Irreducible representations and the superspace formalism

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to Modulated Crystal Structures
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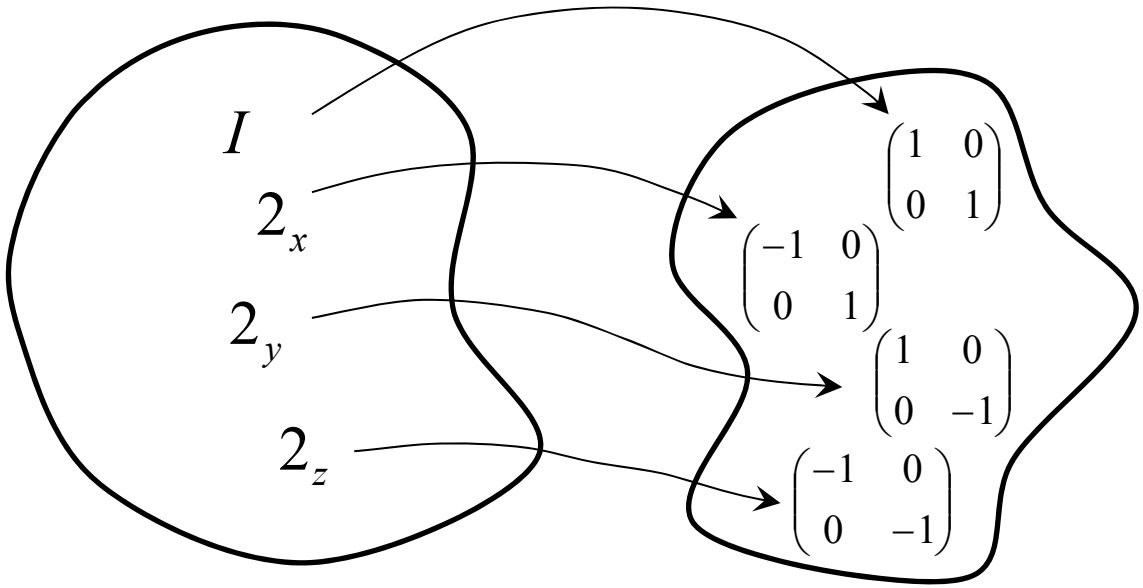
Group representations



Point group: 222

	I	2_x	2_y	2_z
I	I	2_x	2_y	2_z
2_x	2_x	I	2_z	2_y
2_y	2_y	2_z	I	2_x
2_z	2_z	2_y	2_x	I

Representations map group elements onto matrices that obey the same multiplication table as the group.

$$2_x\,2_y = 2_z \qquad \rightarrow \qquad \begin{pmatrix}-1 & 0\\ 0 & 1\end{pmatrix}\begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix} = \begin{pmatrix}-1 & 0\\ 0 & -1\end{pmatrix}$$

Irreducible Representations (irreps)

$R1$: $1 \to (1)$ $2_x \to (\bar{1})$ $2_y \to (1)$ $2_z \to (\bar{1})$

$R2$: $1 \to (1)$ $2_x \to (1)$ $2_y \to (\bar{1})$ $2_z \to (\bar{1})$

$R3$: $1 \to \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}$ $2_x \to \begin{pmatrix} \bar{1} & 0 \\ 0 & 1 \end{pmatrix}$ $2_y \to \begin{pmatrix} 1 & 0 \\ 0 & \bar{1} \end{pmatrix}$ $2_z \to \begin{pmatrix} \bar{1} & 0 \\ 0 & \bar{1} \end{pmatrix}$

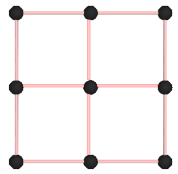
Reducible representation: $R3 = R1 \oplus R2 = \begin{pmatrix} R1 & 0 \\ 0 & R2 \end{pmatrix}$

Irreducible representations can't be separated into smaller pieces!

Irreps are **recipes for symmetry breaking**!

Irrep recipe for symmetry breaking

Example: Γ_5 irrep of space group $P4mm$



$$\begin{array}{cccccccc} 1 & m_y & m_x & 2_z & m_{\bar{x}z} & +4_z & -4_z & m_{xz} \\ g_1 & g_2 & g_3 & g_4 & g_5 & g_6 & g_7 & g_8 \end{array}$$

$$G = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & \bar{1} \end{pmatrix} \begin{pmatrix} \bar{1} & 0 \\ 0 & 1 \end{pmatrix} \begin{pmatrix} \bar{1} & 0 \\ 0 & \bar{1} \end{pmatrix} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \begin{pmatrix} 0 & \bar{1} \\ 1 & 0 \end{pmatrix} \begin{pmatrix} 0 & 1 \\ \bar{1} & 0 \end{pmatrix} \begin{pmatrix} 0 & \bar{1} \\ \bar{1} & 0 \end{pmatrix}$$

Find the group elements whose matrices leave some vector invariant.

$\begin{pmatrix} a \\ 0 \end{pmatrix} \Rightarrow \{g_1, g_2\}$ $\quad \{1, m_y\}$ $\quad Pm$

$\begin{pmatrix} a \\ a \end{pmatrix} \Rightarrow \{g_1, g_5\}$ $\quad \{1, m_{\bar{x}z}\}$ $\quad Cm$

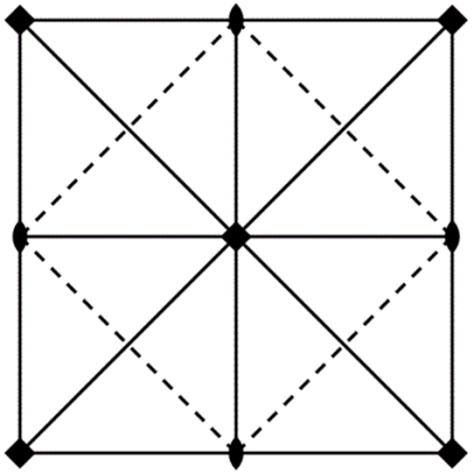
$\begin{pmatrix} a \\ b \end{pmatrix} \Rightarrow \{g_1\}$ $\quad \{1\}$ $\quad P1$

The vector used is called the *order parameter direction* or OPD.
The resulting symmetry is called an *isotropy subgroup* of the parent.

Abstract vs Concrete

Irrep (Γ_5)

$$\begin{pmatrix}1 & 0\\0 & 1\end{pmatrix}\ \begin{pmatrix}1 & 0\\0 & \bar{1}\end{pmatrix}\ \begin{pmatrix}\bar{1} & 0\\0 & 1\end{pmatrix}\ \begin{pmatrix}\bar{1} & 0\\0 & \bar{1}\end{pmatrix}$$
$$\begin{pmatrix}0 & 1\\1 & 0\end{pmatrix}\ \begin{pmatrix}0 & \bar{1}\\1 & 0\end{pmatrix}\ \begin{pmatrix}0 & 1\\\bar{1} & 0\end{pmatrix}\ \begin{pmatrix}0 & \bar{1}\\\bar{1} & 0\end{pmatrix}$$



Space group ($P4mm$)

OPDs

$$\begin{pmatrix}a\\0\end{pmatrix}\quad\begin{pmatrix}a\\a\end{pmatrix}\quad\begin{pmatrix}a\\b\end{pmatrix}$$

Basis functions / order parameters

Shear strains along a and b axes.

Invariant/isotropy subgroups

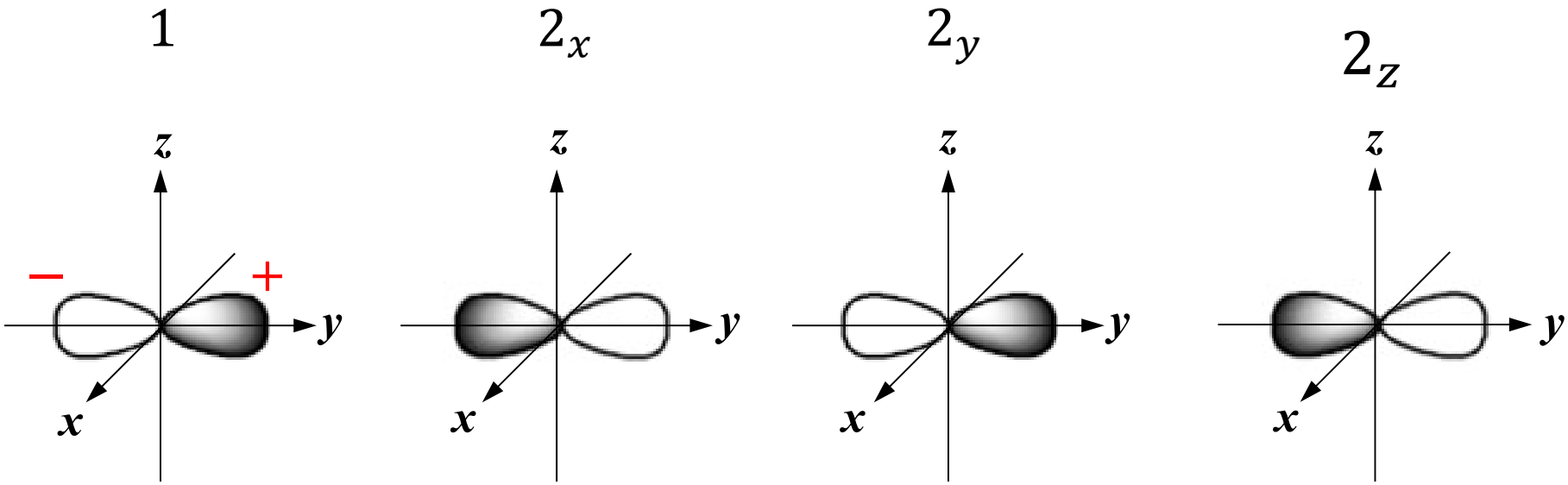
$$\begin{pmatrix}1 & 0\\0 & 1\end{pmatrix}\begin{pmatrix}1 & 0\\0 & \bar{1}\end{pmatrix}\ \begin{pmatrix}1 & 0\\0 & 1\end{pmatrix}\begin{pmatrix}0 & 1\\1 & 0\end{pmatrix}\ \begin{pmatrix}1 & 0\\0 & 1\end{pmatrix}$$

