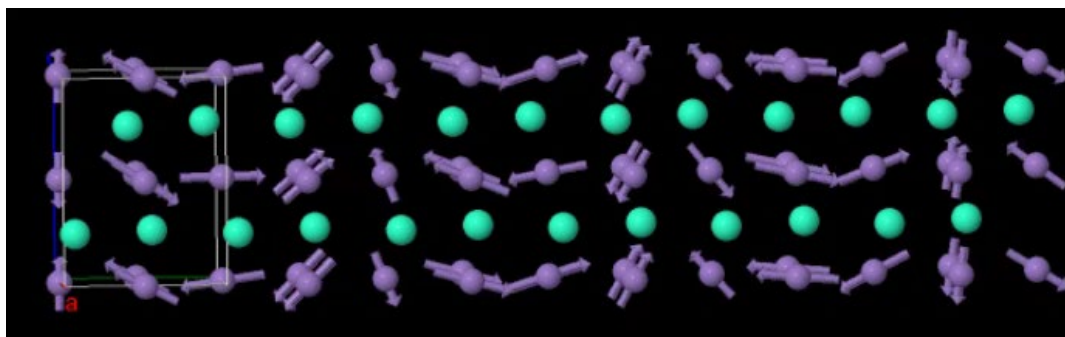
- 5 $x1, x2, x3, x4+1/2, -1$
- 6 $-x1, -x2, x3+1/2, -x4+1/2, -1$
- 7 $x1+1/2, -x2+1/2, x3+1/2, -x4+1/2, -1$
- 8 $-x1+1/2, x2+1/2, x3, x4+1/2, -1$

Type-4 MSSG with anti-translation $(0,0,0, 1/2)'$





Multi-ferroic TbMnO₃



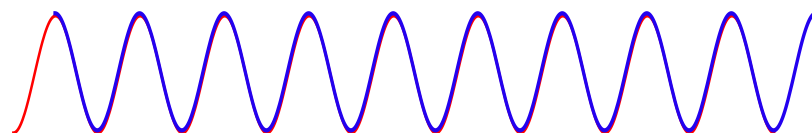
TbMnO₃ (#[1.1.7](#)) $Pbn2_1.1'(0\beta0)000s$ 33.1.9.5.m145.2

- | | | | |
|---|------------------------------------|---|--|
| 1 | $x1, x2, x3, x4, +1$ | 5 | $x1, x2, x3, x4+1/2, -1$ |
| 2 | $-x1, -x2, x3+1/2, -x4, +1$ | 6 | $-x1, -x2, x3+1/2, -x4+1/2, -1$ |
| 3 | $x1+1/2, -x2+1/2, x3+1/2, -x4, +1$ | 7 | $x1+1/2, -x2+1/2, x3+1/2, -x4+1/2, -1$ |
| 4 | $-x1+1/2, x2+1/2, x3, x4, +1$ | 8 | $-x1+1/2, x2+1/2, x3, x4+1/2, -1$ |

Mn_Mx	0.00000	0.00000	Mxc1	Mxs1
Mn_My	0.00000	-2.82843	Myc1	Mys1
Mn_Mz	-2.82843	0.00000	Mzc1	Mzs1

