Icosahedral quasicrystals

higher dimensional description
&
structure determination

Hiroyuki Takakura

北海道大学
HOKKAIDO UNIVERSITY

Division of Applied Physics, Faculty of Engineering,
Hokkaido University

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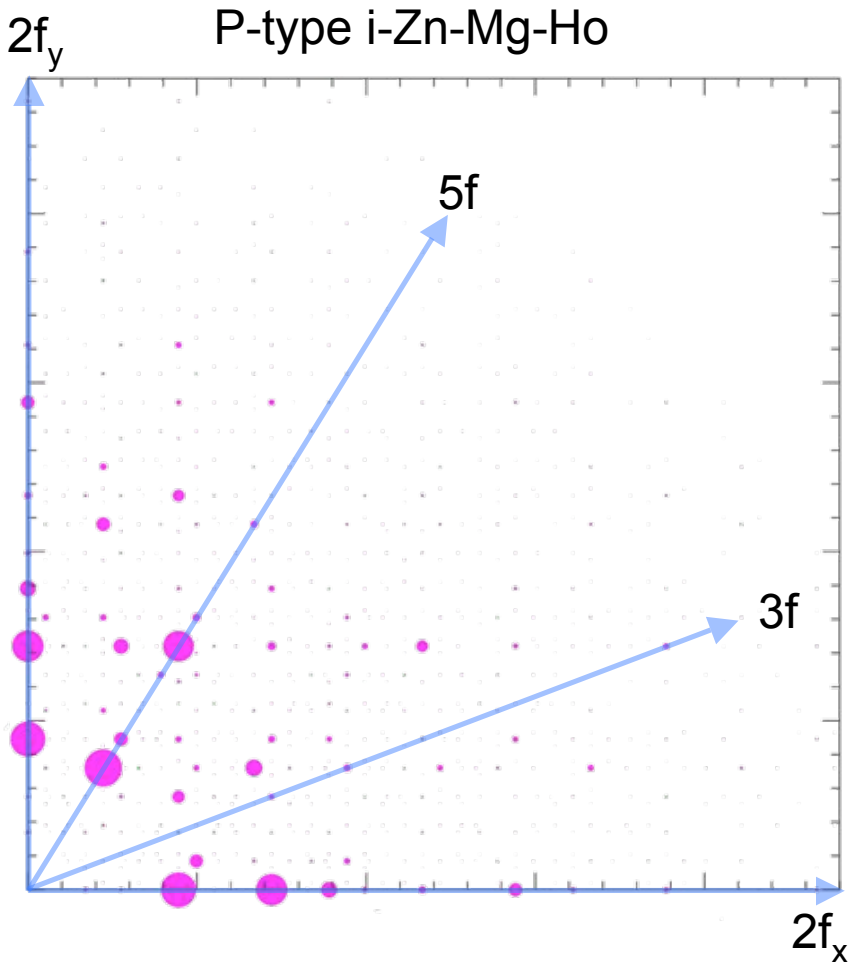
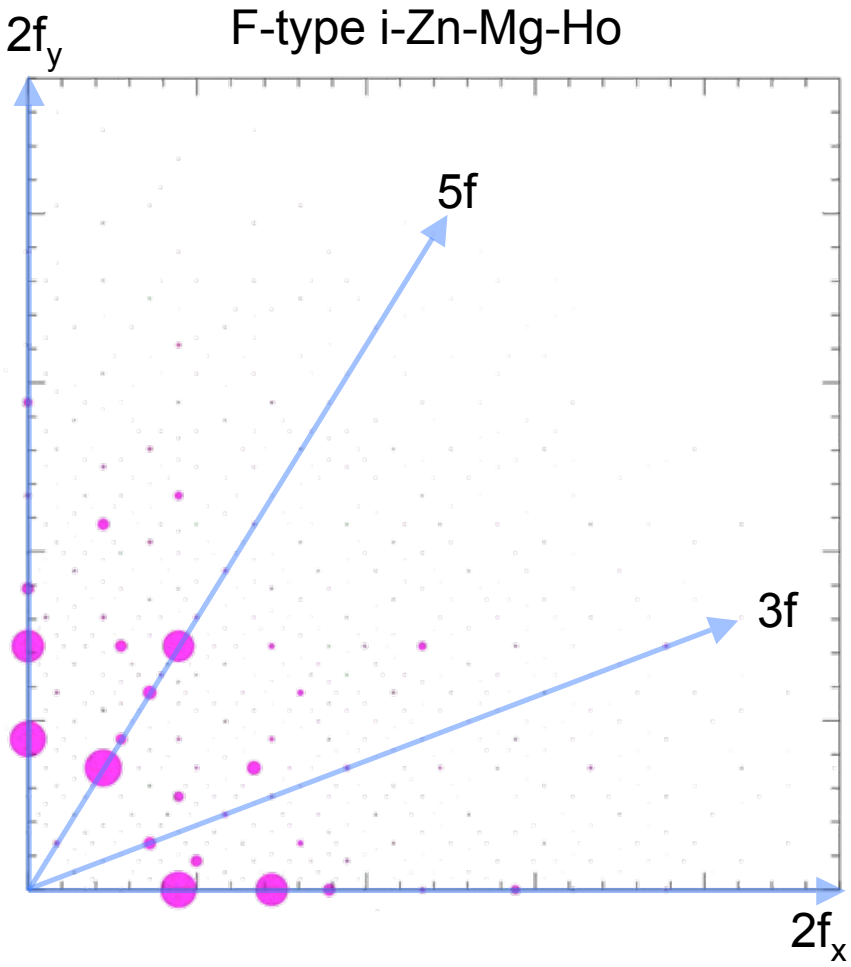
Overview

- Diffraction symmetry & Space groups of i-QC's
- Unit vectors in icosahedral system
  1. Reciprocal space (external & internal)
  2. Direct space (external & internal)
  3. Vectors for defining occupation domains
- Description of i-QC structures
  1. 3D Penrose tiling (Ammann tiling)
  2. Simple decoration model
- Low density elimination method
- Structure determination of i-YbCd (→ 2nd part)

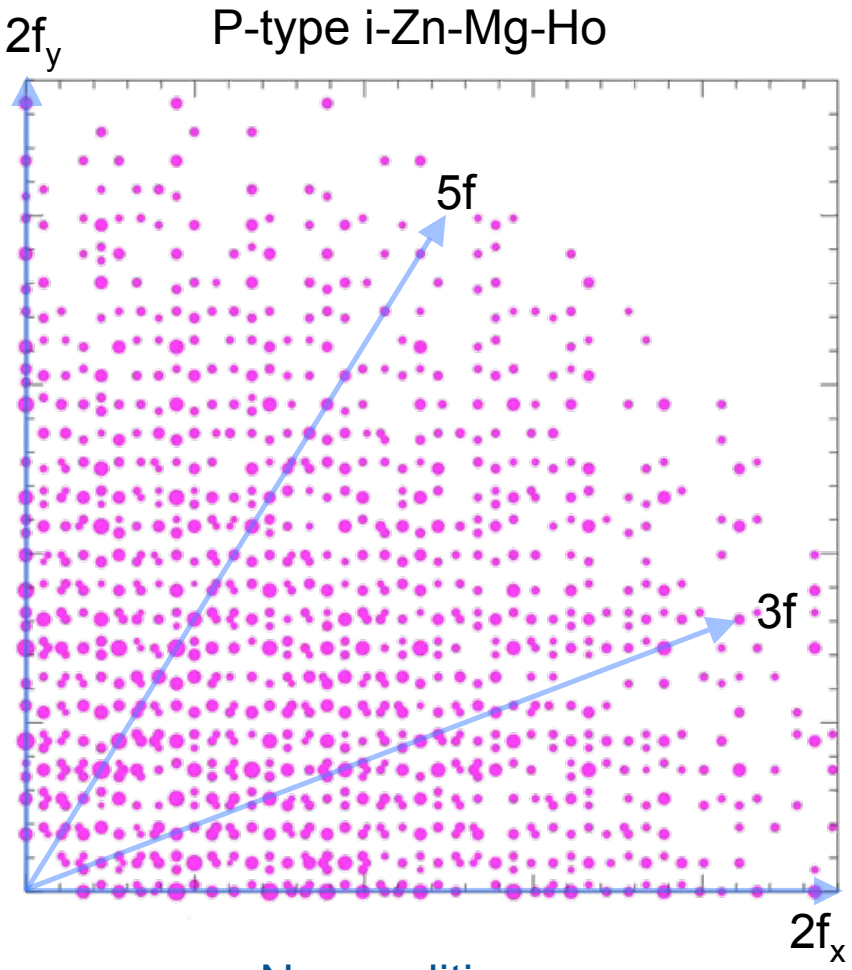
Diffraction symmetry
&
Space group of i-QC's