Actual subgroups

$\{1, m_y\}$	$\{1, m_{\bar{x}z}\}$	$\{1\}$
Pm	Cm	$P1$

Irrep basis functions

Parent symmetry = 222

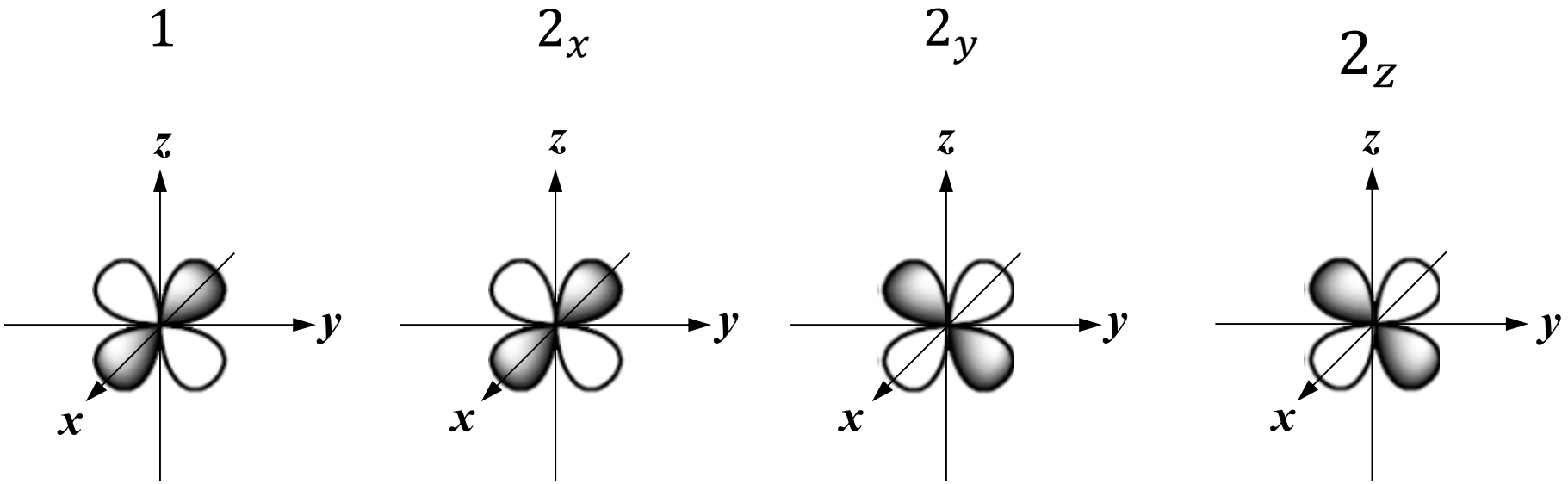


Under the symmetry operations of the group, a p_y orbital transforms the same way as which irrep?

	1	$\mathbf{2}_x$	$\mathbf{2}_y$	$\mathbf{2}_z$	
Γ_1	1	1	1	1	
Γ_2	1	−1	−1	1	p_z
Γ_3	1	1	−1	−1	p_x
Γ_4	1	−1	1	−1	p_y

Irrep basis functions

Parent symmetry = 222



Under the symmetry operations of the group, a d_{yz} orbital transforms the same way as which irrep?

	1	$\mathbf{2}_x$	$\mathbf{2}_y$	$\mathbf{2}_z$	
Γ_1	1	1	1	1	$d_{x^2-y^2}, d_{z^2}$
Γ_2	1	−1	−1	1	p_z, d_{xy}
Γ_3	1	1	−1	−1	p_x, d_{yz}
Γ_4	1	−1	1	−1	p_y, d_{xz}

Wonderful Orthogonality Theorem (WOT)

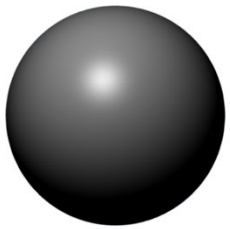
Ferdinand Frobenius

Issai Schur

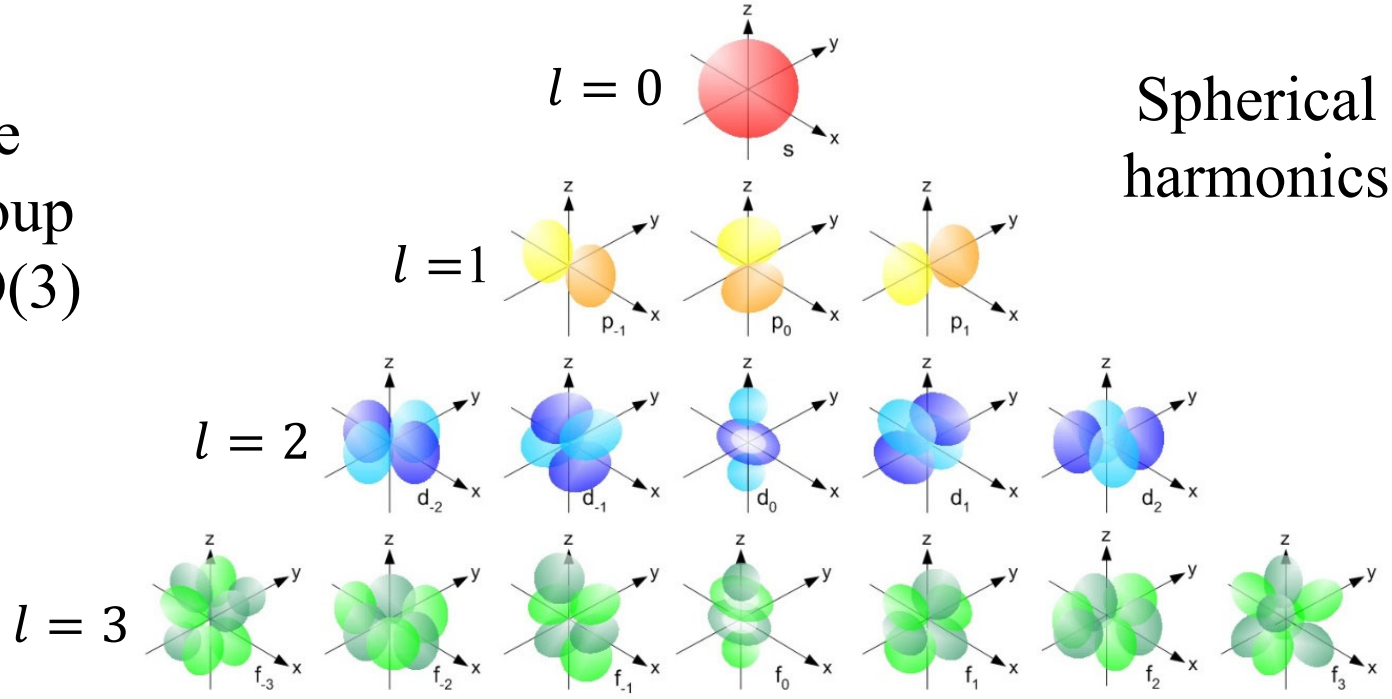
Frobenius discovered irreducible group representations in 1895-97. His student Schur discovered their orthogonality and completeness relations in 1904-07. They imply a natural symmetry-based coordinate system for parameterizing symmetry loss in any system. The new parameters are amplitudes of irrep basis functions, also called basis vectors, symmetry-adapted modes, or just "symmetry modes".