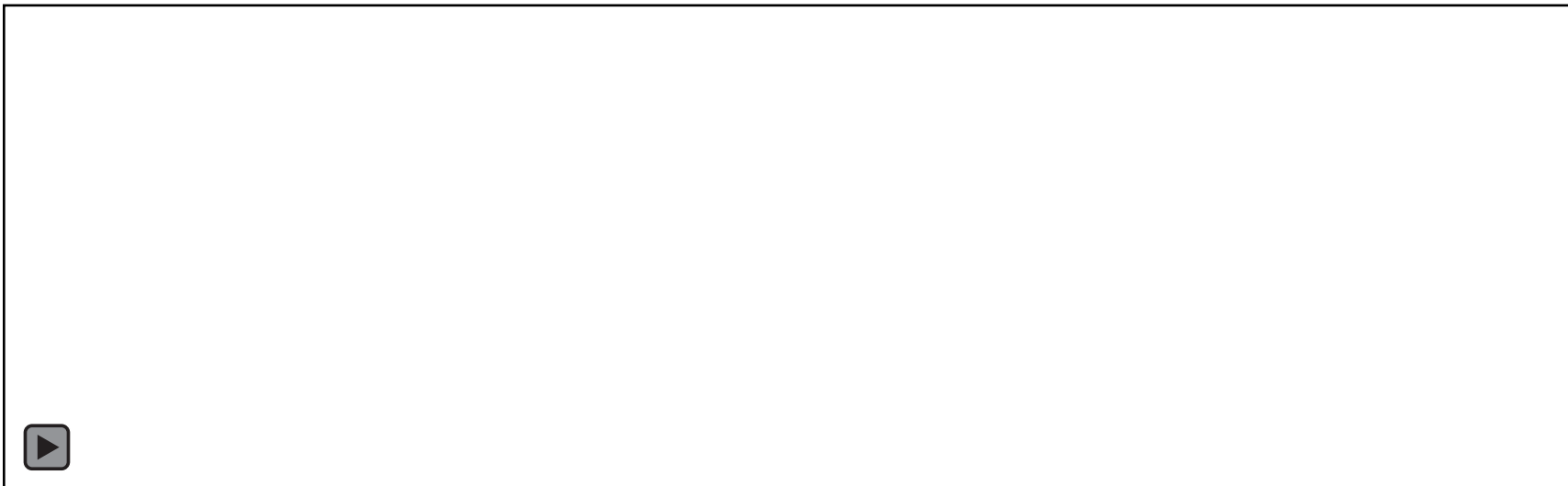
Type-4 MSSG with anti-translation $(0,0,0, 1/2)'$

No MSSG symmetry constraints on the modulation amplitudes, which are instead associated with relevant multi-dimensional irreps/OPDs: $m\Sigma_3(a, 0) + m\Sigma_2(b, 0)$.





Multi-ferroic TbMnO₃



TbMnO₃ (#[1.1.7](#)) $Pbn2_1.1'(0\beta0)000s$ 33.1.9.5.m145.2

Transverse and longitudinal modulations are superposed 90° out of phase, forming an incommensurate single- k magnetic cycloid.

Acting together, these two magnetic modulations break inversion symmetry and couple to a secondary ferroelectric moment, making the material an important multiferroic.