Diffraction patterns of i-QC's

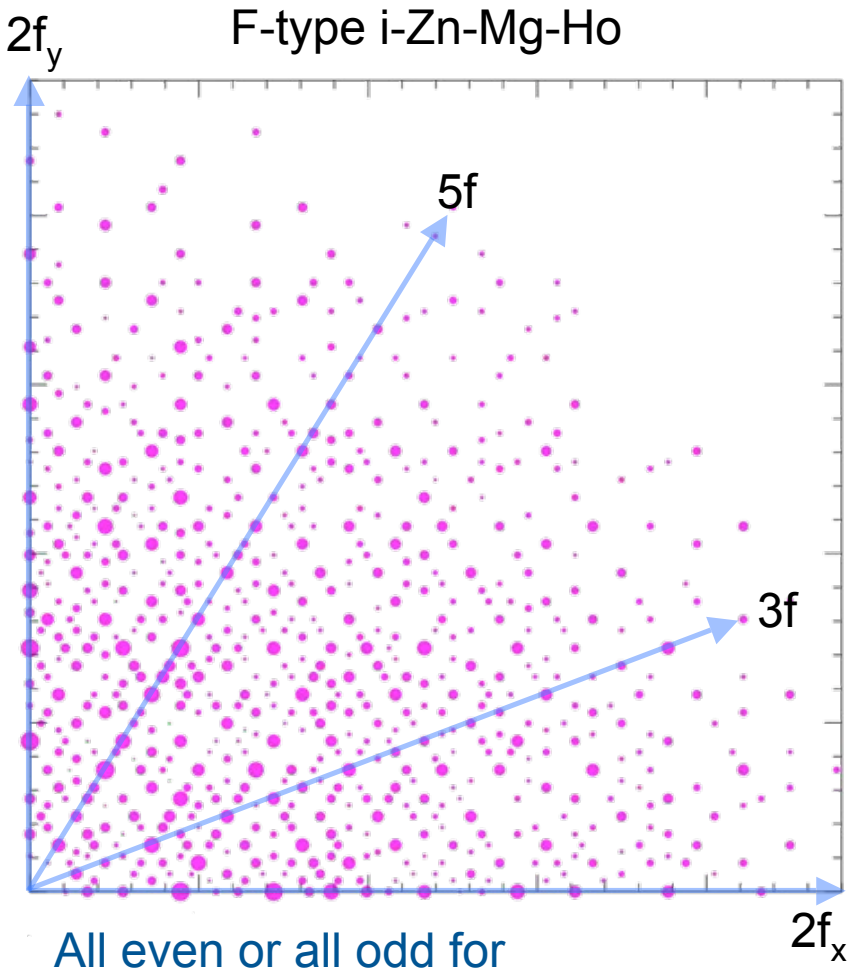




Their log plots



No condition



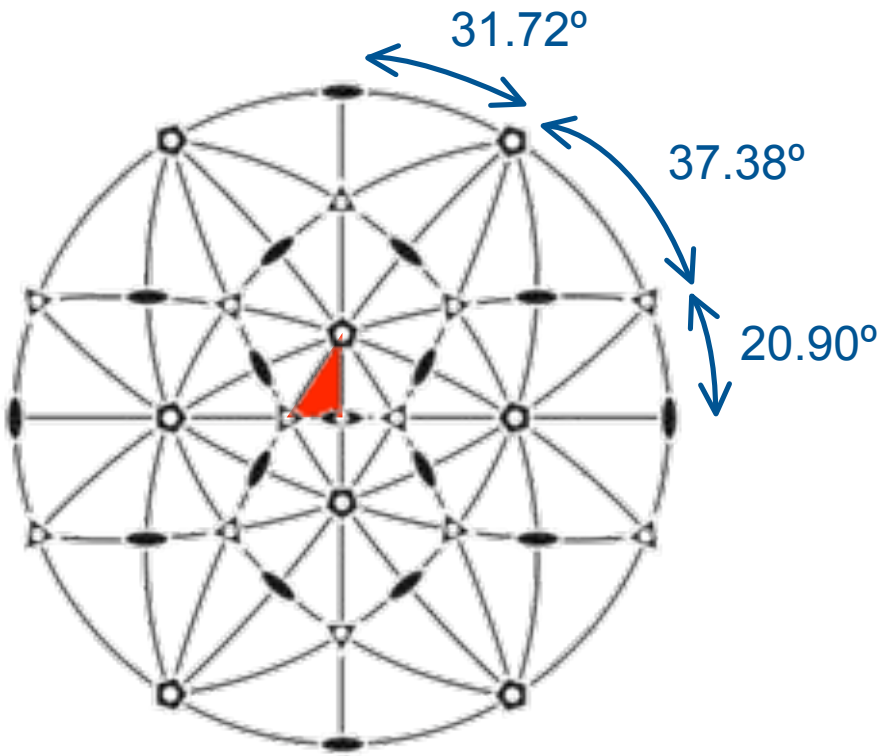
All even or all odd for

$h_1 h_2 h_3 h_4 h_5 h_6$

Symmetry of i-QC's

Point group $m\bar{3}\bar{5}$

Order : 120



Asymmetric region: $\frac{1}{120}$

6 +10 +15 + *m* + center

6D Icosahedral lattices

3 lattice types	Reflection condition
• Primitive $Pm\bar{3}\bar{5}$	No condition
• Body-centered $Im\bar{3}\bar{5}$	$\sum_{i=1}^{6} h_i = 2n$ for $h_1h_2h_3h_4h_5h_6$
• Face-centered $Fm\bar{3}\bar{5}$	All even or all odd for $h_1h_2h_3h_4h_5h_6$

Face-centered lattice

32 centering translations

$$(0,0,0,0,0,0)$$
$$(\tfrac{1}{2},\tfrac{1}{2},0,0,0,0)$$
$$(\tfrac{1}{2},0,\tfrac{1}{2},0,0,0)$$
$$\ldots$$

Body-centered lattcie

2 centering translations

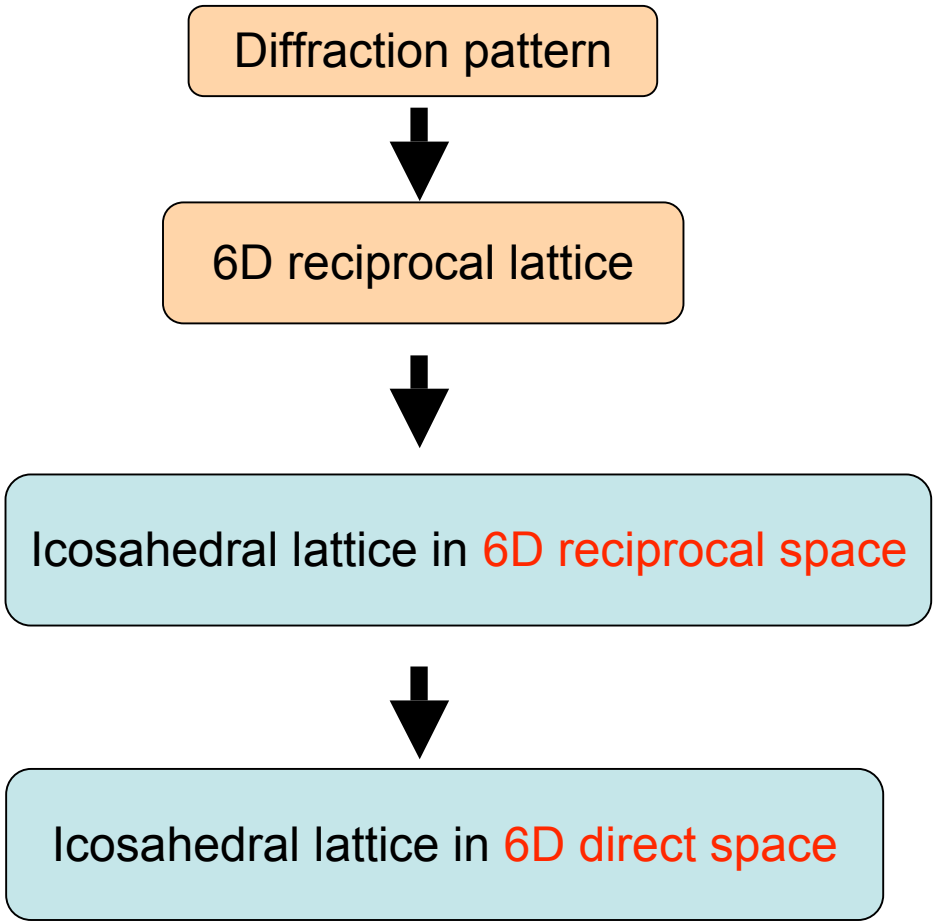
$$(0,0,0,0,0,0)$$

$$(\tfrac{1}{2},\tfrac{1}{2},\tfrac{1}{2},\tfrac{1}{2},\tfrac{1}{2},\tfrac{1}{2})$$

- Body-centered lattice has not observed yet experimentally

Unit vectors in icosahedral system

A flowchart representing a process for determining the unit vectors of i-QC's



Icosahedral symmetry

$$\mathbf{d}_i^{*\mathrm{e}} \quad (i = 1, \ldots, 6)$$

6 indices: $h_1 h_2 h_3 h_4 h_5 h_6$

Lattice constant: a^*

$$\mathbf{d}_i^* = \mathbf{d}_i^{*\mathrm{e}} + \mathbf{d}_i^{*\mathrm{i}} \quad (i = 1, \ldots, 6)$$

(external) (internal)

$$\mathbf{d}_i = \mathbf{d}_i^{\mathrm{e}} + \mathbf{d}_i^{\mathrm{i}} \quad (i = 1, \ldots, 6)$$

Lattice constant: $a = \frac{1}{2a^*}$

Unit vectors in 6D reciprocal space

$\mathbf{d}_i^* \ (i = 1, 2, \ldots, 6)$: unit reciprocal lattice vectors

$\mathbf{a}_1, \mathbf{a}_2, \mathbf{a}_3, \mathbf{a}_4, \mathbf{a}_5, \mathbf{a}_6$: orthonormal base vectors

$\mathbf{a}_1, \mathbf{a}_2, \mathbf{a}_3$: span the external space（3D）

$\mathbf{a}_4, \mathbf{a}_5, \mathbf{a}_6$: span the internal space (3D)

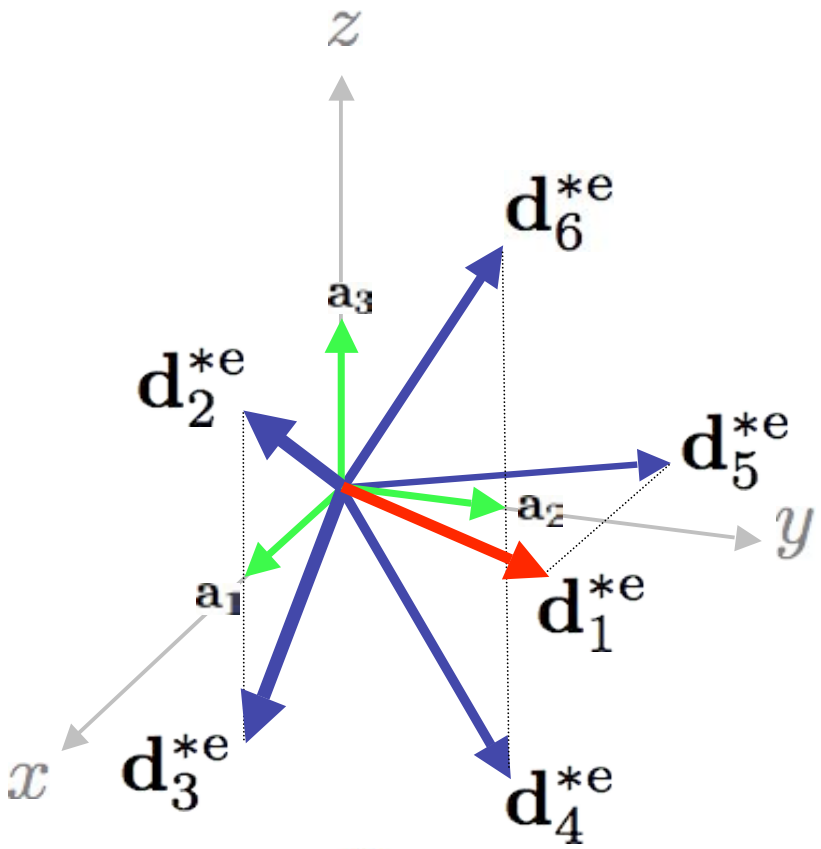
$$\begin{pmatrix} \mathbf{d}_1^* \\ \mathbf{d}_2^* \\ \mathbf{d}_3^* \\ \mathbf{d}_4^* \\ \mathbf{d}_5^* \\ \mathbf{d}_6^* \end{pmatrix} = \frac{a^*}{\sqrt{2+\tau}} \begin{pmatrix} 1 & \tau & 0 & \tau & -1 & 0 \\ \tau & 0 & 1 & -1 & 0 & \tau \\ \tau & 0 & -1 & -1 & 0 & -\tau \\ 0 & 1 & -\tau & 0 & \tau & 1 \\ -1 & \tau & 0 & -\tau & -1 & 0 \\ 0 & 1 & \tau & 0 & \tau & -1 \end{pmatrix} \begin{pmatrix} \mathbf{a}_1 \\ \mathbf{a}_2 \\ \mathbf{a}_3 \\ \mathbf{a}_4 \\ \mathbf{a}_5 \\ \mathbf{a}_6 \end{pmatrix}$$