Irrep basis functions

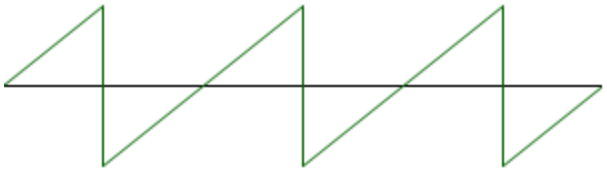
Irreps of the symmetry group of a sphere: O(3)

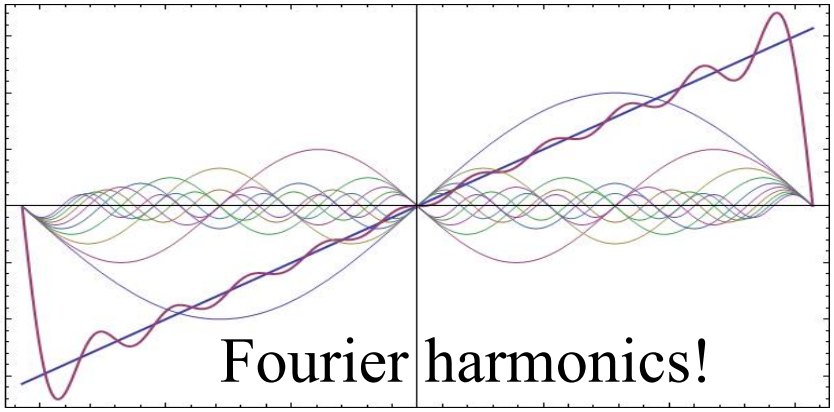


Spherical harmonics

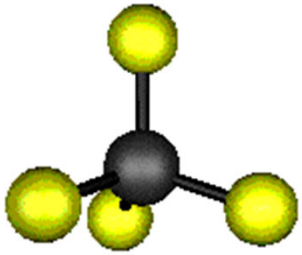


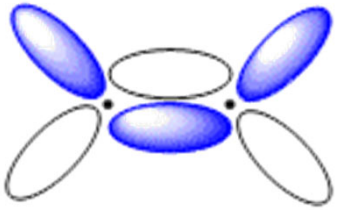
Irreps of the translational group of a periodic signal

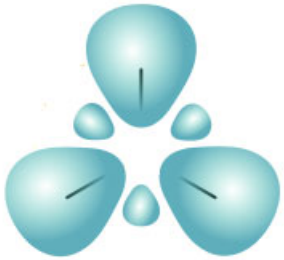


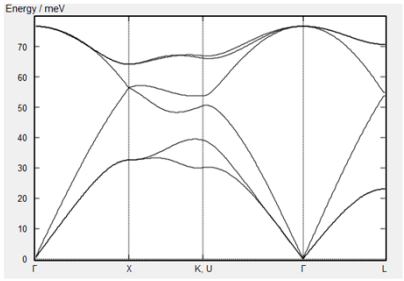


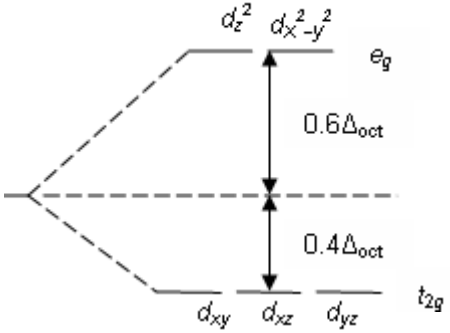
Applications of the irrep basis



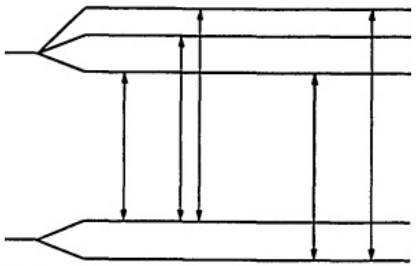


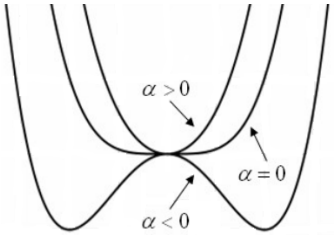




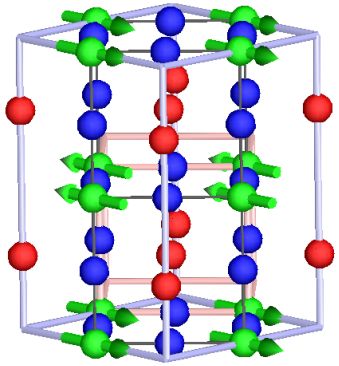


Molecular vibrations
Hybridized and molecular orbitals
Crystal-field splitting
Electronic-transition selection rules
Crystal band structure
Landau theory
General order parameters in crystals!