Magnetic skyrmion lattice

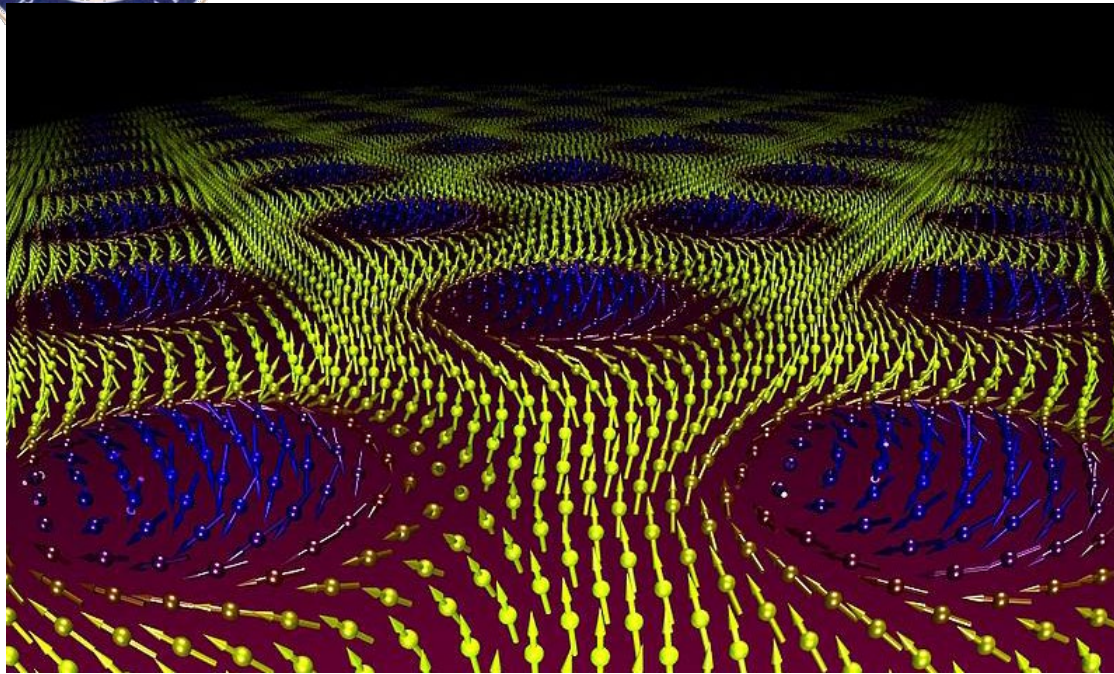
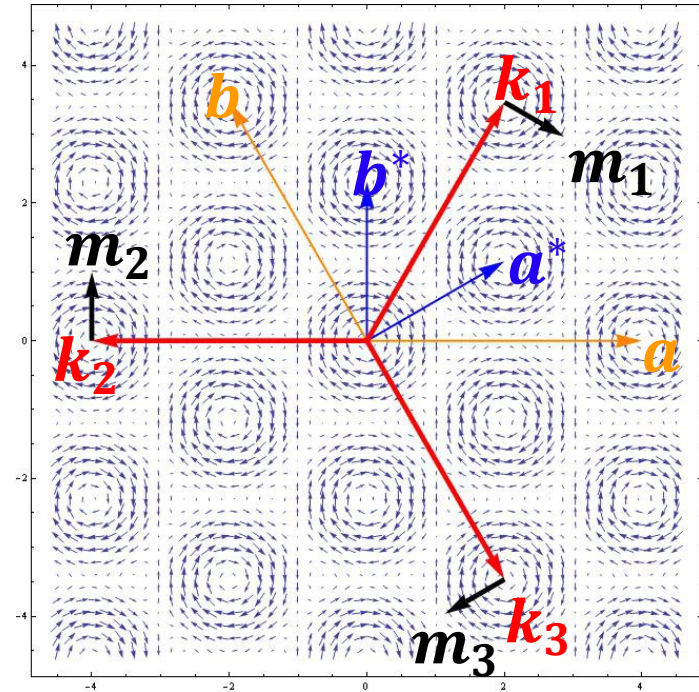


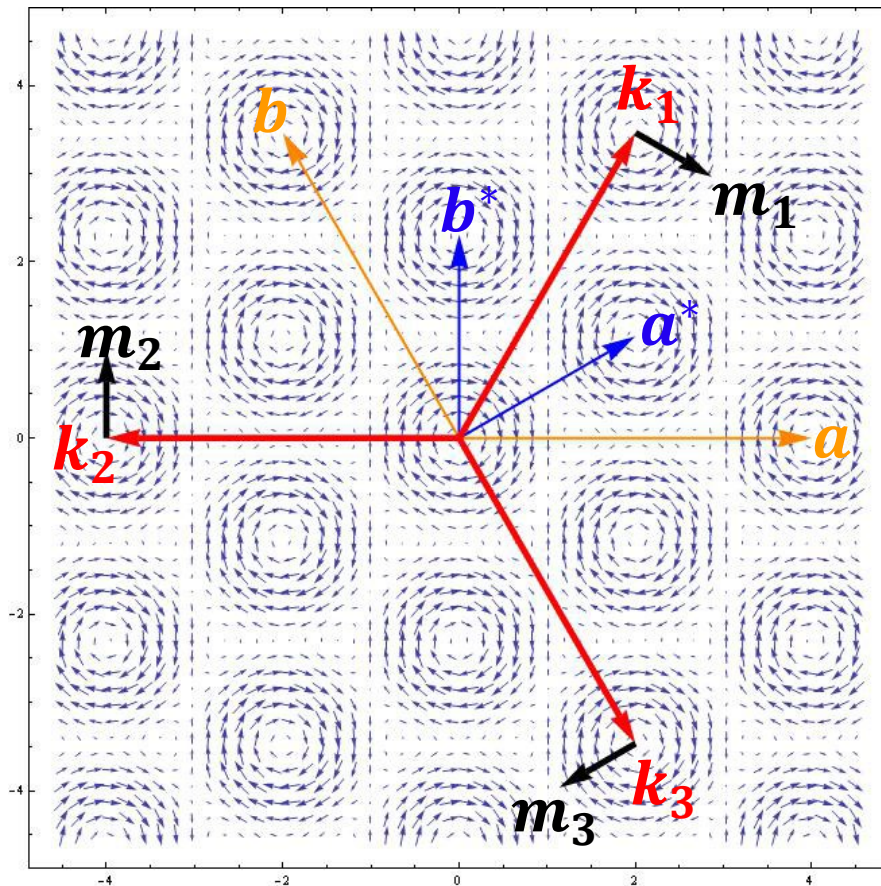
Image from Mühlbauer et al., Science 323, 915-919 (2009).