$$\tilde{M}^{-1}$$

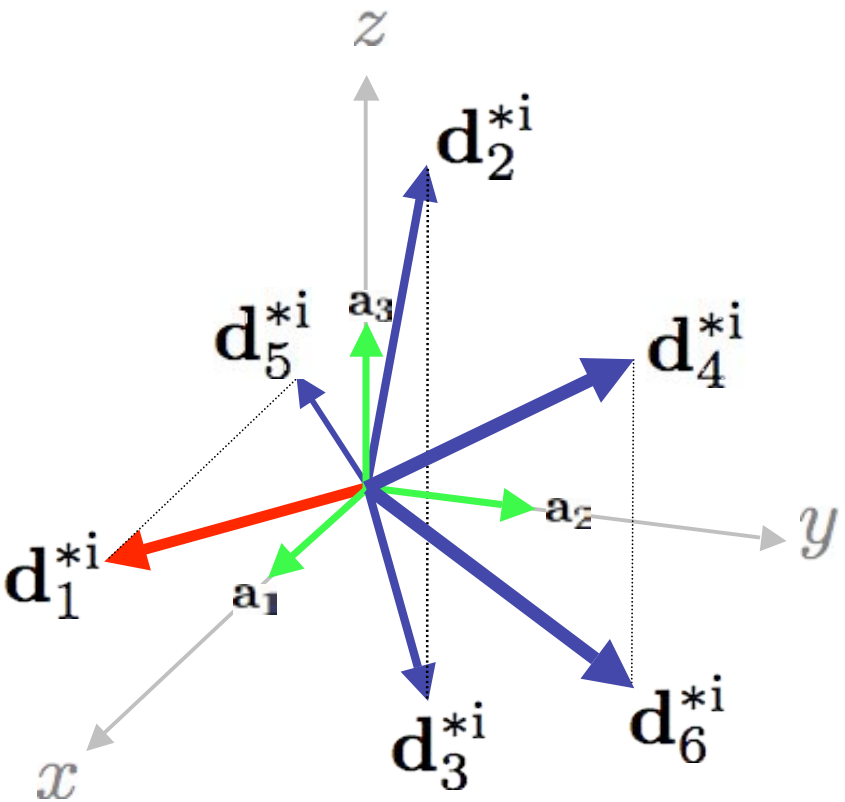
a^* : lattice constant in reciprocal space

Projection of the unit vectors $\mathbf{d}_i^*$

External space



Internal space



$$\mathbf{h}^e = \sum_{i=1}^{6} h_i \mathbf{d}_i^{*e}$$

Unit vectors in 6D direct space

Reciprocal lattice vectors $\mathbf{d}_i^* \ (i = 1, 2, \ldots, 6)$ → Direct lattice vectors $\mathbf{d}_i \ (i = 1, 2, \ldots, 6)$

$$\mathbf{d}_i^* = \sum_{j=1}^{6} \tilde{M}_{ij}^{-1} \mathbf{a}_j \qquad \mathbf{d}_i = \sum_{j=1}^{6} M_{ij} \mathbf{a}_j$$

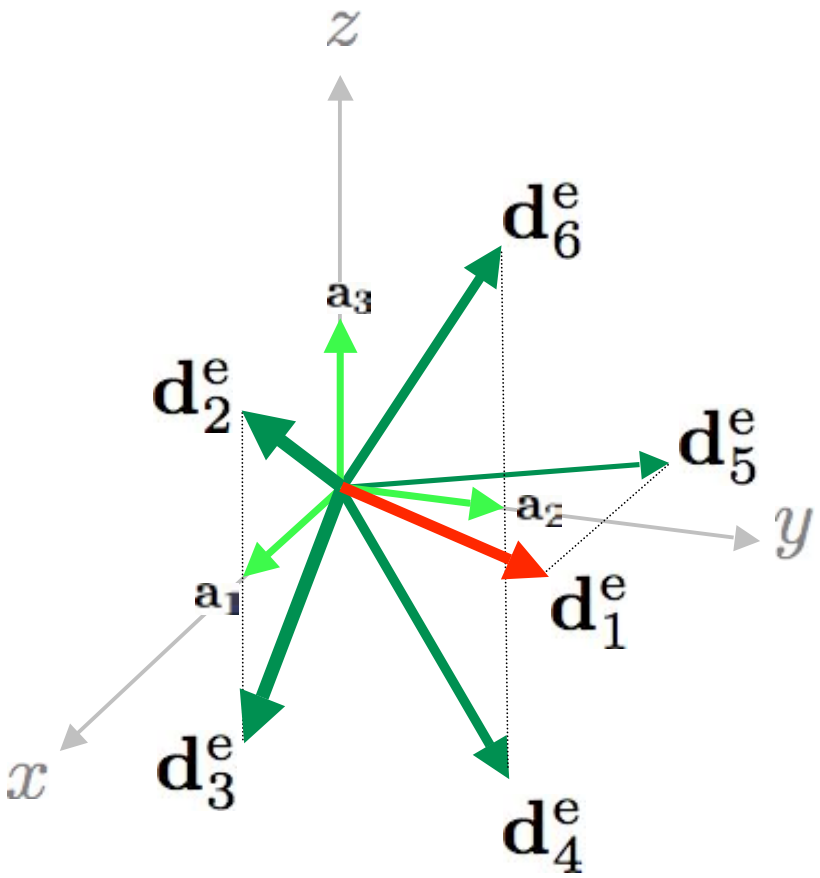
$$\begin{pmatrix} \mathbf{d}_1 \\ \mathbf{d}_2 \\ \mathbf{d}_3 \\ \mathbf{d}_4 \\ \mathbf{d}_5 \\ \mathbf{d}_6 \end{pmatrix} = \frac{a}{\sqrt{2+\tau}} \begin{pmatrix} 1 & \tau & 0 & \tau & -1 & 0 \\ \tau & 0 & 1 & -1 & 0 & \tau \\ \tau & 0 & -1 & -1 & 0 & -\tau \\ 0 & 1 & -\tau & 0 & \tau & 1 \\ -1 & \tau & 0 & -\tau & -1 & 0 \\ 0 & 1 & \tau & 0 & \tau & -1 \end{pmatrix} \begin{pmatrix} \mathbf{a}_1 \\ \mathbf{a}_2 \\ \mathbf{a}_3 \\ \mathbf{a}_4 \\ \mathbf{a}_5 \\ \mathbf{a}_6 \end{pmatrix}$$

M

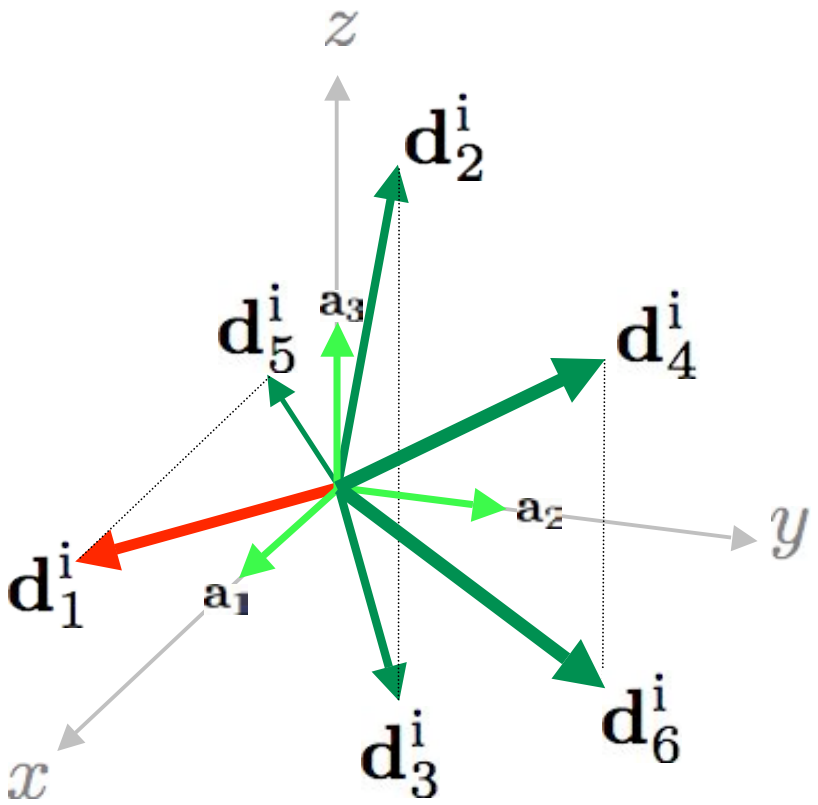
$a = \frac{1}{2a^*}$: lattice constant

Projection of the unit vectors $\mathbf{d}_i$

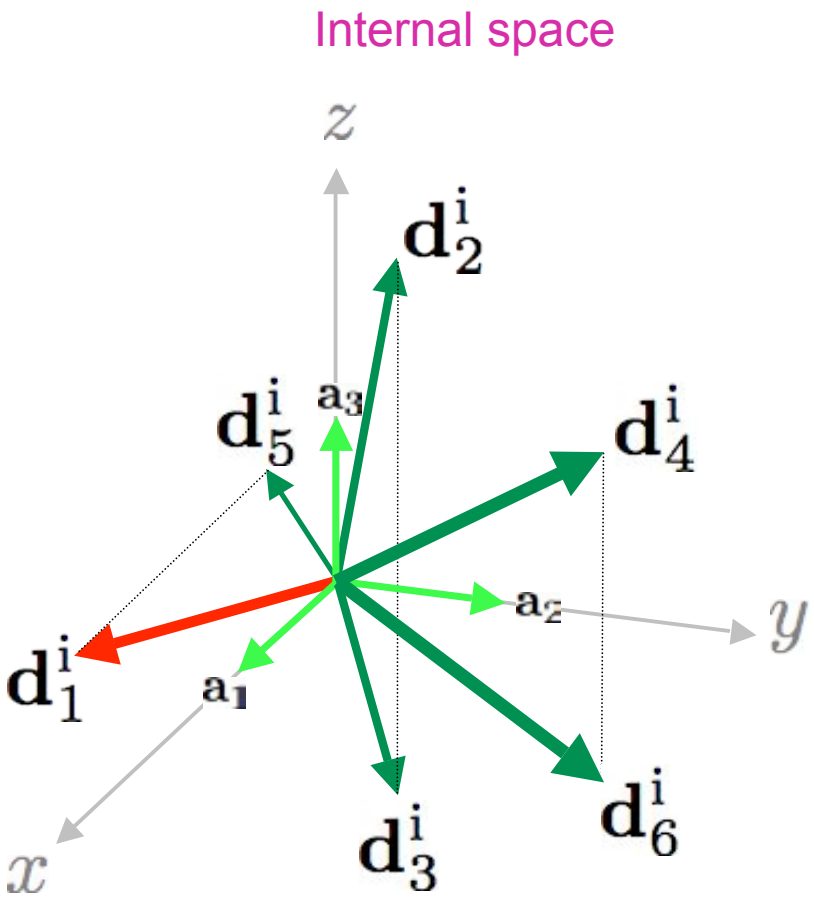
External space



Internal space



Vectors employed for defining the occupation domains of i-QC's

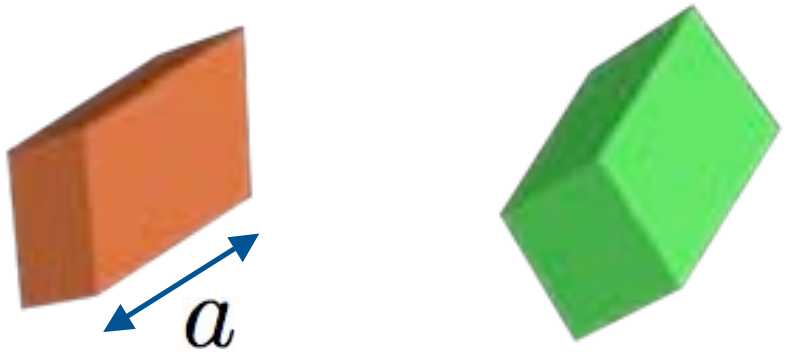


Description of i-QC structures

Three dimensional Penrose tiling (or Ammann tiling)