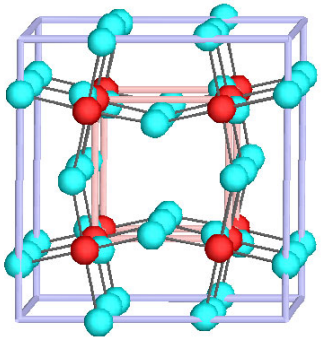




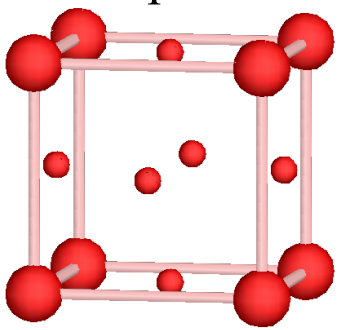
magnetic



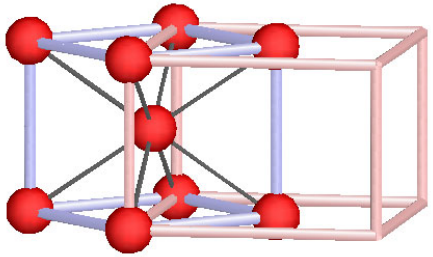
displacive



occupational

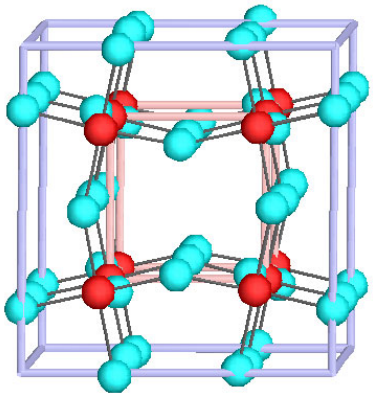


lattice strain

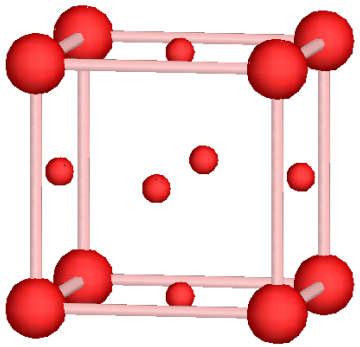


Application to materials

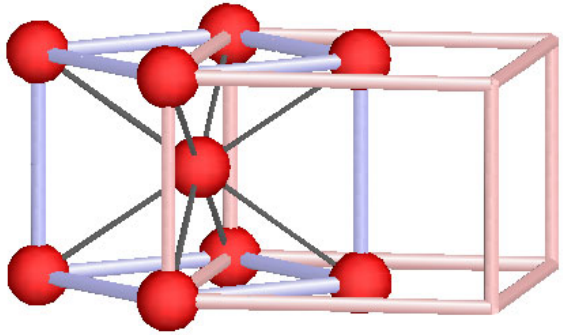
displacive



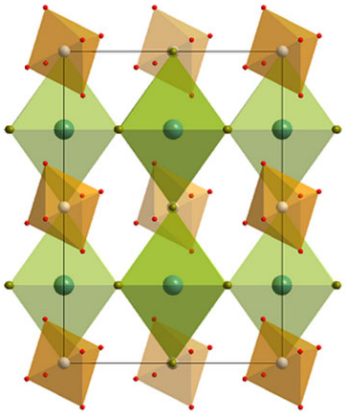
occupational



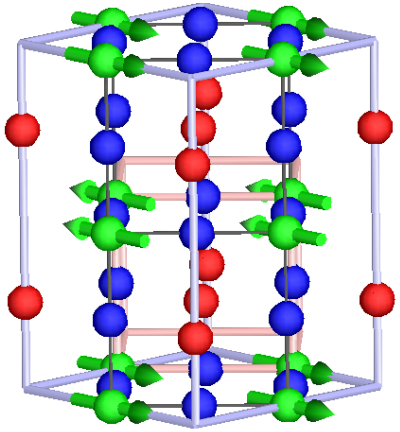
lattice strain



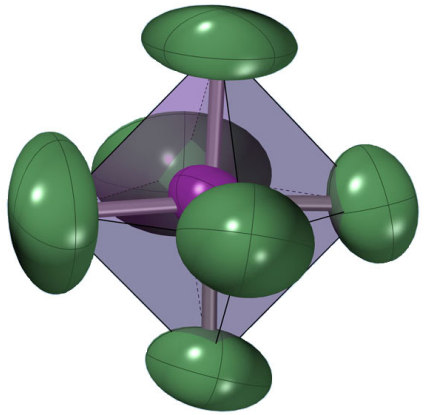
rotational



magnetic

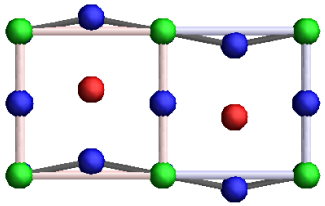


ADPs

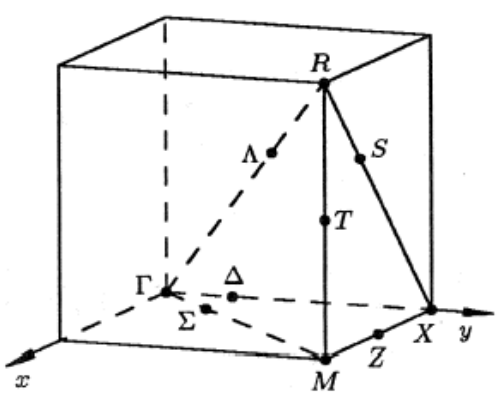


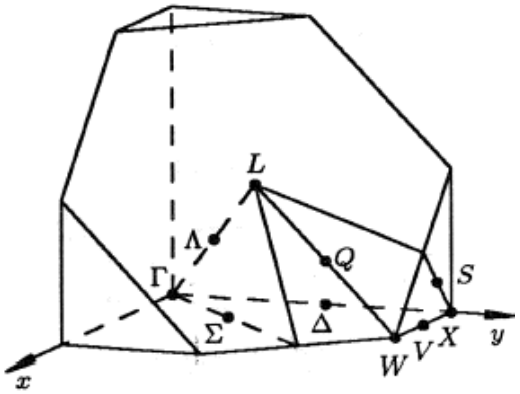
In crystals, symmetry modes provides an orthogonal and complete basis for describing the physical *order parameters* that arise in phase transitions. Each mode yields a *pattern* of changes that breaks the parent symmetry in a unique way.

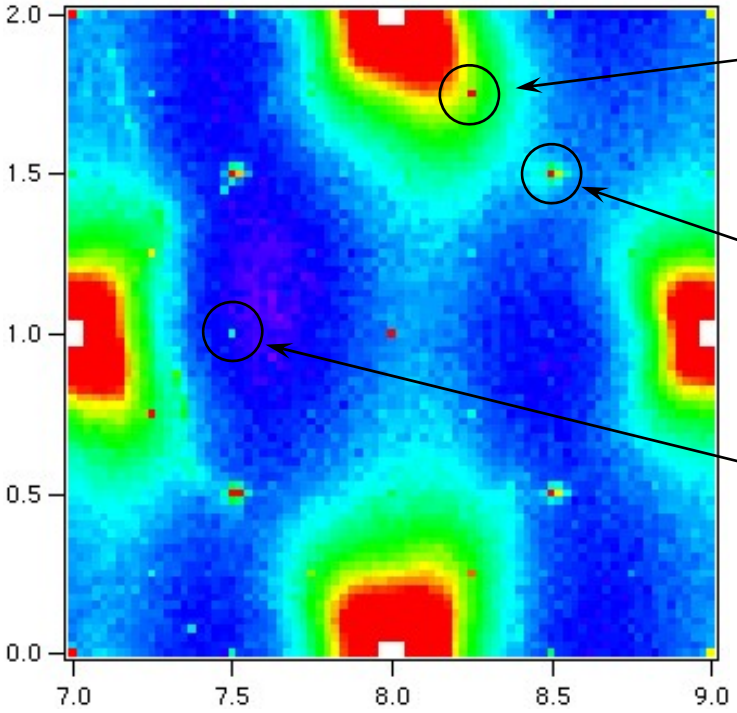
Crystal irreps defined at specific *k*-points



→ (½,0,0)





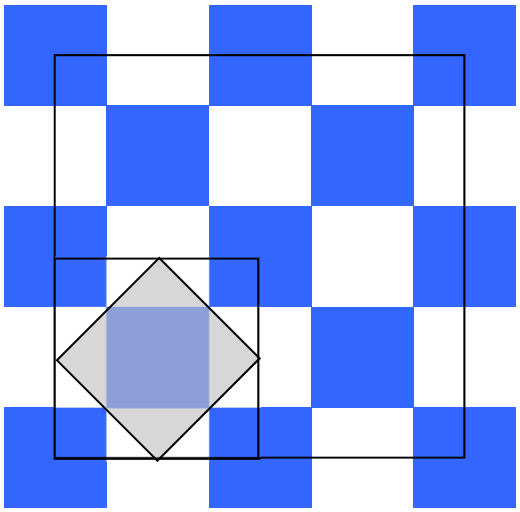


(¼,¼,0)

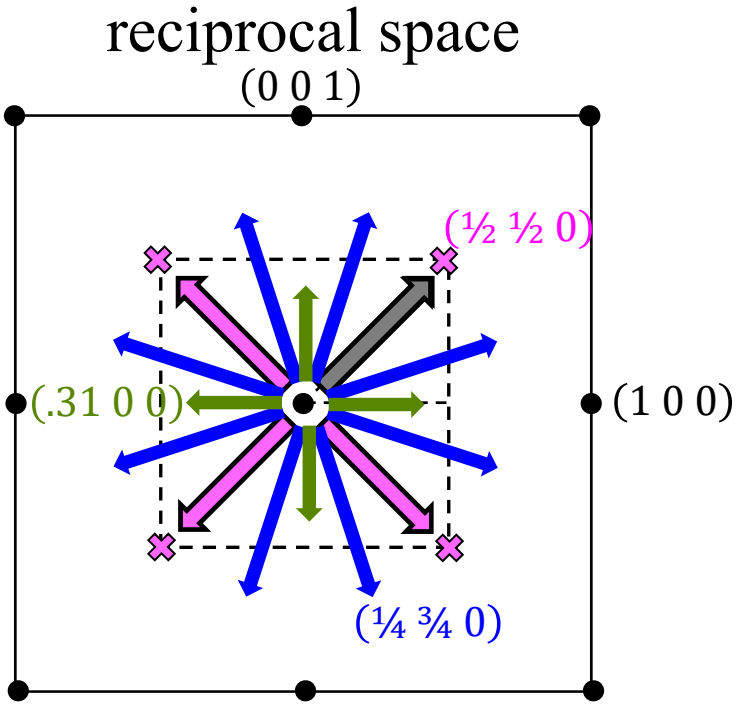
(½,½,0)

(½,0,0)

$2\sqrt{2} \times 2\sqrt{2}$



Space-group irrep calculations



Little-*k* group irreps

Faddeyev; Kovalev; Zak, Casher, Glück & Gur; Bradley, Cracknell, Davies, Miller, Love (1964-1979)

Complete space-group irreps at special-*k* points

Simultaneous action of entire ***k*** star.
8 cases worked *manually* (1968-1984).

Tables of Stokes & Hatch (1984, 1987): all 4777 space groups irreps at special ***k***; 15239 isotropy subgroups [green book].

Complete space-group irreps (any commensurate or incommensurate *k*)

ISO-IR, Stokes & Campbell (2014)
Tabulated – not real-time!

Complete space-group irreps at any commensurate *k* point

Karep (1992), ISOTROPY (1998)
real-time calculations

$$e^{2\pi i \boldsymbol{k}\cdot\boldsymbol{x}}$$

Irreps to (3+*d*)D superspace

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Tabulation of irreducible representations of the crystallographic space groups and their superspace extensions

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- New irreps matrices for all space groups at commensurate $\boldsymbol{k}$ vectors. Similar to earlier matrices ($g' = AgA^{-1}$ for $g \in G$), modern form.
- Separated form makes tabulation possible for the first time!
- Irrep matrices tabulated for superspace extensions of all space groups for the first time. Their isotropy subgroups are superspace groups.

Selecting an isotropy subgroup:

parent space-group symmetry

↓

k-point

finite number of types (e.g. Γ, Δ, X), but ∞ number of points

↓

irrep

finite number for each k-point (e.g. X_1^+, X_3^+, X_5^-)

↓

order-parameter direction (OPD)

