Why use symmetry?

- Electronic properties and structures are complex
  - Magnetism is the spin-dependent part
- Never as much information about them as we would like
  - Experimentalists typically deal with under-defined problems (there are too many possible solutions)
  - Symmetry is useful to introduce a grand simplifying structure (makes rules - followed and broken, classes of behaviour, and thus to simplify, clarify and reveal…)

An Introduction to Magnetic Structures
- Working with the propagation vector
  (Crystalline Solids)

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Overview of today

• Why do we need to invoke symmetry?

• Taking symmetry theory from point groups to magnetic structures
  – Translational periodicity
    • Increases complexity of the irreducible representations
    • Rotation-translation operations
    • The propagation vector, the k-vector

• A more sophisticated language
  – The little group of the propagation vector \( G_k \)
  – Permutation representation
  – Axial and polar vectors, representations
  – Magnetic representation
  – Basis vectors
  – Couplings - Time reversal and Landau theory

• Symmetries and frameworks
  – Representations and irreducible representations
  – Magnetic space groups (time reversal)

➡ Gives the language for understanding magnetic structures, for posing questions
Overview of this lecture
- Working with the propagation vector

- Using the symmetry language
  - Away from a shaken box → Frameworks and information
  - Reveal what magnetic structures are
  - Building up descriptions to make the range of possible magnetic structures
  - Pulling together the different symmetry ideas together within representation theory

Why should an experimentalist use symmetry?
- Never enough information...

- Magnetic structures are complex
  - Information is destroyed in many ways
  - The magnetic form factor: \( J(Q) \)
  - The magnetic structure factor: \( F_{\text{M}\perp}(Q) \)
  - Powder averaging
  - Domain averaging (powder, single crystal)
Some simple magnetic structures

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• Domain averaging (powder, single crystal)
Complex incommensurate magnetic ordering in B-Mn$_{1-x}$Ru$_x$ ($x=0.12$)

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Why should an experimentalist use symmetry?  
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  ➔ Under-defined problem  
  ➔ Hidden (unconsidered) possibilities

Definition of magnetic structures, phonons, electronic orbitals

  - A linear combination of plane waves (basis vectors, Fourier components)
  - Bloch waves - Eigenfunctions of a periodic Hamiltonian can be constructed from Fourier components

\[
\begin{align*}
\psi^k_j \cdot \nu & = \psi^k_i \cdot \nu e^{-2\pi i \mathbf{k} \cdot \mathbf{r}_{ij}} \\
\mathbf{m}_{ij} & = \sum_{\nu; \mathbf{k}} C_{\nu; \mathbf{k}} \psi^k_i \cdot \nu e^{-2\pi i \mathbf{k} \cdot \mathbf{r}_{ij}}
\end{align*}
\]

⇒ Once the moments in the primitive unit cell are defined, the $\mathbf{k}$ vector defines every other spin in the structure
The propagation vector

\[ \psi_{j,\nu} = \psi_{i,\nu} e^{-2\pi i \vec{k} \cdot \vec{t}_{ij}} \]

with \( \vec{k} = \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} \)

\[ \begin{align*}
\psi_{j,\nu} &= \psi_{i,\nu} \exp \left[ -2\pi i \begin{pmatrix} 0 \\ 0.5 \\ 0 \end{pmatrix} \cdot \begin{pmatrix} 0 \\ 0 \\ 3 \end{pmatrix} \right] = -\psi_{i,\nu} \\
\psi_{j,\nu} &= \psi_{i,\nu} \exp \left[ -2\pi i \begin{pmatrix} 0 \\ 0 \\ 0.5 \\ 0 \\ 0 \\ 2 \end{pmatrix} \right] = \psi_{i,\nu} \\
\psi_{j,\nu} &= \psi_{i,\nu} \exp \left[ -2\pi i \begin{pmatrix} 0 \\ 0 \\ 0.5 \\ 0 \\ 0 \\ 1 \end{pmatrix} \right] = \psi_{i,\nu} \exp [-\pi i] = -\psi_{i,\nu} \\
\psi_{j,\nu} &= \psi_{i,\nu} \exp \left[ -2\pi i \begin{pmatrix} 0 \\ 0 \\ 0.5 \\ 0 \\ 0 \\ 0 \end{pmatrix} \right] = \psi_{i,\nu} \exp [-2\pi \theta] = \psi_{i,\nu}
\end{align*} \]