3D Penrose tiling

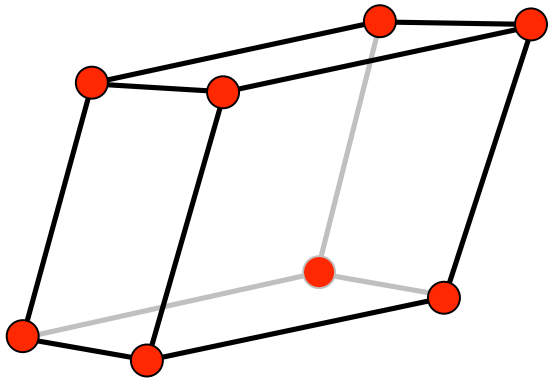
- A 3D analog of Penrose tiling.
- The 3DPT consists of two primitive rhombohedra: acute rhombohedron (AR) and obtuse rhombohedron (OR).
- Space-filling structure with icosahedral symmetry.



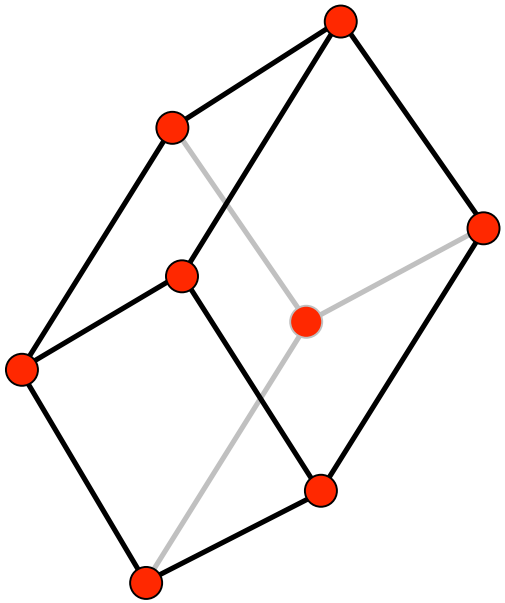
AR

OR

Vertex decoration of the rhombohedra in the 3DPT



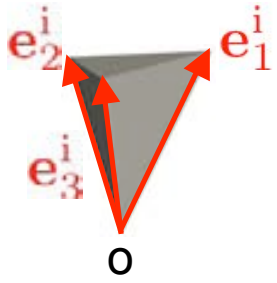
AR



OR

Construction of 3DPT by the section method

Occupation Domain



Asymmetric part

Corner vectors

$$\mathbf{e}_1 = (1, \bar{1}, \bar{1}, \bar{1}, \bar{1}, \bar{1})/2$$
$$\mathbf{e}_2 = (1, 1, \bar{1}, \bar{1}, \bar{1}, \bar{1})/2$$
$$\mathbf{e}_3 = (1, 0, \bar{1}, \bar{1}, 0, \bar{1})/2$$

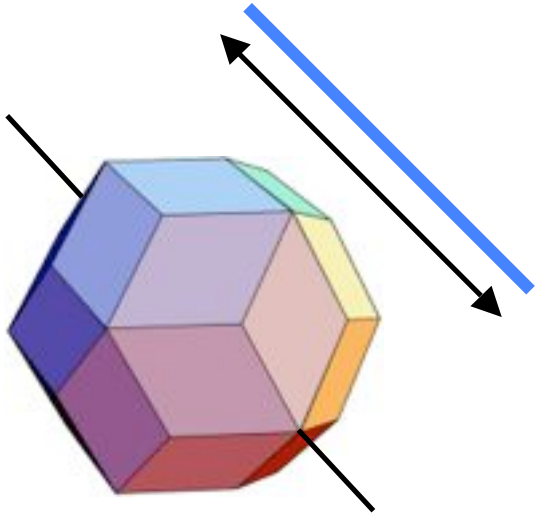
Site symmetry

$(0, 0, 0, 0, 0, 0)$

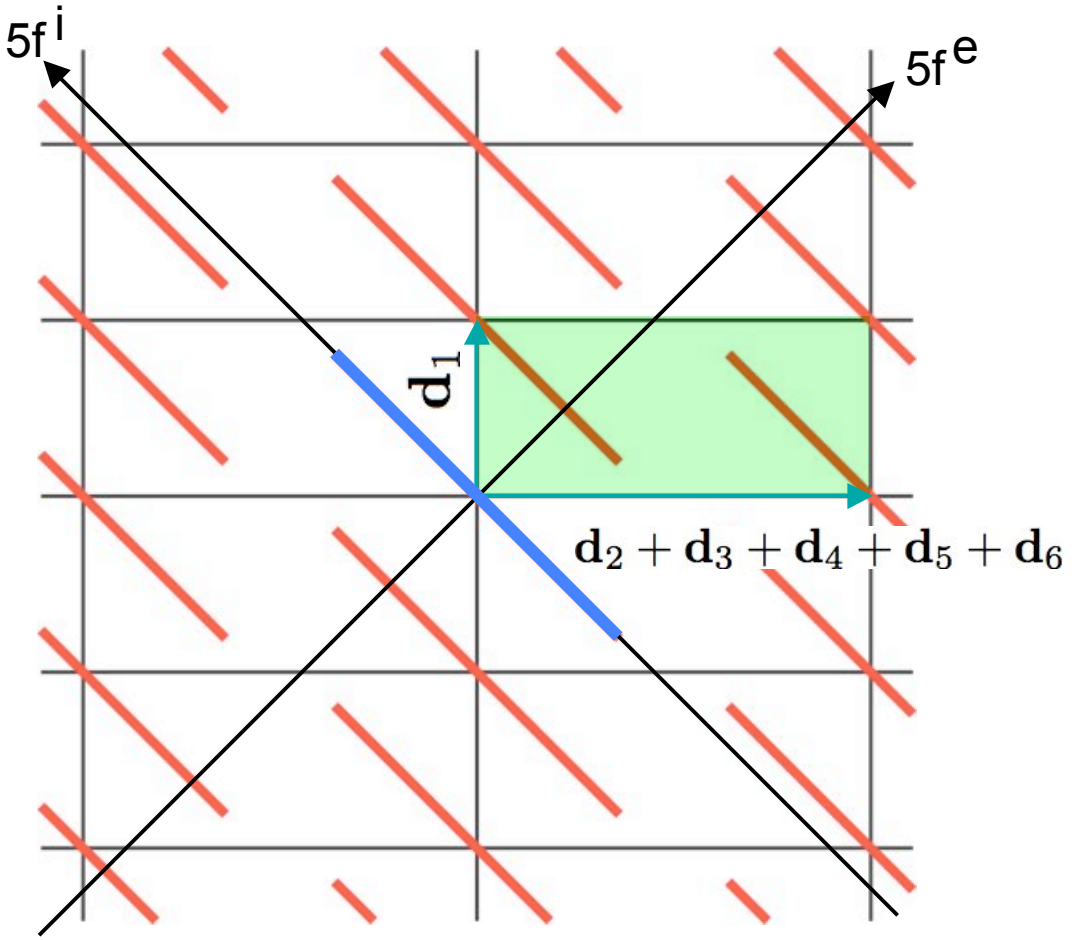
$m\bar{3}\bar{5}$

The rhombic triacontahedron is the projection of 6D unit cell onto the 3D internal space.

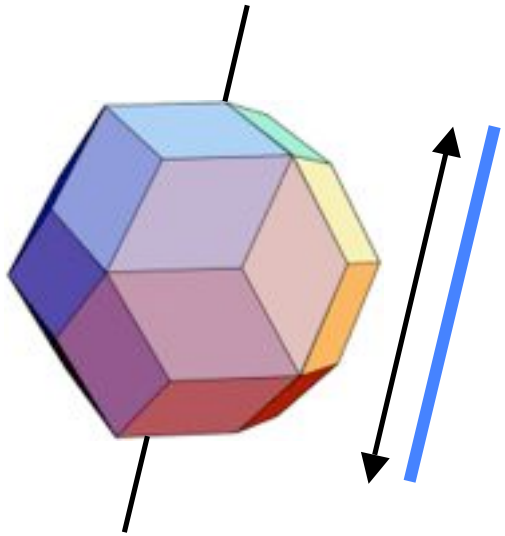
2D section of the 6D structure including 5f axes



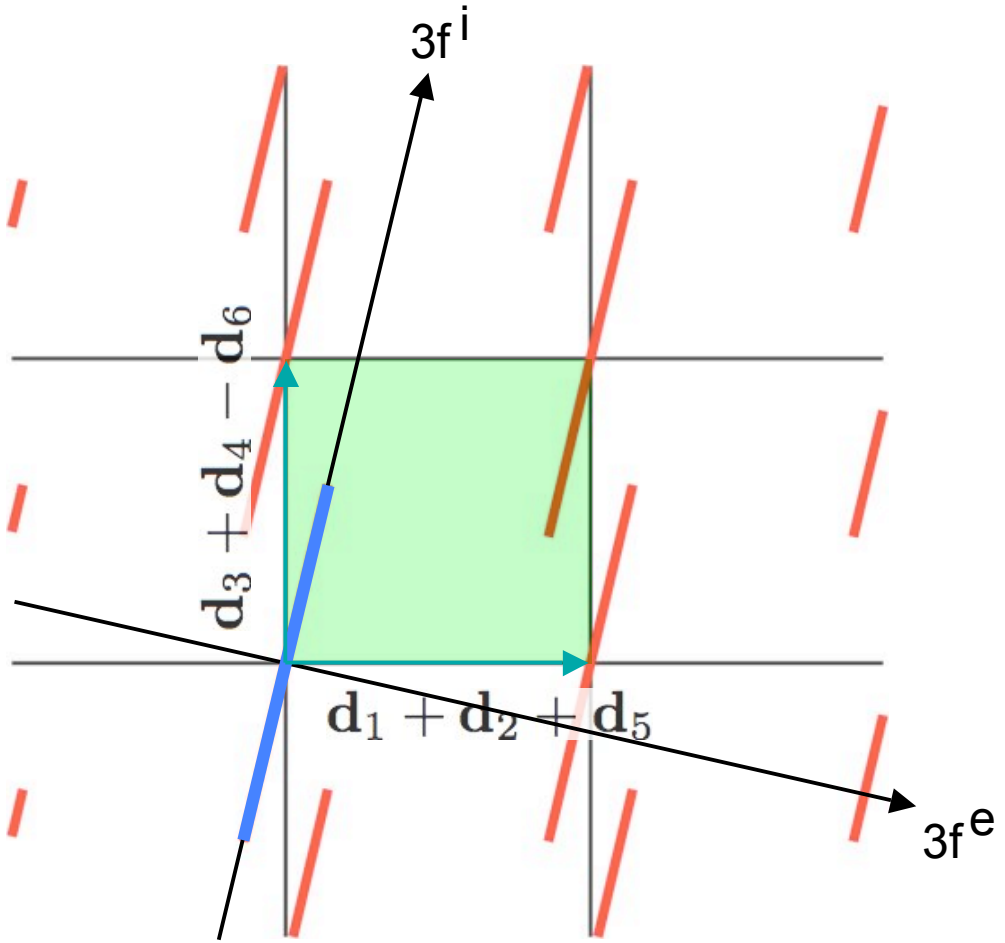
The bar is the 1D section of the OD along a 5f axis.