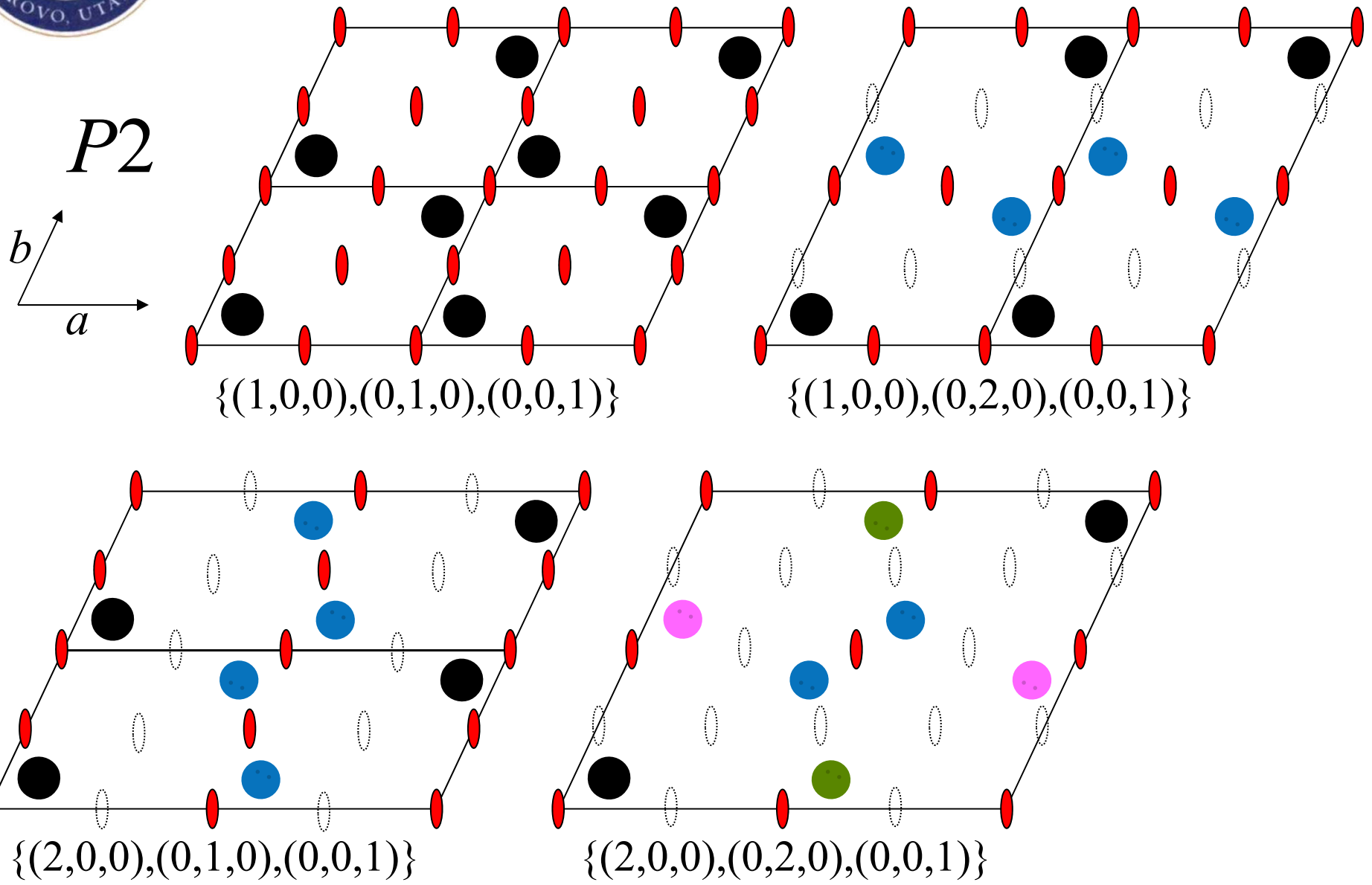
finite number for each IR; special points/lines/planes in abstract representations space

↓

isotropy subgroup

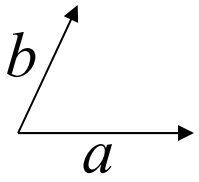
[1] space-group type (1 to 230)
[2] supercell basis (relative to parent)
[3] origin of supercell (relative to parent).

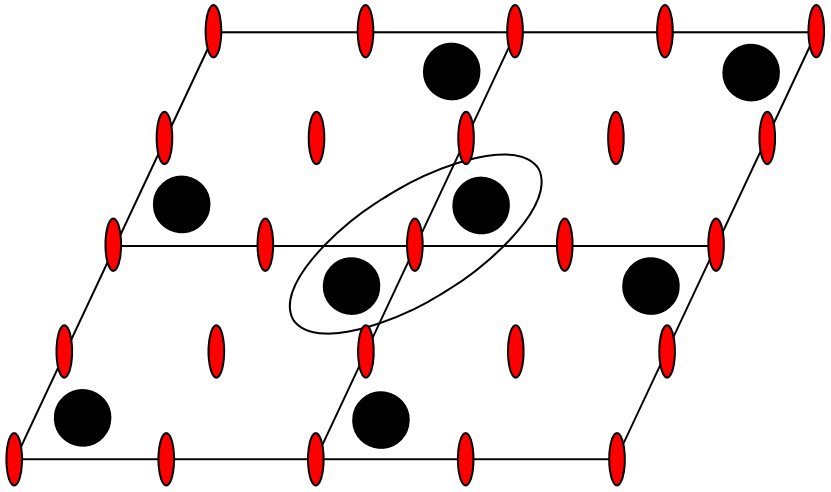
Distortion: (basis, SG-type)



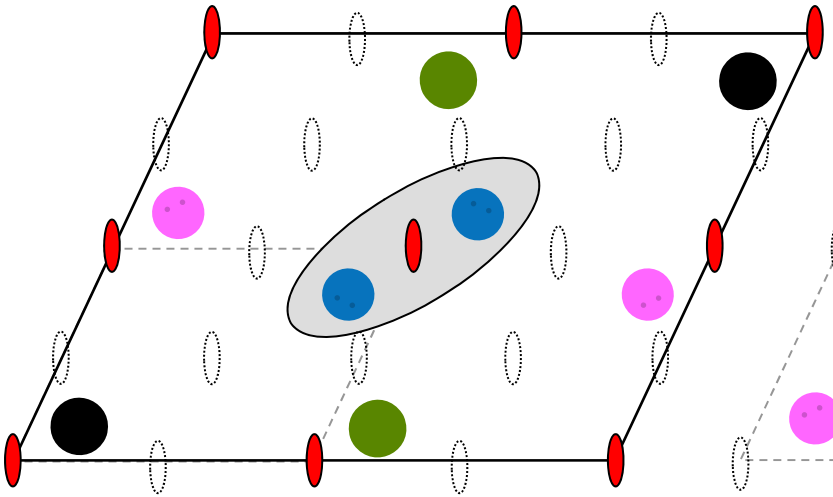
Distortion: (basis, origin, SG-type)

