Talk Outline

• The ‘Golden Ratio’ in nature.
• Quasicrystals and incommensurate structures.
• Periodic functions vs. quasiperiodic functions.
• Aperiodic tiling.
• Embedding in higher dimensional space.
• Indexing in higher dimensional crystallography.
The ‘Golden Ratio’ in nature

- The Golden Ratio, τ or φ, can be found in many patterns in nature.
- Maximal packing.
- First defined by Euclid.
- Accurately describes the geometry of pentagons.\(^1\)

\[
\tau = \frac{AB}{AC} = 1.618034…
\]

Incommensurately modulated structures (IMS)

- Periodic distortions incommensurable with the translation periods of the basic lattice.
- Satellite peaks around main peaks.
- Structure factor has an extra contribution from reflections.  

\[
\begin{align*}
{r_{u,j}} & = r_j^0 + r_u \\
{r_{u,j}} & = r_j^0 + r_u + f_j(q \cdot (r_j^0 + r_u) - \phi) \\
g & = h a_1^* + k a_2^* + l a_3^* + m q
\end{align*}
\]

Electron diffraction pattern of \( \text{Ba}_{0.85}\text{Ca}_{2.15}\text{In}_6\text{O}_{12} \) showing satellite peaks due to incommensurate modulation.  


Five-fold symmetry in quasicrystals

- Discovery in 1984 by Shechtman leads to the term **quasicrystals** - any crystal that displays forbidden crystallographic symmetries.

- In 3-D, cannot be tiled by a single parallelepiped.

- Exhibits long-range order but not periodicity.

- Special kind of incommensurate structures.


Diffraction pattern of Al-Mn alloy showing icosahedral point symmetry m-5-3.
The math of quasiperiodic functions

• Periodic function \( f(x) \) can be expanded in a Fourier series.

\[
f(x) = f(x + np) \\
f'(x) = \sum_{n_1=-\infty}^{\infty} f^*(n_1) \exp[2\pi i n_1 x] \\
f(x_1,\ldots,x_N) = \sum_{n_1,\ldots,n_N} f^*(n_1,\ldots,n_N) \exp[2\pi i (n_1 x_1 + \cdots + n_N x_N)] \\
g(x) = \sum_{n_1,\ldots,n_N} f^*(n_1,\ldots,n_N) \exp[2\pi i (n_1 \nu_1 + \cdots + n_N \nu_N) x]
\]

• 2D to 1D example:

\[
f(x, y) = A_1 \sin(2\pi x) + A_2 \sin(2\pi y) \\
y = \alpha x \\
f'(x) = A_1 \sin(2\pi x) + A_2 \sin(2\pi \alpha x)
\]

• \( g(x) \) and \( f'(x) \) are quasiperiodic functions if \( \nu \) and \( \alpha \) are irrational numbers.
Approximations with periodic functions

- Model with a series of delta functions (crosses and open circles).

\[
\sum_n \delta(x - na) \\
\sum_m \delta(x - ma \tau/2)
\]

\[
\tau = 1 + \frac{1}{1 + \frac{1}{1 + \frac{1}{1 + \ldots}}}
\]

\[
\tau_N = \tau
\]

\[
\tau_0 = 1
\]

\[
\tau_1 = 1 + 1 = 2
\]

\[
\tau_2 = 1 + 1/(1+1) = 3/2
\]

\[
\tau_3 = 1 + 1/(1+1/(1+1)) = 5/3
\]
Penrose tiling

- Example of aperiodic tiling.

- Properties of Penrose quasicrystal.
  - Quasiperiodic translational order.
  - Minimal separation between atoms.
  - Orientational order.

- The two prototiles have matching rules.

- Matching rules correspond to physical forces in a material.

- Recently, Steinhardt and Jeong showed that you can tile with decagon and maximize density.\(^5\)

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Fourier transforms of Penrose tiling and quasiperiodic functions

- In 1982 Mackay proves that aperiodic tilings can show sharp Bragg peaks.

Optical diffraction of Penrose tiling displays 10-fold symmetry.\(^7\)

Optical diffraction of Penrose tiling with red laser.\(^8\)

8. T. R. Welberry website http://rsc.anu.edu/~welberry/Optical_transform/
Embedding in higher-dimensional crystallography

• Several methods
  – Multigrid method.
  – Cut-and-project method.
  – Projection method.

• Can extend the methods to higher dimensions.

• For icosahedral 3-D groups, can be thought of cut-and-project strips in 6-D.

• Would use $h/h'$, $k/k'$, $l/l'$ to index an icosahedral superspace group.

Example of cut-and-project method in which a cut with an irrational slope through 2-D leads to Fibonacci sequence in 1-D.\textsuperscript{5}
N-dimensional crystallography

- Deal with periodic functions and therefore the usual formalism of crystallographic space groups.