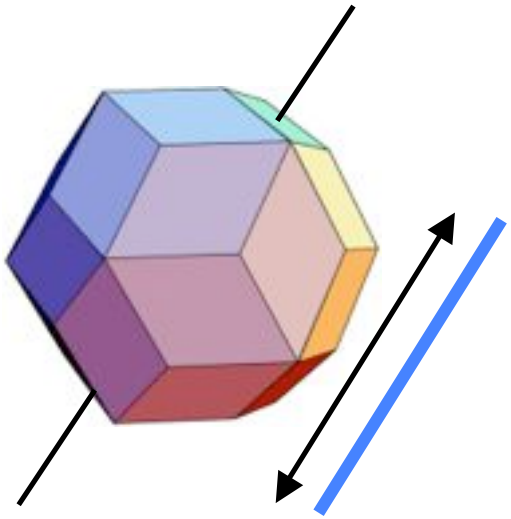
2D section of the 6D structure including 3f axes



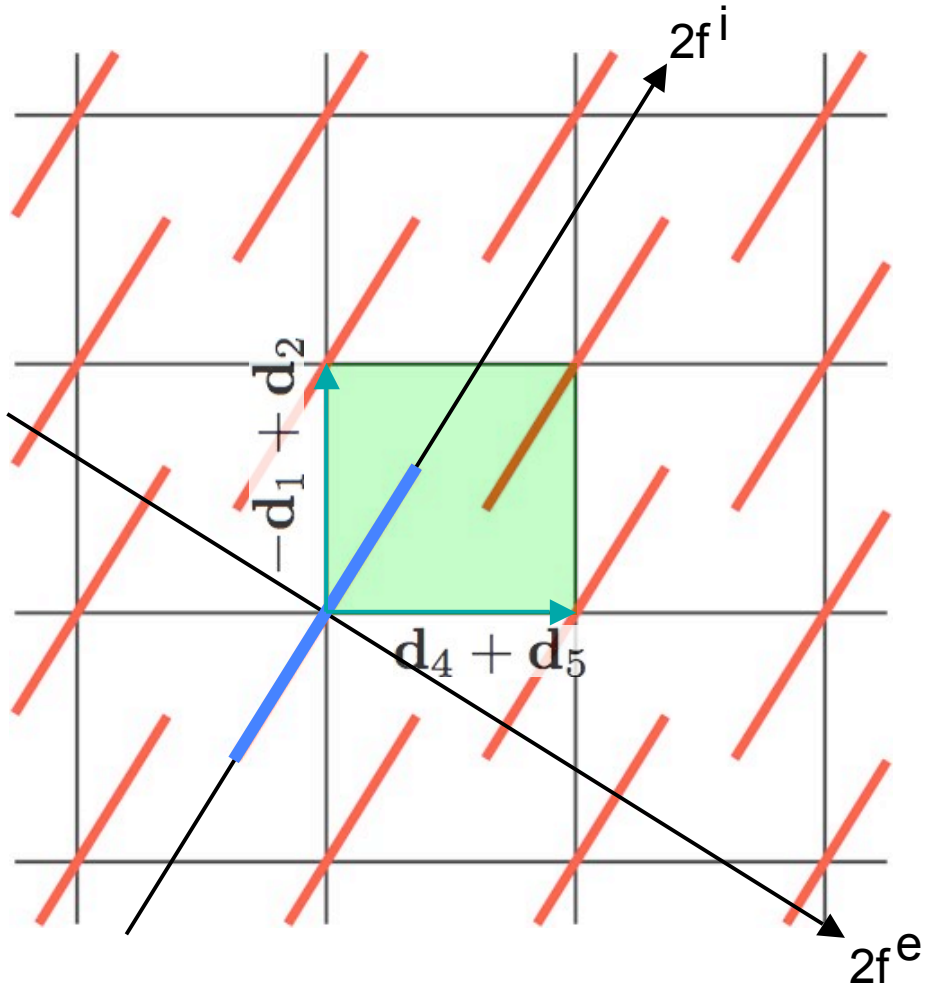
The bar is the 1D section of the OD along a 3f axis.



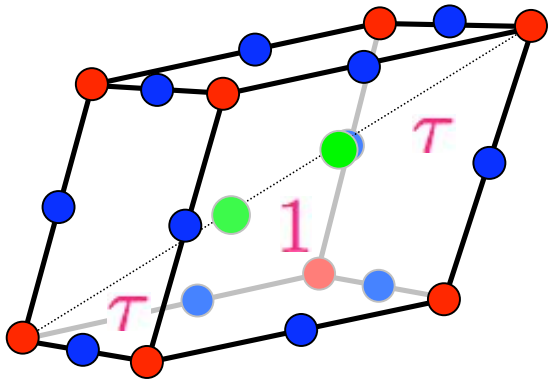
2D section of the 6D structure including 2f axes

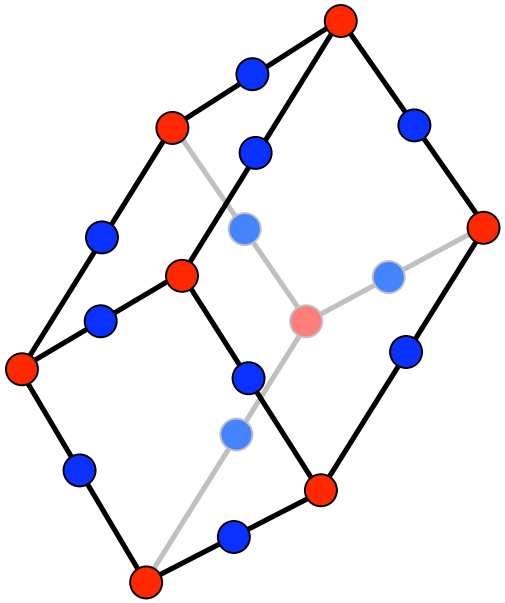


The bar is the 1D section of the OD along a 2f axis.