P2

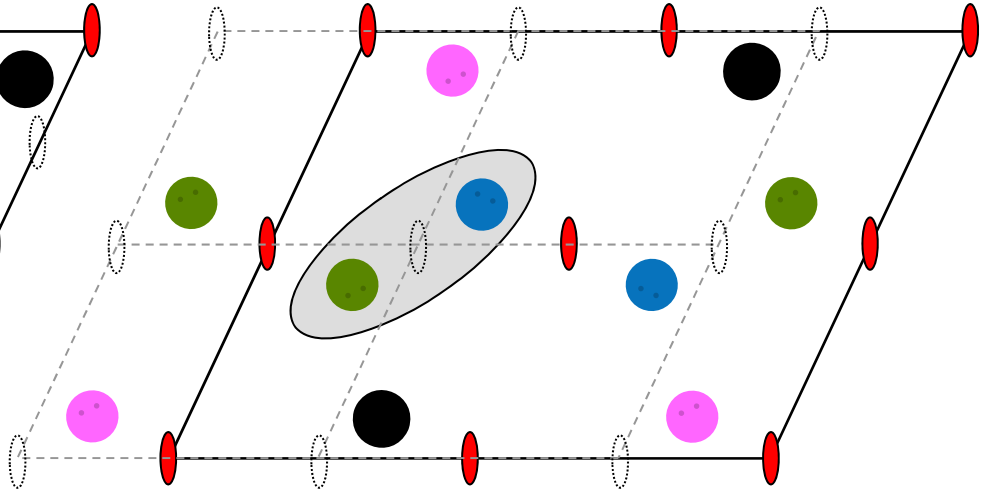




Two distinct cases with same basis = {(2,0,0),(0,2,0),(0,0,1)}



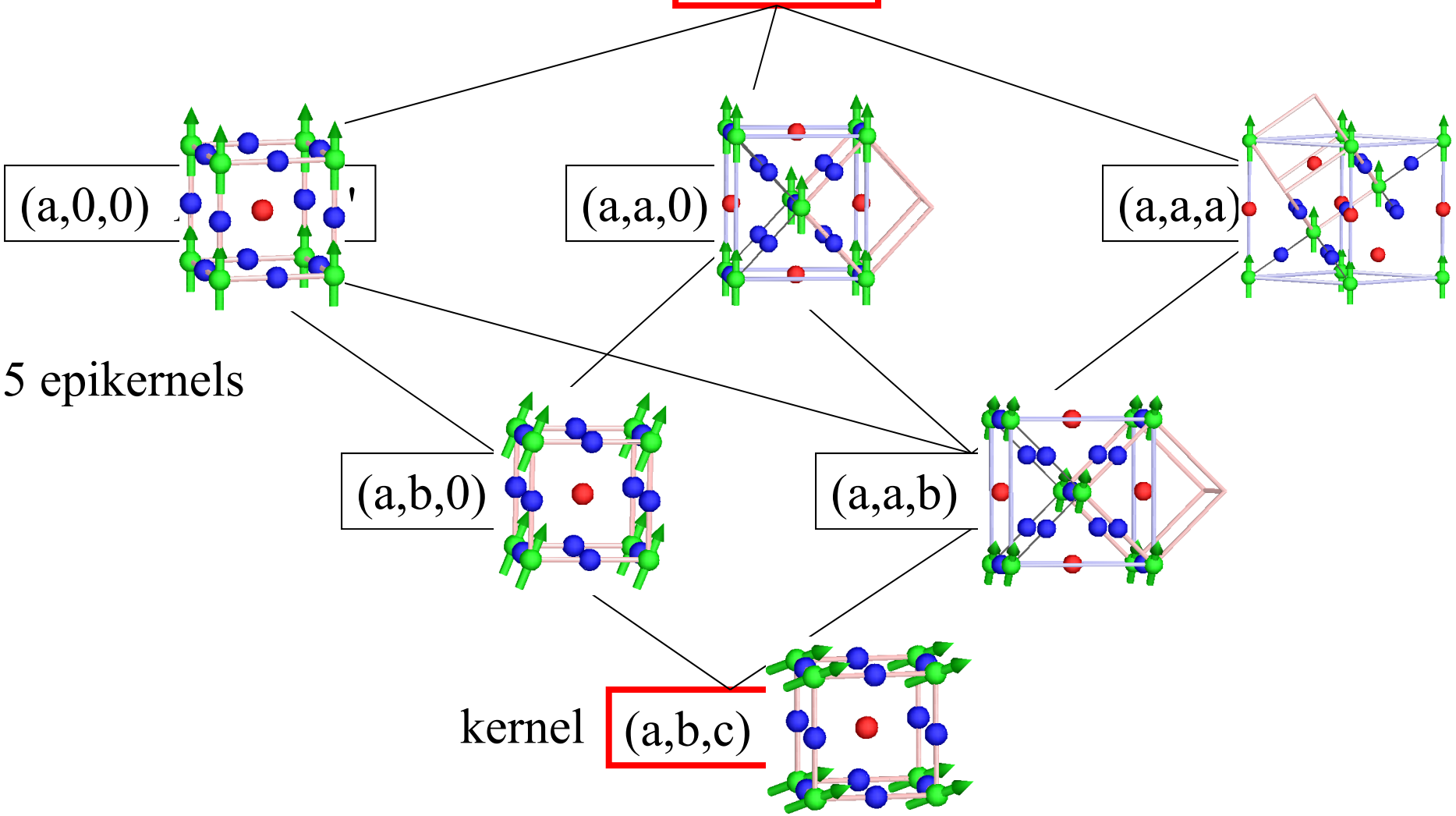
origin: (0,0,0)



origin: (½,0,0)

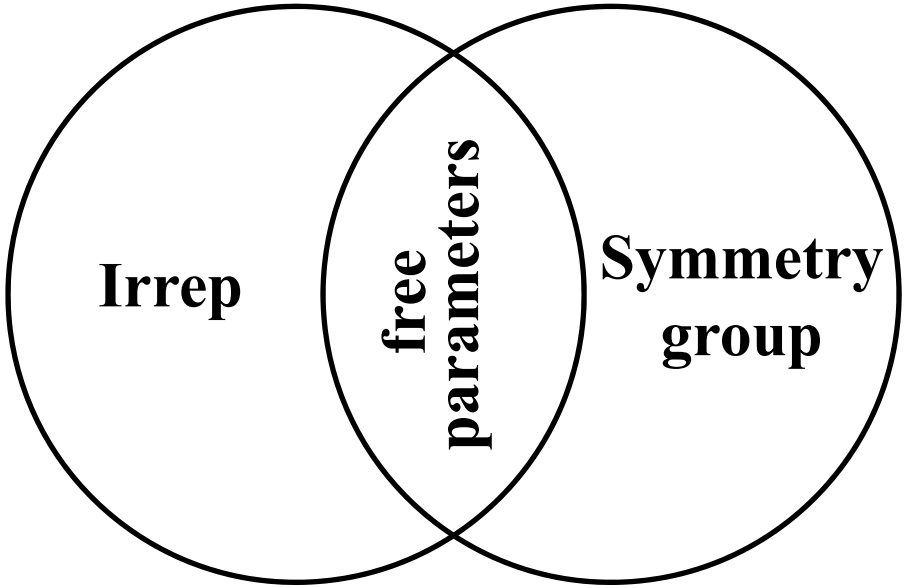
Irreps vs symmetry groups

3-dimensional irrep mΓ_4^+ irrep of $Pm\bar{3}m1'$ has 6 OPDs.



Irreps vs symmetry groups

Symmetry groups and irreps basis functions provide complementary constraints! Neither approach negates or weakens the other. They're fully compatible and actually work best together.



Especially in the area of magnetic-structure analysis, the practical use of irrep basis functions developed first, though the symmetry group infrastructure eventually caught up. One can now refine irrep basis functions that obey specific symmetry-group constraints.

Incommensurate-structure descriptions

Supercell description (approximate commensurate cell)

List moment vectors of atoms in the asymmetric unit of the MSG of the superstructure.

Wave description (incommensurate)

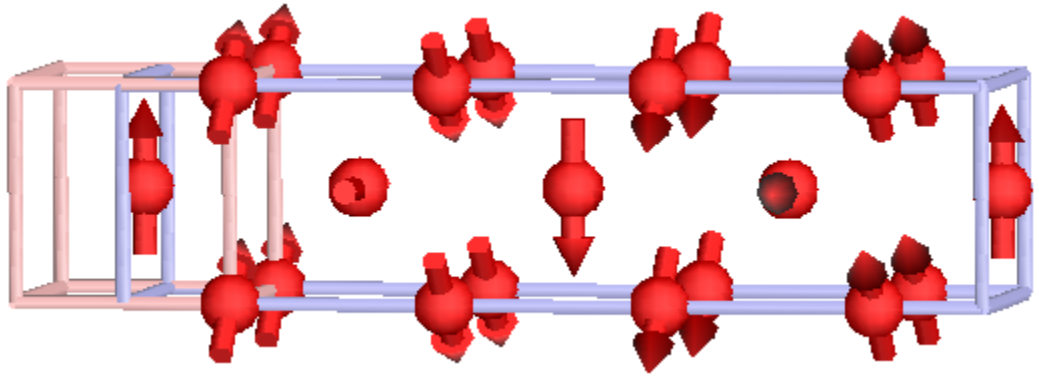
List complex Fourier amplitude vectors (sine and cosine) of atoms in the asymmetric unit of the MSG of the basic unit cell at all satellites of non-negligible intensity in the reciprocal parent cell.

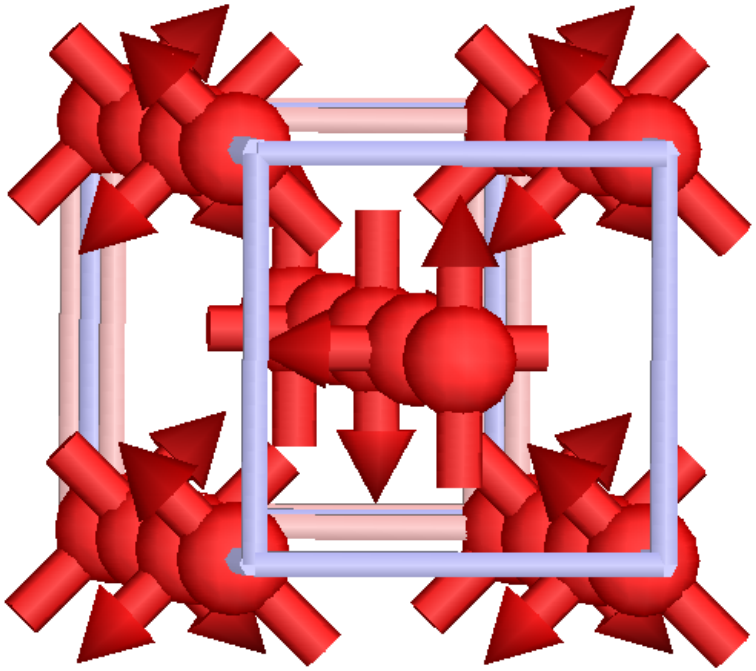
Irrep description (commensurate or incommensurate)

List the amplitudes of the irrep symmetry modes consistent with the MSG or MSSG. Include secondary irreps, multiple instances of same irrep, and all components of multi-dimensional order params.

If presented with sufficient generality, all three descriptions have the same number of free parameters, whether or not symmetry is applied.