- For quasicrystals, new point groups can be achieved (e.g. pentagonal, octagonal, icosahedral).

- Crystallographic point groups have different ranks, or the number of dimensions to make the spacing periodic.

### Point groups in higher dimensions

<table>
<thead>
<tr>
<th>Point group</th>
<th>Rank</th>
<th>( R_1 )</th>
<th>( R_2 )</th>
<th>( R_3 )</th>
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<td>((x, u, v, z, y))</td>
<td>((x + 1, u + 1, x + 1, y + 1))</td>
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<td>5m</td>
<td>5</td>
<td>( (y, z, u, v, x) )</td>
<td>((x + 1, u + 1, x + 1, y + 1))</td>
<td>((x, v, u, z, y))</td>
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<td>((x + 1, v + 1, u + 1, z + 1, y + 1))</td>
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<tr>
<td>( \bar{5} )</td>
<td>5</td>
<td>( (y, z, u, v, x) )</td>
<td>((x, v, u, z, y))</td>
<td>((x + 1, v + 1, u + 1, z + 1, y + 1))</td>
</tr>
<tr>
<td>( \bar{5}m )</td>
<td>5</td>
<td>( (y, z, u, v, x) )</td>
<td>((x, v, u, z, y))</td>
<td>((x + 1, v + 1, u + 1, z + 1, y + 1))</td>
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<tr>
<td>5</td>
<td>001, (100)</td>
<td>( (x + p/5, z, u, v, \bar{s}) )</td>
<td>(p = 0, 1, 2)</td>
<td>(x, v, u, z, y)</td>
</tr>
<tr>
<td>5m1</td>
<td>5</td>
<td>( (x, z, u, v, \bar{s}) )</td>
<td>(x, z, u, v, x)</td>
<td>(x + 1, z, u, v, x)</td>
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<tr>
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<td>(x, z, u, v, x)</td>
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<tr>
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<td>(x, \bar{z}, u, v, \bar{s})</td>
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<td>(x, v, S, y, \bar{z})</td>
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<td>10/m</td>
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<td>(x, v, S, y, \bar{z})</td>
<td>(x, v, S, y, \bar{z})</td>
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<tr>
<td>100mm</td>
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<td>( (x + 1, v, S, y, \bar{z}) )</td>
<td>(x, v, S, y, \bar{z})</td>
<td>(x, v, S, y, \bar{z})</td>
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<tr>
<td>1022</td>
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<td>(p = 0, \ldots, 5)</td>
<td>(x, v, S, y, \bar{z})</td>
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<tr>
<td>10 2m</td>
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<td>(x, v, S, y, \bar{z})</td>
</tr>
<tr>
<td>10  mm2</td>
<td>5</td>
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<td>(x, v, S, y, \bar{z})</td>
<td>(x, v, S, y, \bar{z})</td>
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<tr>
<td>10/mmm</td>
<td>5</td>
<td>( (x, v, S, y, \bar{z}) )</td>
<td>(x, v, S, y, \bar{z})</td>
<td>(x, v, S, y, \bar{z})</td>
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<tr>
<td>8</td>
<td>001, (100)</td>
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<td>(x, v, u, z, y)</td>
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<tr>
<td>8/m</td>
<td>5</td>
<td>( (x, v, u, z, y) )</td>
<td>(x, v, u, z, y)</td>
<td>(x, v, u, z, y)</td>
</tr>
</tbody>
</table>

Materials systems solved with higher dimensions

Dendritic aggregate of Al-Li-Cu alloy.

Pentagonally twinned Ge precipitate in Al matrix.

Single grain of the Al-Li-Cu alloy.

Programs for solving in higher dimensions

• **DIMS Direct Methods for Incommensurate Modulated/Composite Structures** - Fan, Hai-fu & colleagues
  [http://cryst.iphy.ac.cn/VEC/Tutorials/DIMS/DIMS.html](http://cryst.iphy.ac.cn/VEC/Tutorials/DIMS/DIMS.html)

• **Fullprof Rietveld** - Juan Rodriguez-Carvajal and **WinPlotr Interface** - T. Roisnel Jana2000
  [http://www-llb.cea.fr/fullweb/powder.htm](http://www-llb.cea.fr/fullweb/powder.htm)

• **JANA2000 Single Crystal and Powder Diffraction Software** - Vaclav Petricek

• **XND Rietveld** - Jean-Francois Berar
In Summary

• Special structural properties IMS and quasicrystals show the need for a more detailed crystallography.

• Quasiperiodic functions and aperiodic tilings help prove that long-range order does not require periodicity, just non-random packing.

• Embedding quasicrystals and IMS in higher dimensions leads to periodic structures that follow the rules of ordinary crystallography.