Simple decoration model of the 3DPT



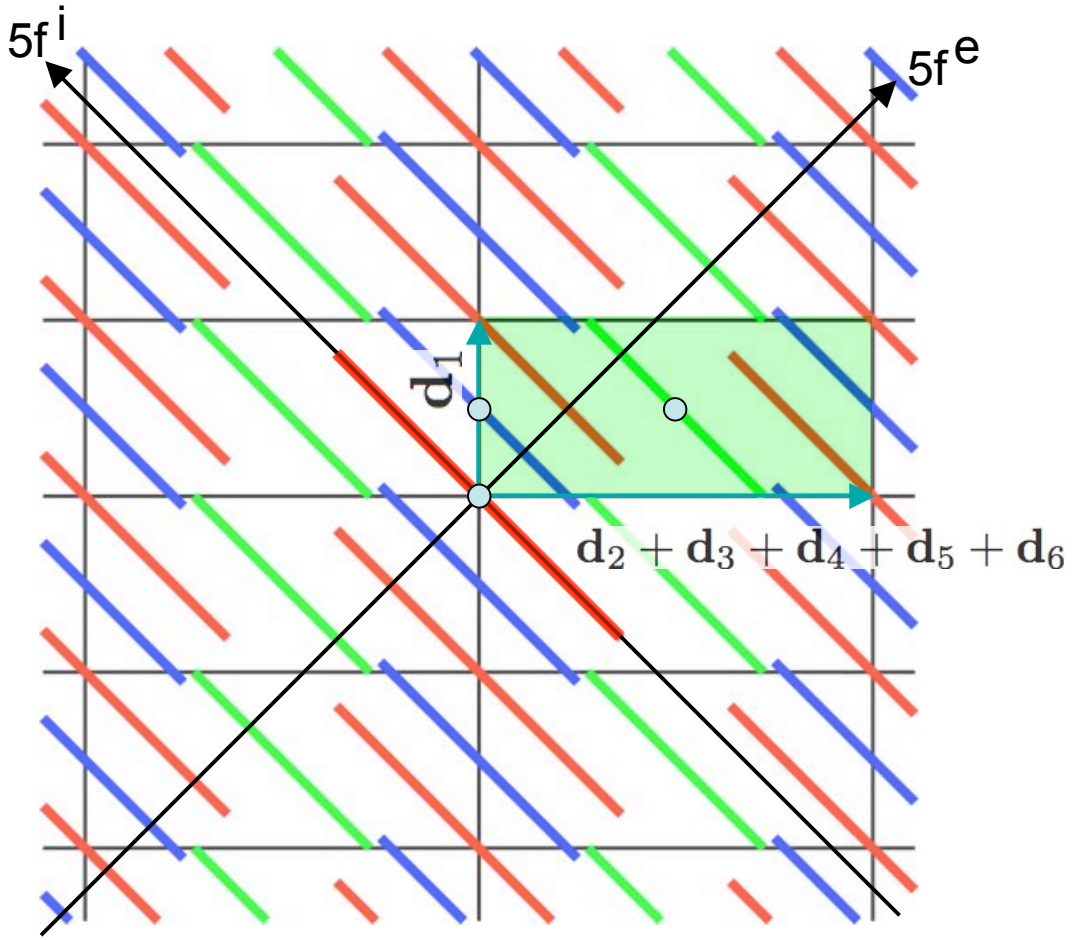


AR

OR

- Vertices
- Mid edges
- Body diagonal two positions in AR's

Simple decoration model in 6D



Site-symmetry group

RT  $m\bar{3}\bar{5}$

V: $(0,0,0,0,0,0)$

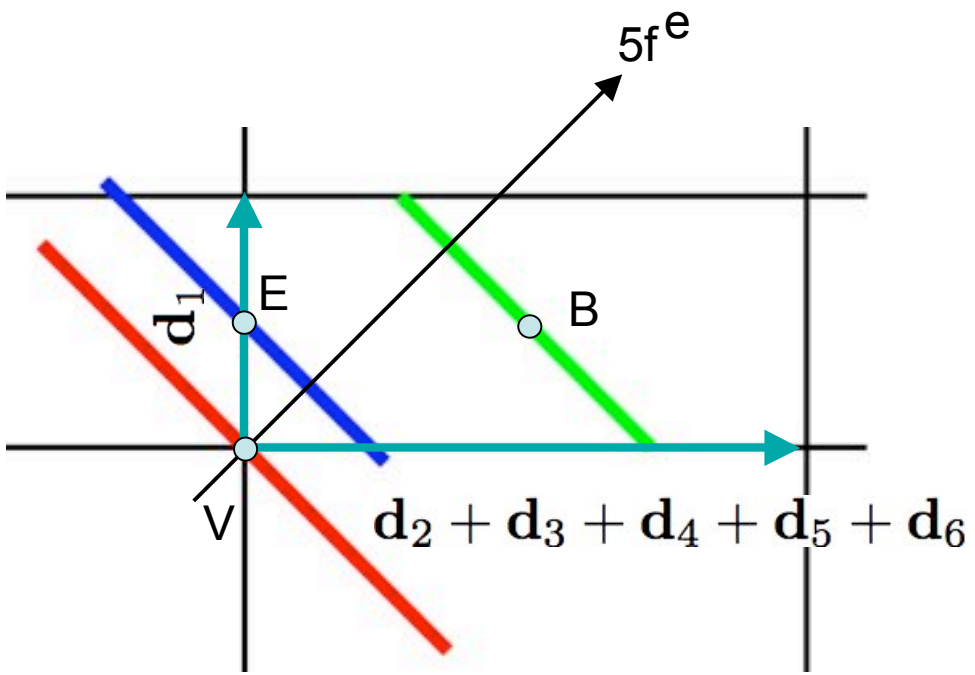
FD  $m\bar{3}\bar{5}$

B: $(\frac{1}{2},\frac{1}{2},\frac{1}{2},\frac{1}{2},\frac{1}{2},\frac{1}{2})$

RI  $m\bar{5}$

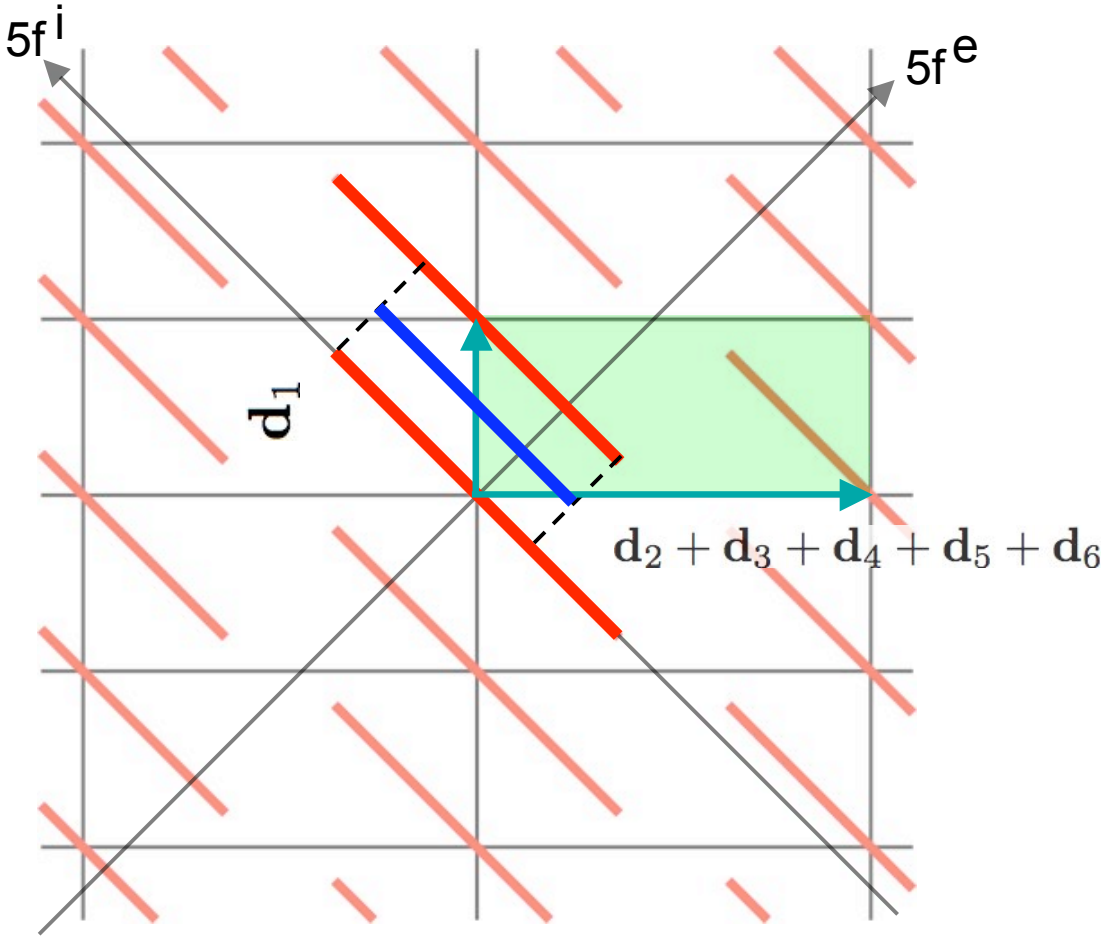
E: $(\frac{1}{2},0,0,0,0,0)$

Independent occupation domains and their shapes in the internal space.



How to obtain the OD at E

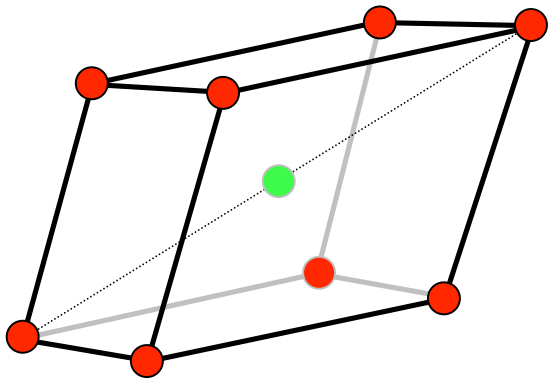
$(\frac{1}{2},0,0,0,0,0)$

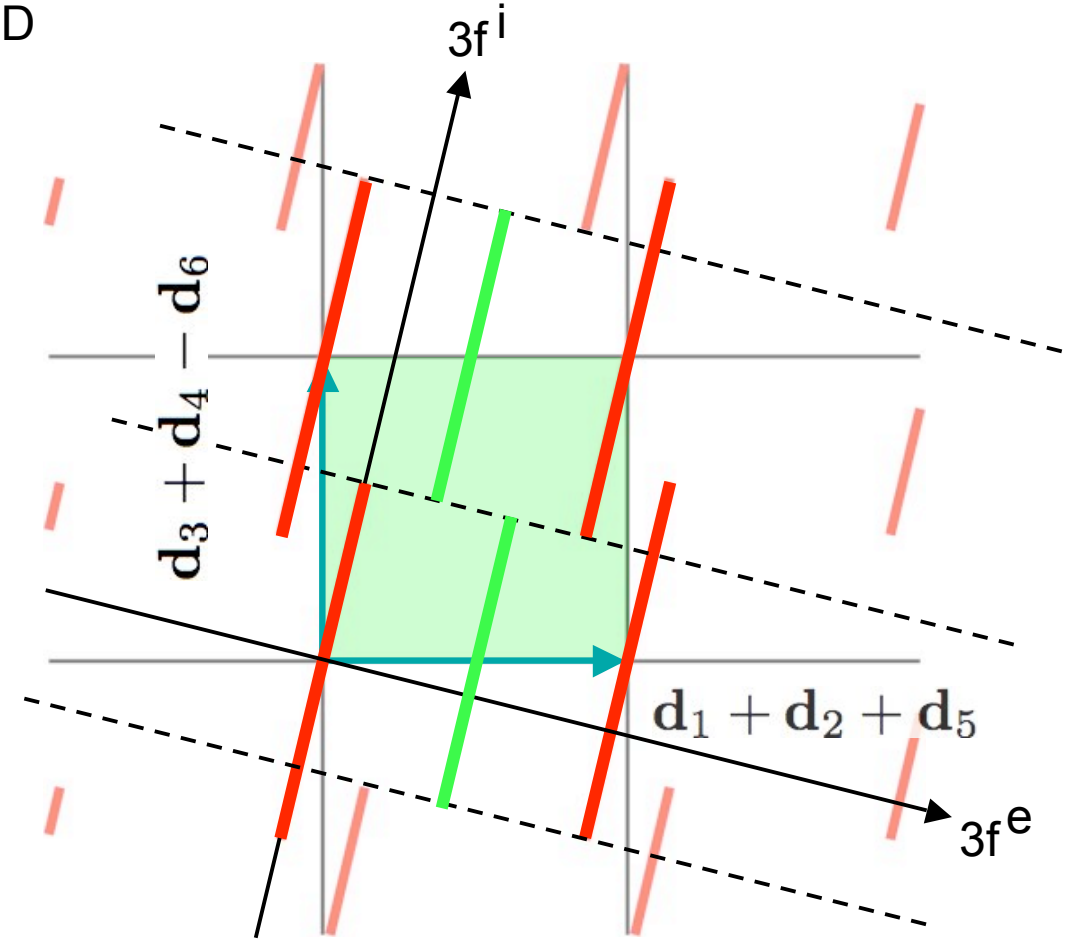


How to obtain the OD at B (step no.1)

Acute rhombohedral-shaped OD

Center: $(\frac{1}{2}, \frac{1}{2}, 0, 0, \frac{1}{2}, 0)$





Length of the diagonal: $|\mathbf{d}_1^{\mathrm{e}} + \mathbf{d}_2^{\mathrm{e}} + \mathbf{d}_5^{\mathrm{e}}| = 1 + \tau$

(step no.2)