4-cell magnetic helix (bcc)





4-cell magnetic helix (bcc)

Commensurate: 95.140 P_c4_322

#	x'	m'	θ	#	x'	m'	θ
1	x,y,z	mx,my,mz	+1	9	x,y,z+1/2	-mx,-my,-mz	-1
2	x,-y,-z+1/2	mx,-my,-mz	+1	10	x,-y,-z	-mx,my,mz	-1
3	-x,y,-z	-mx,my,-mz	+1	11	-x,y,-z+1/2	mx,-my,mz	-1
4	-x,-y,z+1/2	-mx,-my,mz	+1	12	-x,-y,z	mx,my,-mz	-1
5	-y,-x,-z+3/4	-my,-mx,-mz	+1	13	-y,-x,-z+1/4	my,mx,mz	-1
6	-y,x,z+3/4	-my,mx,mz	+1	14	-y,x,z+1/4	my,-mx,-mz	-1
7	y,-x,z+1/4	my,-mx,mz	+1	15	y,-x,z+3/4	-my,mx,-mz	-1
8	y,x,-z+1/4	my,mx,-mz	+1	16	y,x,-z+3/4	-my,-mx,mz	-1

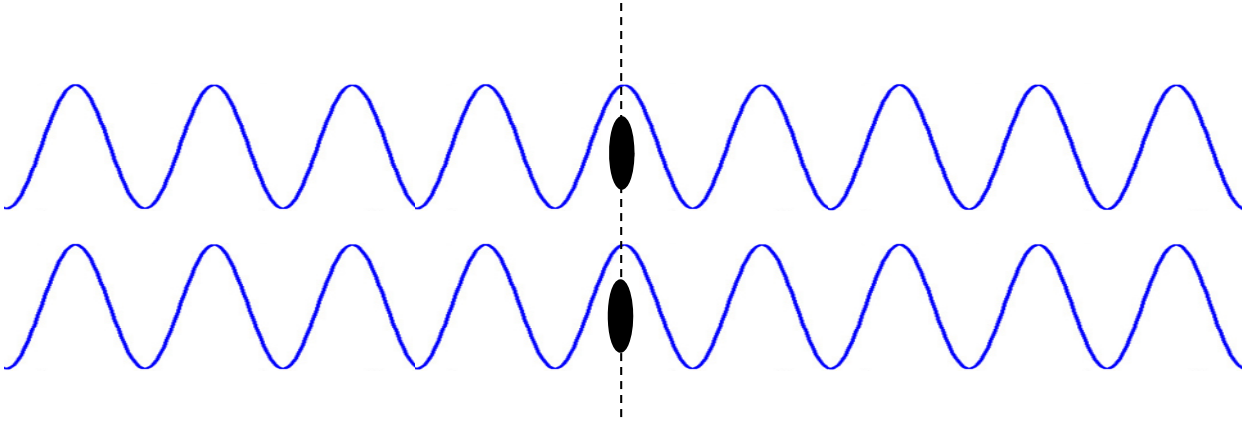
Incommensurate: 97.1.21.2.m152.2 $I4221'(0,0,g)q00s$

#	x'	m'	θ	#	x'	m'	θ
1	x,y,z	mx,my,mz	+1	9	x,y,z+1/2	-mx,-my,-mz	-1
2	x,-y,-z+1/2	mx,-my,-mz	+1	10	x,-y,-z	-mx,my,mz	-1
3	-x,y,-z	-mx,my,-mz	+1	11	-x,y,-z+1/2	mx,-my,mz	-1
4	-x,-y,z+1/2	-mx,-my,mz	+1	12	-x,-y,z	mx,my,-mz	-1
5	-y,-x,-z+3/4	-my,-mx,-mz	+1	13	-y,-x,-z+1/4	my,mx,mz	-1
6	-y,x,z+3/4	-my,mx,mz	+1	14	-y,x,z+1/4	my,-mx,-mz	-1
7	y,-x,z+1/4	my,-mx,mz	+1	15	y,-x,z+3/4	-my,mx,-mz	-1
8	y,x,-z+1/4	my,mx,-mz	+1	16	y,x,-z+3/4	-my,-mx,mz	-1

Traditional vs symmetry-mode parameters

- Symmetry-modes span the same configurational space as traditional coordinates if all relevant *k*-points, irreps, and OPD components are considered simultaneously. Number of free variables is conserved!
- The relationship between traditional and symmetry-mode coordinate systems is linear! Related by an invertible square numerical matrix. One mode can affect many symmetry-distinct atoms, and one atom can be affected by many modes.
- Have you ever constrained a model by manually constructing a linear combination of traditional parameters and fixing them to zero? Symmetry modes can be viewed as linear constraints that you can either refine or fix (at zero).
- Symmetry modes are waves of a specific ***k*** vector. Irreps are defined separately for every point in the first Brillouin zone. Two wave vectors separated by a reciprocal-lattice vector have the same irreps.
- Symmetry modes very often provide the most natural/efficient basis. Nature tends to activate as few symmetry modes as possible. Even complicated magnetic structures are usually described by a single irrep!

Superspace symmetry operations



Space group elements transform atoms into other atoms. Superspace group elements do this too, but must also transform the incommensurate waves attached to those atoms. Regular space group operations can't do that.

Symmetry operations can permute the propagation vectors of the star of k, and hence relate the complex vector amplitudes of the waves to one another. They change **both** the **direction** and **phase** of such a complex vector.

The complex amplitude of a magnetic wave transforms like a magnetic axial vector (includes factors for $\det(R)$ and time reversal θ).

Tabulating superspace groups

222	I	C_{2x}	C_{2y}	C_{2z}
I	I	C_{2x}	C_{2y}	C_{2z}
C_{2x}	C_{2x}	I	C_{2z}	C_{2y}
C_{2y}	C_{2y}	C_{2z}	I	C_{2x}
C_{2z}	C_{2z}	C_{2y}	C_{2x}	I

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

- Start with 3D space group (R, v)
- Establish the k-vector options, based on (3+d)D Bravais classes.
- For given k; calculate M and ϵ.
- Only phases (δ) of generators are unknown. Build matrices for the whole group in terms of them.
- Build multiplication table, with self-consistent phase requirement.
- System of modular equations, can be solved using Smith-normal form.
- Each solution is a candidate. Test equivalence to isolate unique results.

Testing SSG equivalence

I have two sets of operators (G and G') that might be settings of the same SSG. Find a transformation matrix S that simultaneously transforms every $g \in G$ to a corresponding element $g' \in G'$.

$$g = \left(\begin{array}{cc|c} R & 0 & v \\ M & \epsilon & \delta \\ \hline 0 & 0 & 1 \end{array}\right) \qquad S = \left(\begin{array}{cc|c} S_R & 0 & S_v \\ S_M & S_\epsilon & S_\delta \\ \hline 0 & 0 & 1 \end{array}\right) \qquad g' = S\, g\, S^{-1} = \left(\begin{array}{cc|c} R' & 0 & v' \\ M' & \epsilon' & \delta' \\ \hline 0 & 0 & 1 \end{array}\right)$$

$$R' = S_R\, R\, S_R^{-1}$$

$$\epsilon' = S_\epsilon\, \epsilon\, S_\epsilon^{-1}$$

$$M' = S_M\, R \;-\; S_\epsilon\, \epsilon\, S_\epsilon^{-1}\, S_M\, S_R^{-1} \;+\; S_\epsilon\, M$$

$$v' = -\, S_R\, R\, S_R^{-1}\, S_v \;+\; S_R\, v \;+\; S_v$$

$$\delta' = -\, S_M\, R\, S_v \;+\; S_\epsilon\, \epsilon\, (S_\epsilon^{-1}\, S_M\, S_R^{-1}\, S_v - S_\delta) \;-\; S_\epsilon\, M\, S_v \;+\; S_M\, v \;+\; S_\epsilon\, \delta \;+\; S_\delta$$

Tabulating superspace groups

Builds on work of Orlov, Chapuis, and Yamamoto

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

research papers

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

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Equivalence of superspace groups

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Infrastructure for $d = 3$ superspace

Superspace groups

775 for $d = 1$

3338 for $d = 2$

12584 for $d = 3$

Over 16,000 for $d = 1,2,3$

Magnetic superspace groups (NEW)

Almost 350,000 for $d = 1,2,3$

Equivalence testing against our database allows us to determine whether or not two structures with very different presentations are equivalent.

Symmetry modes in (3+1)D superspace

Acta Cryst. A**6**, 365-373 (2007)

research papers

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Order parameters for phase transitions to structures with one-dimensional incommensurate modulations

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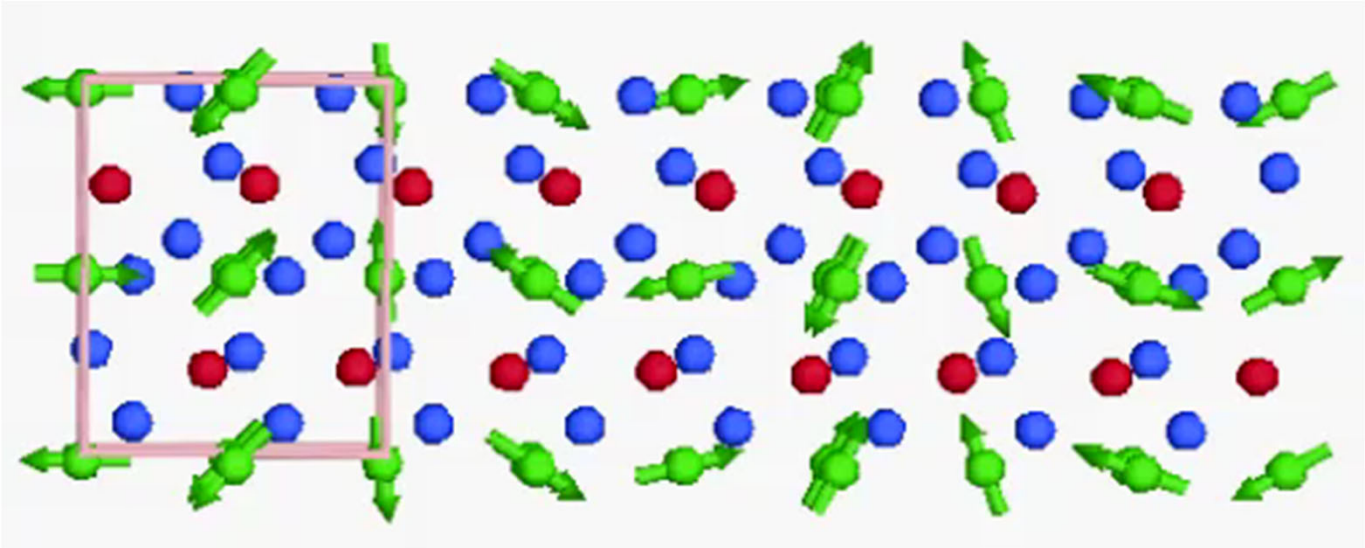
- Irreps and isotropy subgroups tabulated at $d = 1$ incomm $\boldsymbol{k}$ vectors.
- Incomm displacive and occupancy modes in ISODISTORT (2007).
- Superposition of commens and $d = 1$ incomm modes (2010).
- Incommensurate magnetic modes (2011).
- Superposition of commens and $d = 1,2,3$ incomm modes (2014).
- Incommensurate rotational modes in (2016).