The formalism of the propagation vector, \( \vec{k} \)

\[ \tilde{m}_j = \sum_{\nu, \vec{k}} C_{\nu \vec{k}} \psi_{i,\nu} e^{-2\pi i \vec{k} \cdot \vec{t}_{ij}} \]

- I moment in the asymmetric unit (the primitive unit cell)
- Once \( \vec{k} \) is defined, total degrees of freedom = 3
What a magnetic structure (Néel state) is

- An ordered configuration of magnetic moments with a long correlation length
  - The order has some translational symmetry (the moments in different unit cells - related by primitive lattice vectors - are related)
- The orientations of the moments are related by symmetry (what happens in detail depends on where the moments are in the system and the host crystal structure)

What a magnetic structure isn’t

- A haphazard set of arrows (moments) in a box (crystal structure)
  - This could fail to have the translational symmetry relating moments in different unit cells (careful with centred cells!!!)
  - Magnetic structures are pretty well misunderstood and papers giving nonsensical structures an frequent problem…
- There are rules…
  - But they are open ended…
What different types of structures are possible?

- Lots:
  - Simple ferromagnetic structures (identical moments that align parallel)
  - Simple antiferromagnetic and ferrimagnetic structures (neighbouring moments align antiparallel)
  - Complex antiferromagnetic structures
    - Commensurate
    - Incommensurate (sine waves or spin density waves, helices, etc)

- Open ended → Mixtures
Which moments are related by symmetry?

Magnetic structures

- **k-vector**:
  - Propagate (a component of) the magnetic structure through the crystal
  - Define translational periodicity and orientation dependence

- **basis vectors**
  - Build up symmetry within primitive unit cell of $G_0$

$$\vec{m}_j = \sum_{\nu,k} C_{\nu}^{\vec{k}} \psi_{\nu}^{\vec{k}} e^{-2\pi i \vec{k} \cdot \vec{t}_{ij}}$$
How can magnetic structures be described - Simple moments and unit cells?

- People like to think in terms of $m_x, m_y, m_z$ - Don’t!
- Begin with $m_x, m_y, m_z$ (the components along parallel to the crystal axes)
  - This description is intuitive
  - Best used to describe the final structure, not to refine it
  - Instead use functions that are symmetry adapted to the system you are dealing with, these will allow more complex symmetries

- People like to use unit cells (magnetic space groups)
  - This description is intuitive
  - The description of a magnetic structure within MSG framework is equivalent to using representation analysis - it has to be
  - Beauty is in the eye of the beholder:
    - Both MSGs and representation theory need to be treated with care.
    - Couplings are treated differently, elegance of describing a structure depends on what you want and your preferred point of reference.

How can magnetic structures be described -an alternative approach

- Origins
  - The eigenfunctions of an electrons with a periodic Hamiltonian are Bloch waves.
    - with the form: $\tilde{m}_j = \sum_{\nu, \vec{k}} C_{\nu}^{\vec{k}} \tilde{\psi}_{\vec{k}, \nu} e^{-2\pi i \vec{k} \cdot \vec{t}_j}$
  - Magnetic structures are eigenfunctions of the spin-dependent electronic Hamiltonian and have the same form
  - If we expand the exponential, we see that it is made up of a Real cosine part and an Imaginary sine part
    $$\tilde{m}_j = \sum_{\nu, \vec{k}} C_{\nu}^{\vec{k}} \tilde{\psi}_{\vec{k}, \nu} \left[ \cos(-2\pi \vec{k} \cdot \vec{t}_j) + i \sin(-2\pi \vec{k} \cdot \vec{t}_j) \right]$$
  - This formalism very general and we will see that it can describe simple and exotic structures, such as sinusoidal and helical structures
Basis vectors and $k$-vectors

- Simple structures and 'sine or cosine' structures
  - The translational properties of a magnetic structure may be described by
    \[
    \tilde{m}_j = \sum_{\nu,k} C^k_{\nu} \psi^e_{i,\nu} e^{-2\pi i \tilde{k} \cdot \tilde{t}_{ij}}
    \]
  - Working with only one basis vector, ignoring the coefficient for simplicity and expanding the exponential, this becomes
    \[
    \tilde{m}_j = \psi^e_{i,\nu} \left[ \cos(-2\pi \tilde{k} \cdot \tilde{t}_{ij}) + i \sin(-2\pi \tilde{k} \cdot \tilde{t}_{ij}) \right]
    \]
  - If $\psi$ is real and the propagation vector is such that the sine part is zero, e.g., components 0 and 1/2
    - Left with a simple cosine curve with the moments of the same amplitude.