20AR

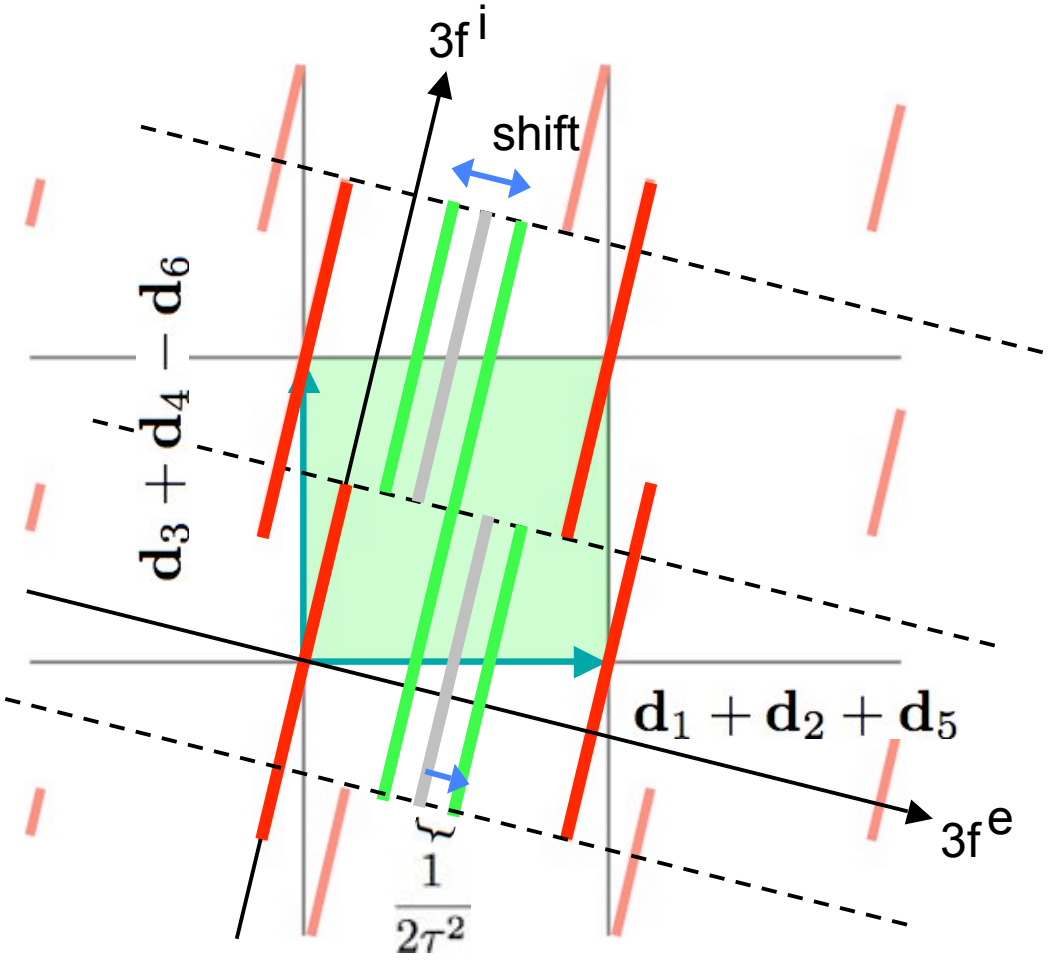
Center: $(\frac{1}{2}, \frac{1}{2}, 0, 0, \frac{1}{2}, 0)$ + shift along E-space

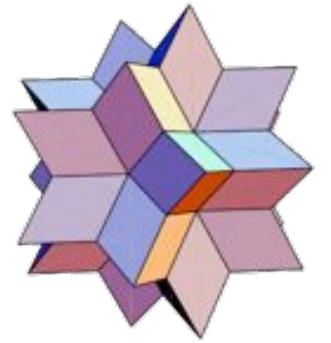
...

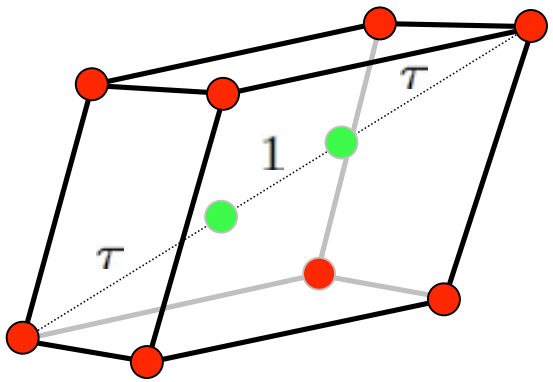
...

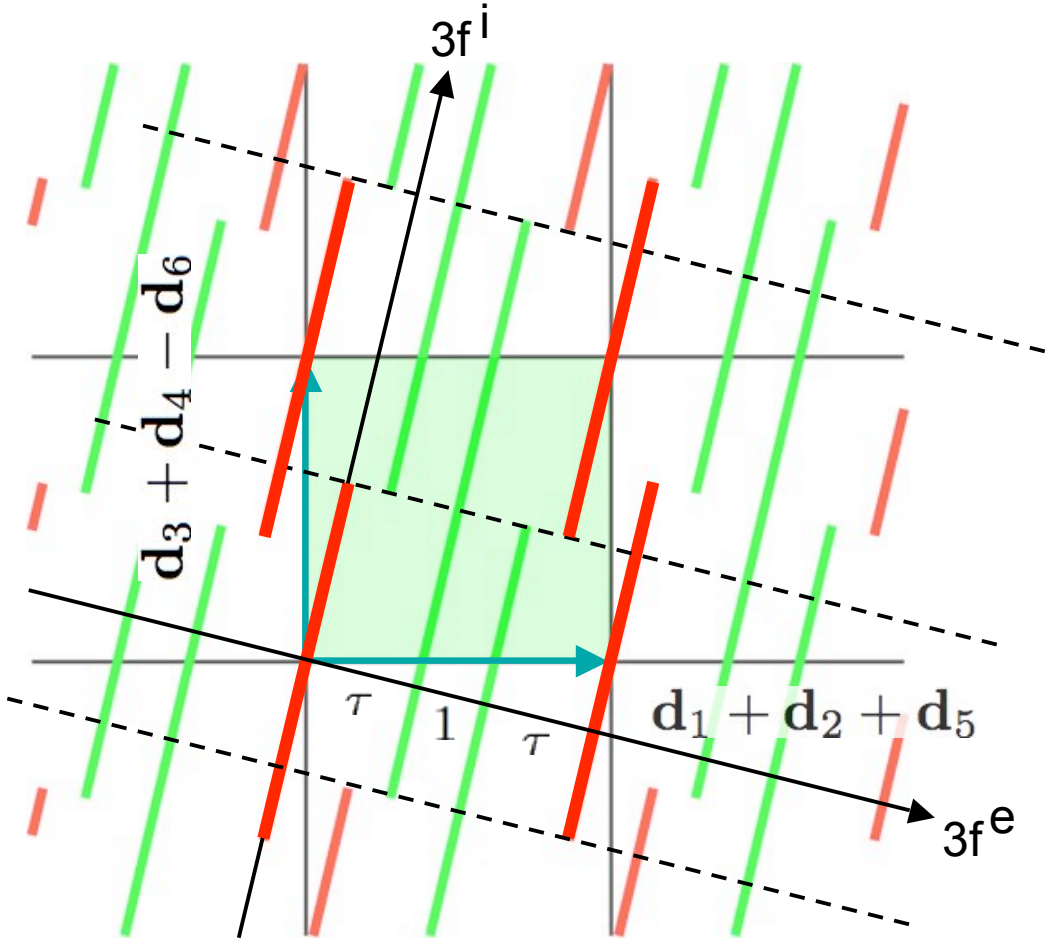
FD

Center: $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}, \frac{1}{2}, \frac{1}{2})$









The result is equivalent to put FD shaped OD on (1,1,1,1,1,1)/2 instead of putting the AR shaped OD's on the edges of 6D unit cell.

Low density elimination method

Phase problem in QC's

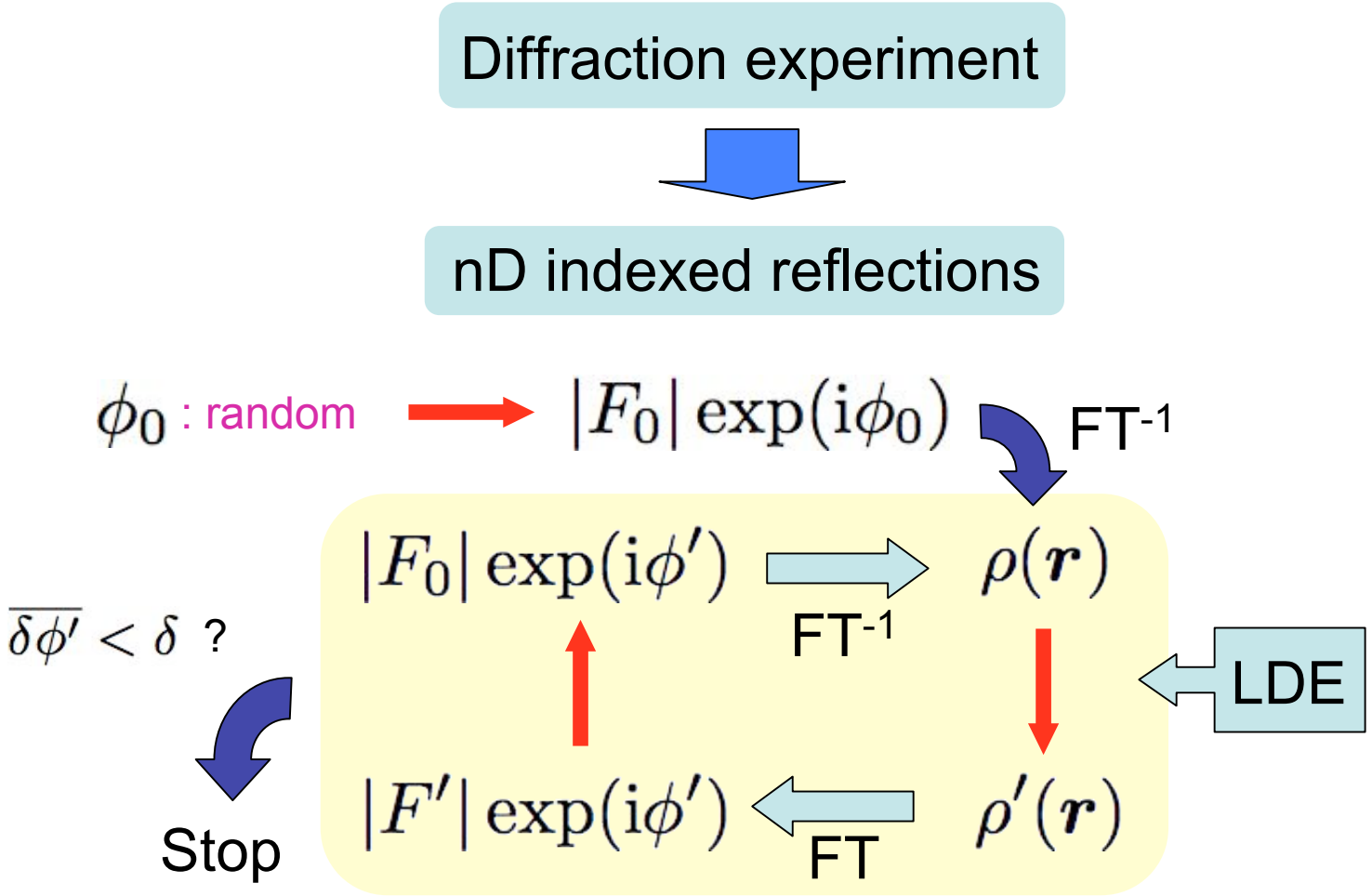
is the same as in Crystals.