Skyrmion lattice. Three phase-locked transverse waves in the plane yield an incommensurate 2D lattice of magnetic vortices on a triangular arrangement of magnetic atom. Their topological stability makes skyrmions interesting for information storage, magneto-electronic devices, and quantum computing.



2D skyrmion lattice



Three incommensurate transverse waves, locked in phase, form this remarkable 2D magnetic skyrmion-lattice. The in-plane component of the pattern has only one adjustable magnetic degree of freedom. The out-of-plane component is not shown.

One k vector depends on the other two, so that there are only two independent superspace dimensions.

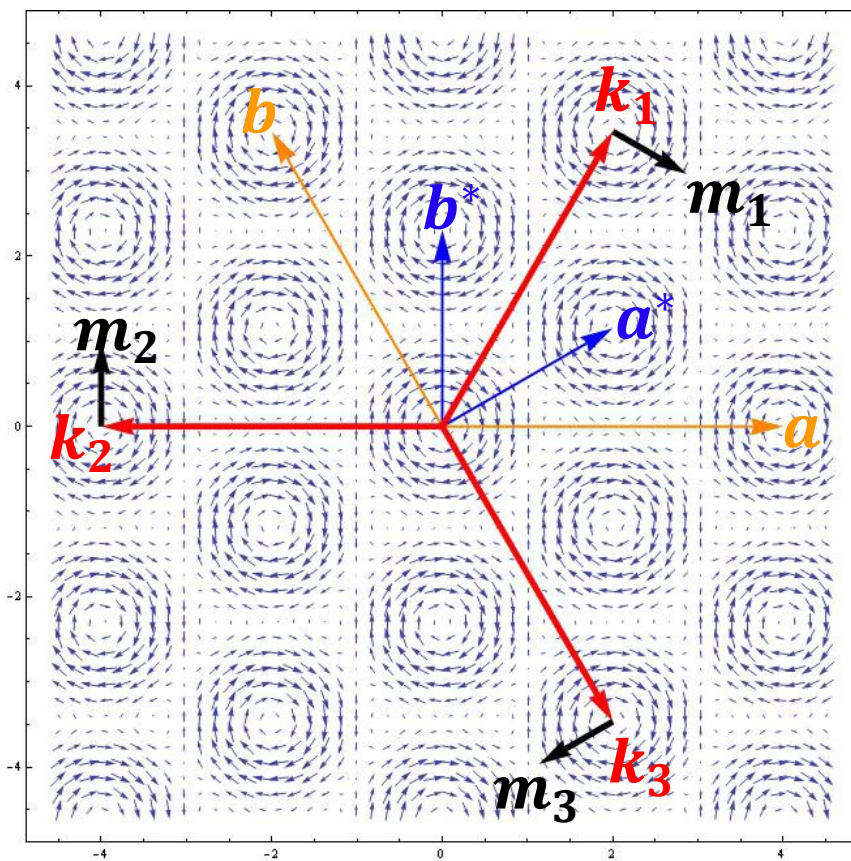
$$k_1 = (\alpha, \alpha, 0), \quad k_2 = (-2\alpha, \alpha, 0), \quad k_3 = (\alpha, -2\alpha, 0) \rightarrow k_1 + k_2 + k_3 = 0$$



2D skyrmion lattice – magnetic superspace group

MSSG: 177.2.83.6.m153.1 $P62'2'(a, a, 0)000(-2a, a, 0)000$

Bravais class: 2.83 $P6/mmm(a, a, 0)(-2a, a, 0)$



(3+2)D type-3 MSSG.