A simple (cosine) structure
Basis vectors and k-vectors

- \( \Psi \) is real and \( k \) is such that the sine component is non-zero
  - Leads to \( m \) being complex, so need to make it real
  - The moment vector for an atom in the \( n \)th cell related to that in the zeroth cell by translation \( t \) is given by
    \[
    m_j = C_k \psi_{i,\nu}^k e^{-2\pi ik \cdot t_{ij}} + C_k \psi_{i,\nu}^{-k} e^{-2\pi i(-k) \cdot t_{ij}} \\
    m_j = C_k \psi_{i,\nu}^k e^{-2\pi ik \cdot t_{ij}} + C_k (\psi_{i,\nu}^k)^* e^{-2\pi i(-k) \cdot t_{ij}}
    \]
  - As
    \[
    \psi_{i,\nu}^{-k} = \psi_{i,\nu}^{*k}
    \]
  - Substitution and expansion of the exponential leads to
    \[
    \tilde{m}_j = 2Re(\psi_{i,\nu}^k) \left[ \cos(-2\pi k \cdot t_{ij}) \right] + 2Im(\psi_{i,\nu}^k) \left[ \sin(-2\pi k \cdot t_{ij}) \right]
    \]
  - Where the second term is zero as \( \Psi \) is real \( \rightarrow \) Amplitude modulated sine structure (spin density wave)
Basis vectors and k-vectors

- \( \Psi \) is complex and \( k \) is incommensurate
  - Leads to \( \mathbf{m} \) being complex, so need to make real moments
  - The atomic vector for an atom in the \( n \)th cell related to that in the zeroth cell by translation \( \mathbf{t} \) is given by
    \[
    \mathbf{m}_j = C^k_{\nu} \psi^k_{i,\nu} e^{-2\pi ik \cdot t_{ij}} + C^k_{\nu} (\psi^k_{i,\nu})^* e^{-2\pi i(-k) \cdot t_{ij}}
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    \bar{m}_j = 2Re(\psi^k_{i,\nu}) \left[ \cos(-2\pi \mathbf{k} \cdot \mathbf{t}_{ij}) \right] + 2Im(\psi^k_{i,\nu}) \left[ \sin(-2\pi \mathbf{k} \cdot \mathbf{t}_{ij}) \right]
    \]
  - If the real and imaginary parts are not parallel → circular or elliptical helix

- If the real and imaginary parts are not parallel → circular or elliptical helix
Building structures: basis vectors and $k$-vectors

- A circular helix
  - $k$ incommensurate (or sine part is non-zero)
  - $\Psi$ is complex, and has non-collinear real and imaginary components (equal magnitude) in a plane that does not contain $k$

- A conical structure
  - $k$ incommensurate (or sine part is non-zero)
  - $\Psi$ is complex, and has non-collinear real and imaginary components (equal magnitude) in a plane that does not contain $k$
  - $k=(000)$
    - $\Psi$ is ferromagnetic and is perpendicular to the helix

- A cycloid
  - $k$ incommensurate (or sine part is non-zero)
  - $\Psi$ is complex, and has non-collinear real and imaginary components (equal magnitude) in a plane that contains $k$

Practice making some magnetic structures
Work with the Brillouin zone and types of $k$

e.g. FCC

\[ \mathbf{k}' = \mathbf{k}h \pm \mathbf{\tau} \]

- The symmetry types of the different points in reciprocal space
- Different points, lines and planes have different compatible symmetry operations; different $G_k$
- (Care with axis system)
- Several notations exist, Kovalev, Miller and Love, etc

But what about unseen complexity?