Unavoidable loss of phase information in the diffraction intensity.

$$\begin{cases} F(\boldsymbol{k}) = \int \rho(\boldsymbol{r}) \exp(2\pi i \boldsymbol{k}\cdot\boldsymbol{r})\ \mathrm{d}\boldsymbol{r} \\ \rho(\boldsymbol{r}) = \dfrac{1}{V}\int F(\boldsymbol{k}) \exp(-2\pi i \boldsymbol{k}\cdot\boldsymbol{r})\ \mathrm{d}\boldsymbol{k} \end{cases}$$

$$F(\boldsymbol{k}) = |F| \exp(\mathrm{i}\phi) \qquad I \propto |F|^2$$

(LDE)
Principle of the low density elimination method



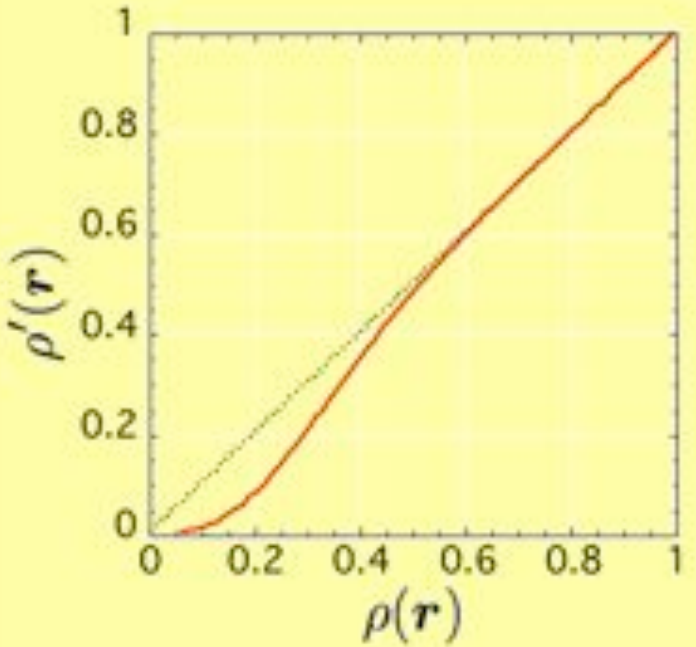
FT: Fourier transform

Density modification function

Density is modified iteratively by

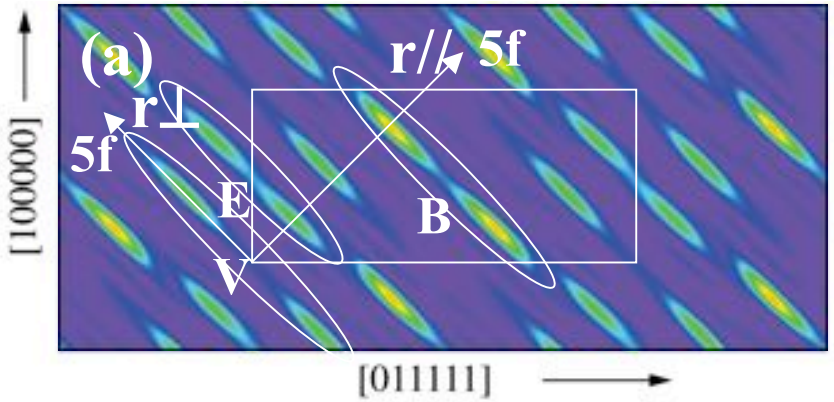
$$\rho'(\mathrm{r}) = \begin{cases} \rho(\mathrm{r})\left\{1 - \exp\left[-\frac{1}{2}\left(\frac{\rho(\mathrm{r})}{0.2\rho_c}\right)^2\right]\right\} & (\rho(\mathrm{r}) \geq 0) \\ 0 & (\rho(\mathrm{r}) < 0) \end{cases}$$

where $\rho(\boldsymbol{r})$ and $\rho'(\boldsymbol{r})$ are the density before and after the modification at a position $\boldsymbol{r}$, respectively. The ρ_c is the average density in unit cell. This function removes negative density and diminishes ripples.



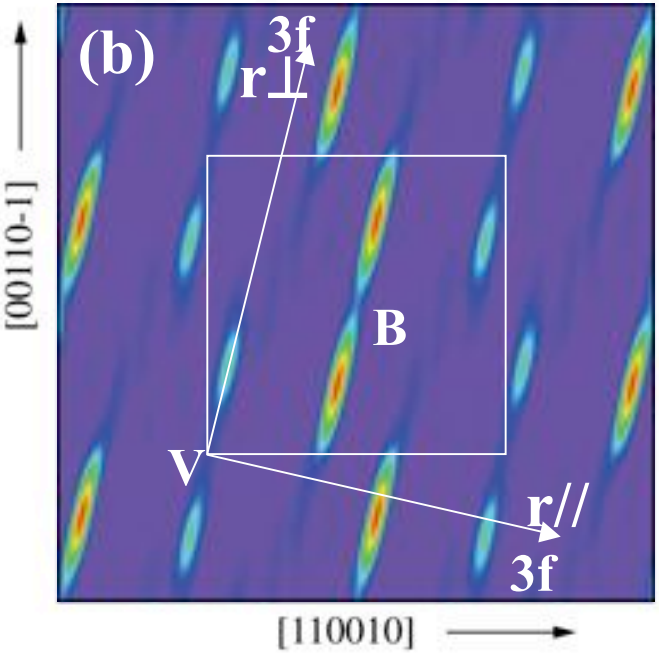
Low Density Elimination (LDE)

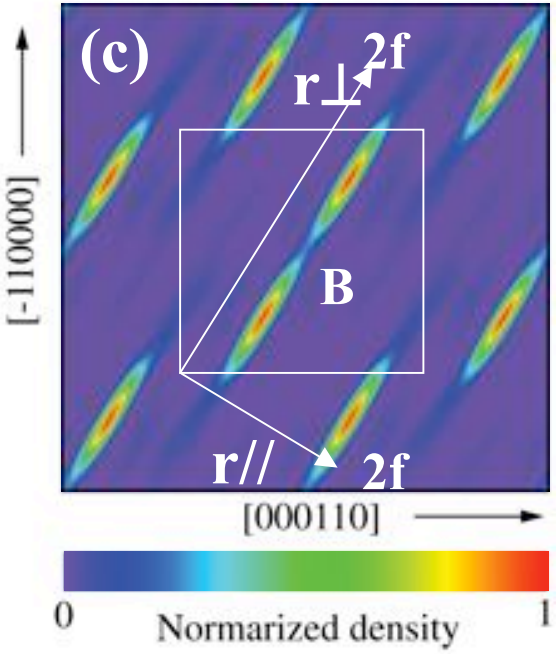
Reconstructed densities of P-type i-Zn-Mg-Ho



space group $Pm\bar{3}5$

V: (0,0,0,0,0,0)
B: (1,1,1,1,1,1)/2
E: (1,0,0,0,0,0)/2
***a* = 0.5137 nm**



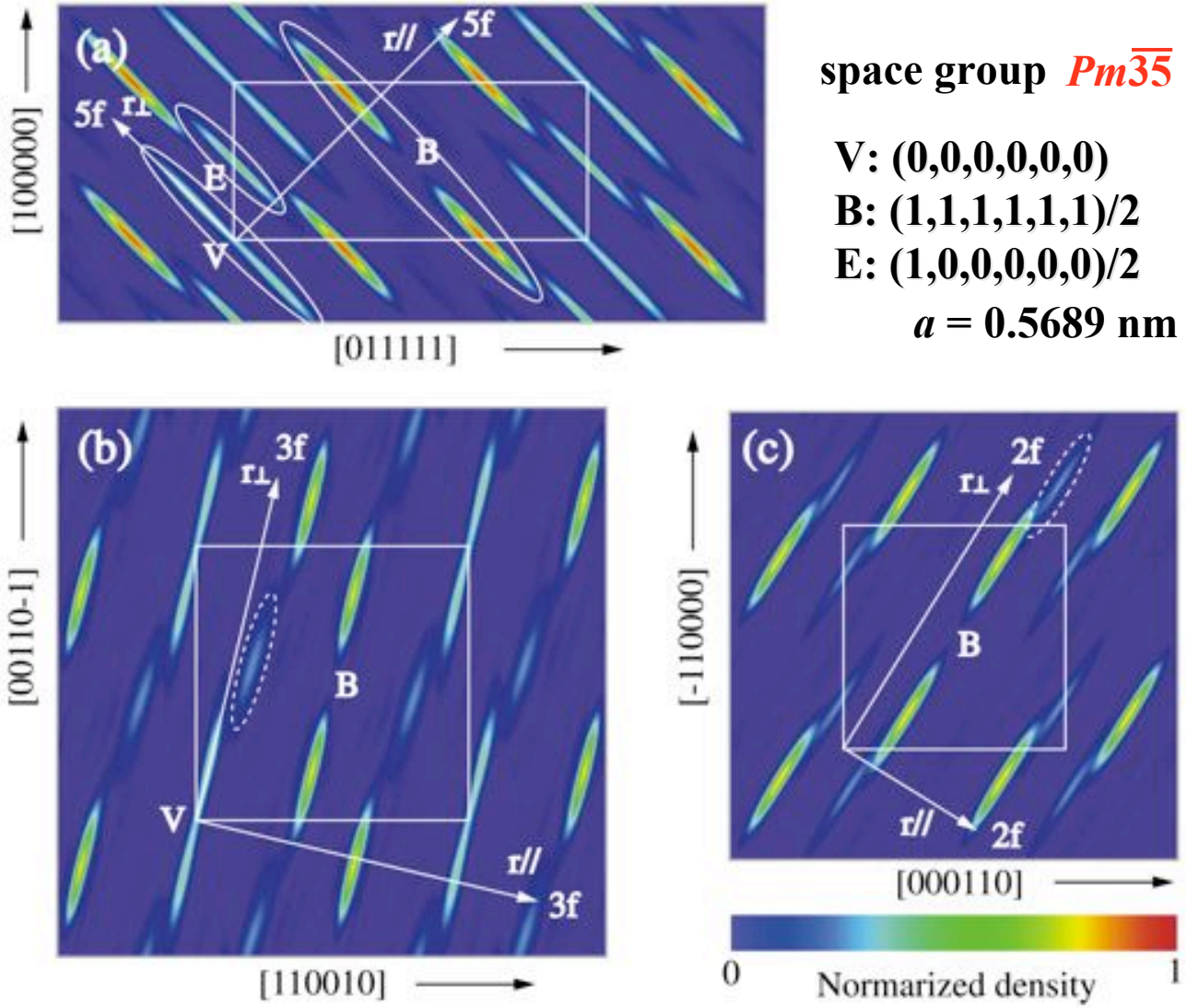


Feature of densities in P-type i-Zn-Mg-Ho

- Similar to the simple decoration model of 3DPT
- Suggests that the position of atoms is similar to that derived from the simple decoration model of 3DPT
- Corresponding approximant crystals consists of Burgman type clusters

⟶ Cluster based model

Reconstructed densities of i-Yb-Cd



space group $Pm\bar{3}5$

V: (0,0,0,0,0,0)
B: (1,1,1,1,1,1)/2
E: (1,0,0,0,0,0)/2
***a* = 0.5689 nm**

Feature of densities in i-Yb-Cd

- There are small OD's shifted along the external space
- Suggests that the position of atoms is not so similar to that derived from the simple decoration model of 3DPT
- Corresponding approximant crystals consists of Tsai type clusters

⟶ Cluster based model

Structure determination of i-YbCd