Representative non-lattice operations:

$$\begin{aligned}
 &(x, y, z, t, u) \\
 &(-x, -y, z, -t, -u) \\
 &(-y, x - y, z, -t - u, t) \\
 &(-x + y, -x, z, u, -t - u) \\
 &(x - y, x, z, -u, t + u) \\
 &(y, -x + y, z, t + u, -t) \\
 &(x - y, -y, -z, -t - u, u)' \\
 &(y, x, -z, t, -t - u)' \\
 &(-x, -x + y, -z, u, t)' \\
 &(x, x - y, -z, -u, -t)' \\
 &(-x + y, y, -z, t + u, -u)' \\
 &(-y, -x, -z, -t, t + u)'
 \end{aligned}$$

Symmetry constraints on the modulations leave only one degree of freedom (m).

$$m_{1(sin)} = (m, -m, 0), \quad m_{2(sin)} = (m, 2m, 0), \quad m_{3(sin)} = (-2m, -m, 0)$$

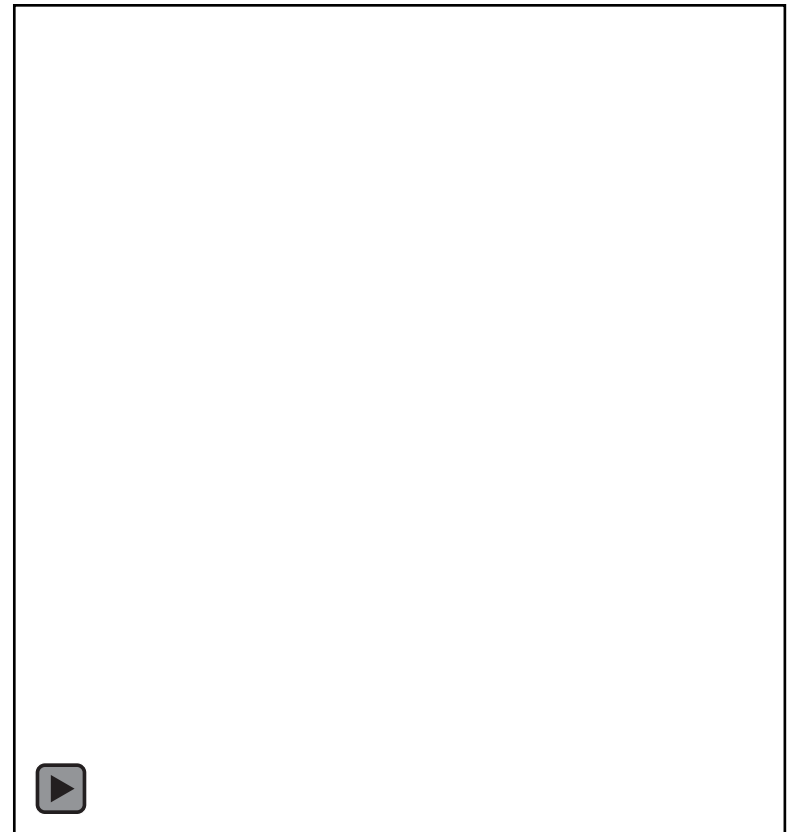
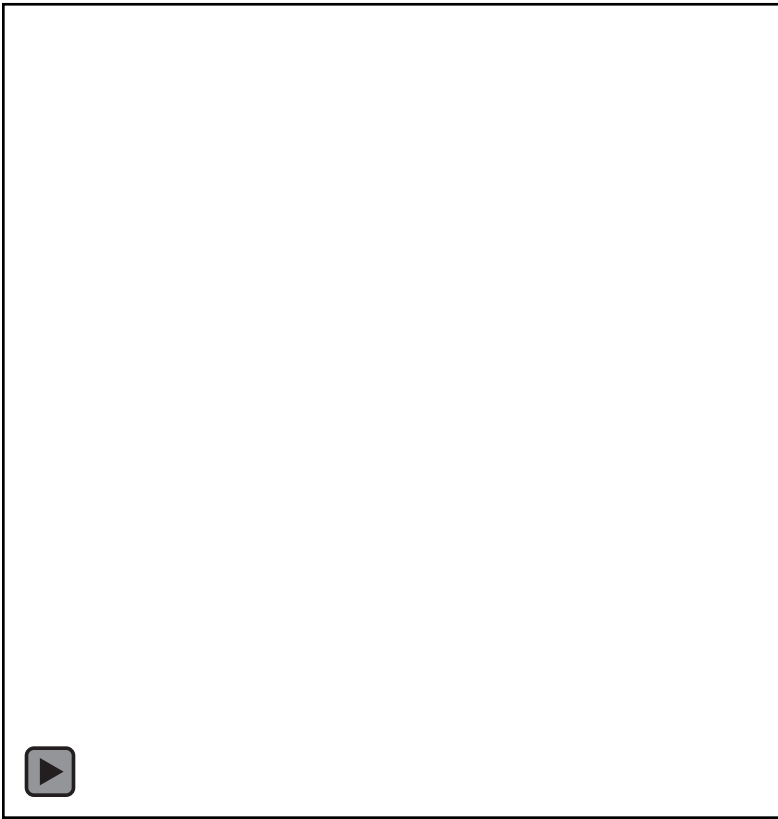