Multi-ferroic $TbMnO_3$



Incommensurate cycloidal magnetic structure. Transverse $m\Sigma_2$ and longitudinal $m\Sigma_3$ order parameters are superposed 90° out of phase.

This irrep combination couples to the secondary ferroelectric Γ_4^+ irrep, making it a multi-ferroic material.

Must consider full space-group irreps (whole *k*-star, $\pm k$) to get the symmetry and physical properties right! This would have avoided a significant controversy in the literature that spanned many years.

Magnetic skyrmion lattice

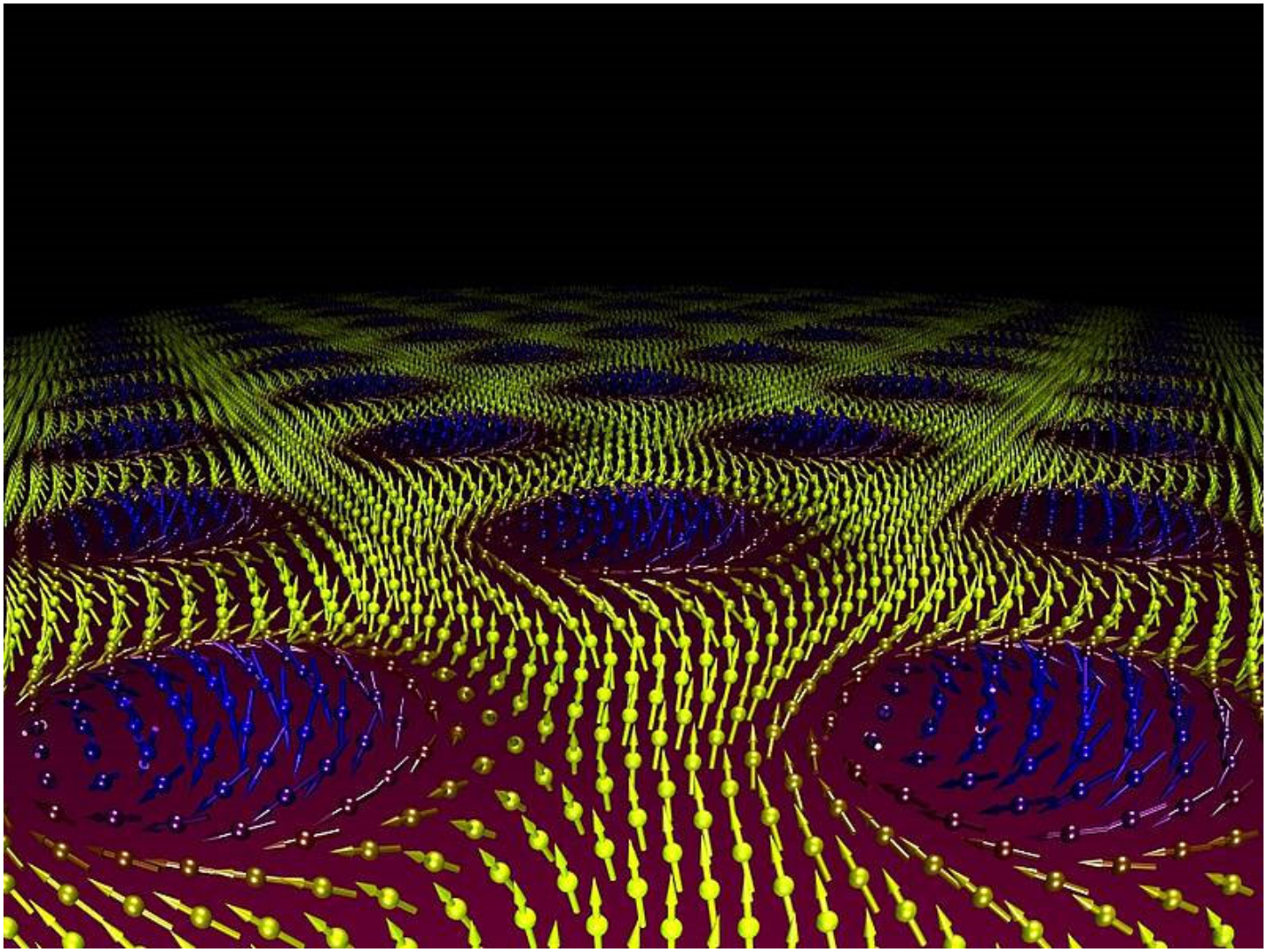
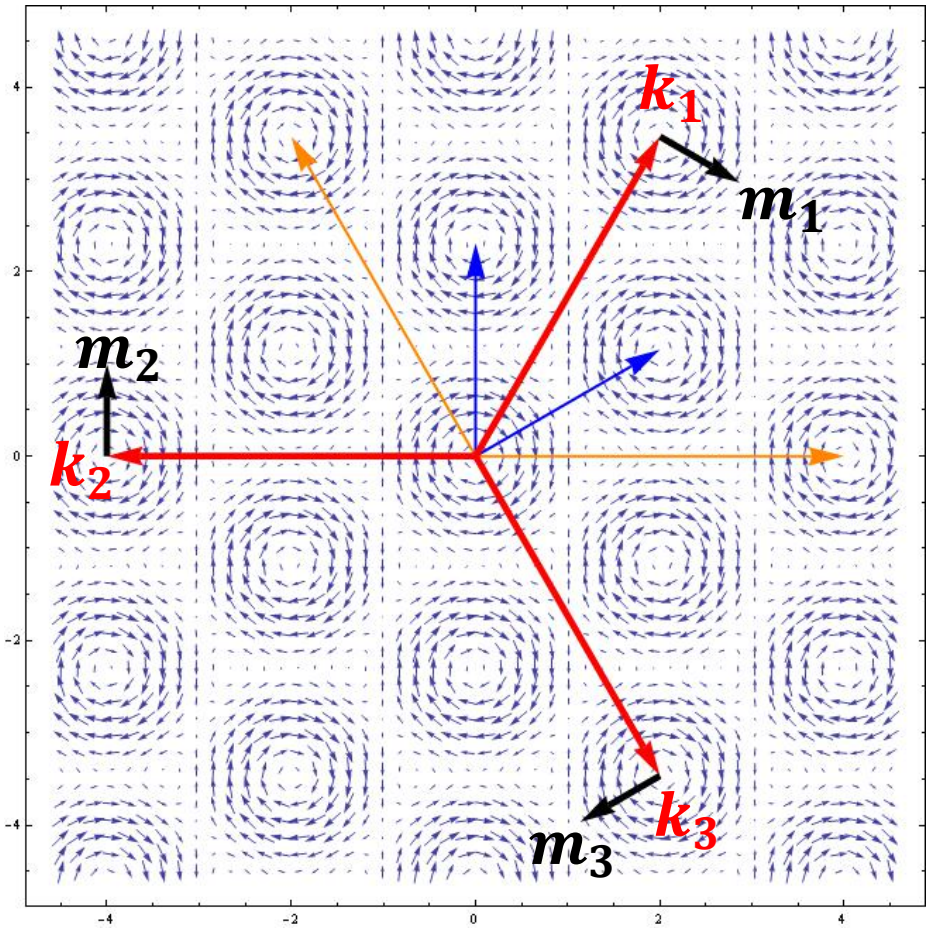


Image from Mühlbauer et al., Science 323, 915-919 (2009).

2D skyrmion-like lattice



Three incommensurate transverse waves, locked in phase, form this remarkable pattern. It has only one adjustable magnetic degree of freedom.

It is readily described in terms of either symmetry-related Fourier wave amplitudes or an irrep basis function.

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

$$m_1 = i(m, -m, 0),\ m_2 = i(m, 2m, 0),\ m_3 = i(-2m, -m, 0)$$

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