- Types of domain (characterised by the types of symmetry elements lost during the magnetic ordering)

<table>
<thead>
<tr>
<th>Configurational (k) domains</th>
<th>(translational symmetry)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pi domains</td>
<td>(time reversal)</td>
</tr>
<tr>
<td>Orientational (S) domains</td>
<td>(rotational symmetry)</td>
</tr>
<tr>
<td>Chiral domains</td>
<td>Centrosymmetry</td>
</tr>
</tbody>
</table>
Configurational domains (k domains)

- Arise if $G_k <> G_0$
- Operating with the paramagnetic symmetry elements on $k$
  generates a set of inequivalent vectors which form the star of $k$,
e.g. $k_1 = k_i E$, $k_2 = k_i R_2$, $k_3 = k_i R_3$, $k_4 = k_i R_4$.
- e.g. FCC lattice

\[
\begin{align*}
\vec{k}_1 &= \left( \frac{1}{2}, \frac{1}{2}, \frac{1}{2} \right) \\
\vec{k}_2 &= \left( -\frac{1}{2}, \frac{1}{2}, \frac{1}{2} \right) \\
\vec{k}_3 &= \left( \frac{1}{2}, -\frac{1}{2}, \frac{1}{2} \right) \\
\vec{k}_4 &= \left( \frac{1}{2}, \frac{1}{2}, -\frac{1}{2} \right)
\end{align*}
\]

- Each vector in the star generates a different (equivalent) configuration domain
- Each configuration domain gives a completely separate set of magnetic reflections at positions $\pm k$ from the reciprocal lattice nodes
- Each set of reflections belongs to a distinct region of the crystal, hence effectively to a single state
**π- domains (time reversal)**
- Regions in which all the moment directions in one domain are reversed with respect to those in the other
- The two domains are related by the time inversion operator
- Ferromagnetic domains provide a simple example
- The intensity and the polarisation scattered by the two domains are identical

**Slip and translational domains**
- Regions in which all the moment directions in one domain are related to another by translational symmetry
- The intensity and the polarisation scattered by the two domains are identical

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Time reversal reverses magnetic moments

Apply translation \( t \)
Orientational domains (S-domains)
- Occurs when the symmetry of the magnetic structure is less than that of the crystal space group
- S domains are related by the symmetry elements that are lost (\( k \) does not change)
- The relationship between \( m \) and \( k \) is the same for all S domains
- Distinguish by single crystal diffraction, not powder diffraction

\[
e.g. \text{cubic with } k = \left(0, 0, \frac{1}{2}\right)
\]

Chiral domains
- Occurs when
  - Paramagnetic space group is centrosymmetric but the magnetic structure is not
  - The magnetic moments on centrosymmetrically related sites are not parallel
  - Incommensurate structures
    - when \( 2k \) is not a reciprocal lattice vector so the configurational group is acentric
    - In this case the two chirality domains correspond to \( +k \) and \( -k \).
    - They both give contributions at (hkl) \( \pm k \).
Fourier description of magnetic structures

- Each single domain follows

\[ m_j^k = \sum_{\nu, \ell} C_{\nu, \ell}^k \psi_\nu^k e^{-2\pi i \vec{k} \cdot \vec{\ell}} \]

- In the absence of unbalancing constraints (applied magnetic or electric field, pressure, etc) these will have the same energy

- Leads to questions
  - are there S-domains
  - multi-k or k-domain?

Example of diffraction pattern (structure) with 2k vectors:

\[ \left( \frac{1}{2}, 0, 0 \right) + \left( 0, \frac{1}{2}, 0 \right) \]

- Both will contribute to reflections at the same (hkl)
- Cannot distinguish by simple diffraction
  - 2k structure
  - 2 k-domains
\[ k_1 \cdot k_2 = 0, 1 \cdot k = 0, 0 \]

\[ k_1 \cdot k_2 = 0, 0, 0 \]

**k-domains vs multi k**

- **External constraint, e.g.**
  - Applied magnetic field
  - Pressure
- Leads to
  - Unbalancing domains
  - Domain repopulation
- Multi-k and k domains structures, S-domain structures will respond differently

\[ k_1 = (1/2, 0, 0) \quad k_2 = (0, 1/2, 0) \]

**Diffraction - Single crystal vs. powder**

- **Single crystal diffraction**
  - (diffraction pattern projected onto a line)
  - Cannot even see k-domains/multi-k
  - But you can always consider the possibilities and effects such as single-ion anisotropy...

- **Powder diffraction**