Other interesting multi-k cases

CeAlGe (#[2.1.1](#))

$I4_1md. 1'(\alpha, 0, 0)000s(0, \alpha, 0)0s0s$

Two orthogonal incommensurate cycloids from same \mathbf{k} -star superpose to yield an unusual topological spin texture.



MnGe (#[3.1.1](#))

$P2_13.1'(\alpha, 0, 0)00s(0, \alpha, 0)00s(0, 0, \alpha)00s$

Three orthogonal incommensurate modulation from same \mathbf{k} -star superpose to yield a fully 3D topological spin texture.



Summary

- Incommensurate modulations break 3D translational periodicity; internal superspace (phase) dimensions restore periodicity, but in $3+d$ dimensions.
- One superspace dimension independent for \mathbf{k} vector (irrational part).
- Every active \mathbf{k} vector is an integer linear combination of the independent ones.
- Modulations can be magnetic, displacive, occupational, or rotational.
- Superspace symmetry operations must transform both the local atom properties (position, occupancy, ADPs) and their waves (\mathbf{k} -vector, vector amplitude, phase).
- MSSGs are constructed from SSGs by adding time reversal to some operations. The four basic constructs are like those of commensurate MSGs. Most structures with type-4 MSSGs have an internal-space anti-translation, e.g. $(000\frac{1}{2})'$.
- There are so many $(3+d)$ D MSSGs; always provide reference transformation!
- Magnetic modulations impact material properties.
- Multi- \mathbf{k} structures from a single \mathbf{k} -star have especially interesting spin textures.



Tools available

- ISOSPACEGROUP and ISO(3+D) list generators, centering vectors, and non-lattice operations for each of 325,127 MSSGs in a standard reference setting, along with those of a supercentered setting where appropriate. Released for testing in May 2019 and corrected/updated June 2020 (<https://iso.byu.edu>).
- Since Jul 2020, FINDSYM and ISOCIF can detect and enforce the SSG/MSSG symmetry (or pseudosymmetry within tolerance) of an incommensurate structure, and ISOCIF can transform the symmetrized result into any desired SSG/MSSG setting.
- ISODISTORT finds incommensurate isotropy subgroups, uniquely identifies the SSG/MSSG and the transformation to a reference setting, projects the modulation waves, and symmetry-restricts the modulation amplitudes.
- The reliable identification and comparison of virtually any two incommensurate magnetic structures is accomplished by combining an exhaustive MSSG tabulation, a MSSG setting-transformation tool, a robust MSSG detection routine, and a very fast MSSG equivalence test.