Grainspotter: overview, indexing unknown polycrystalline compounds

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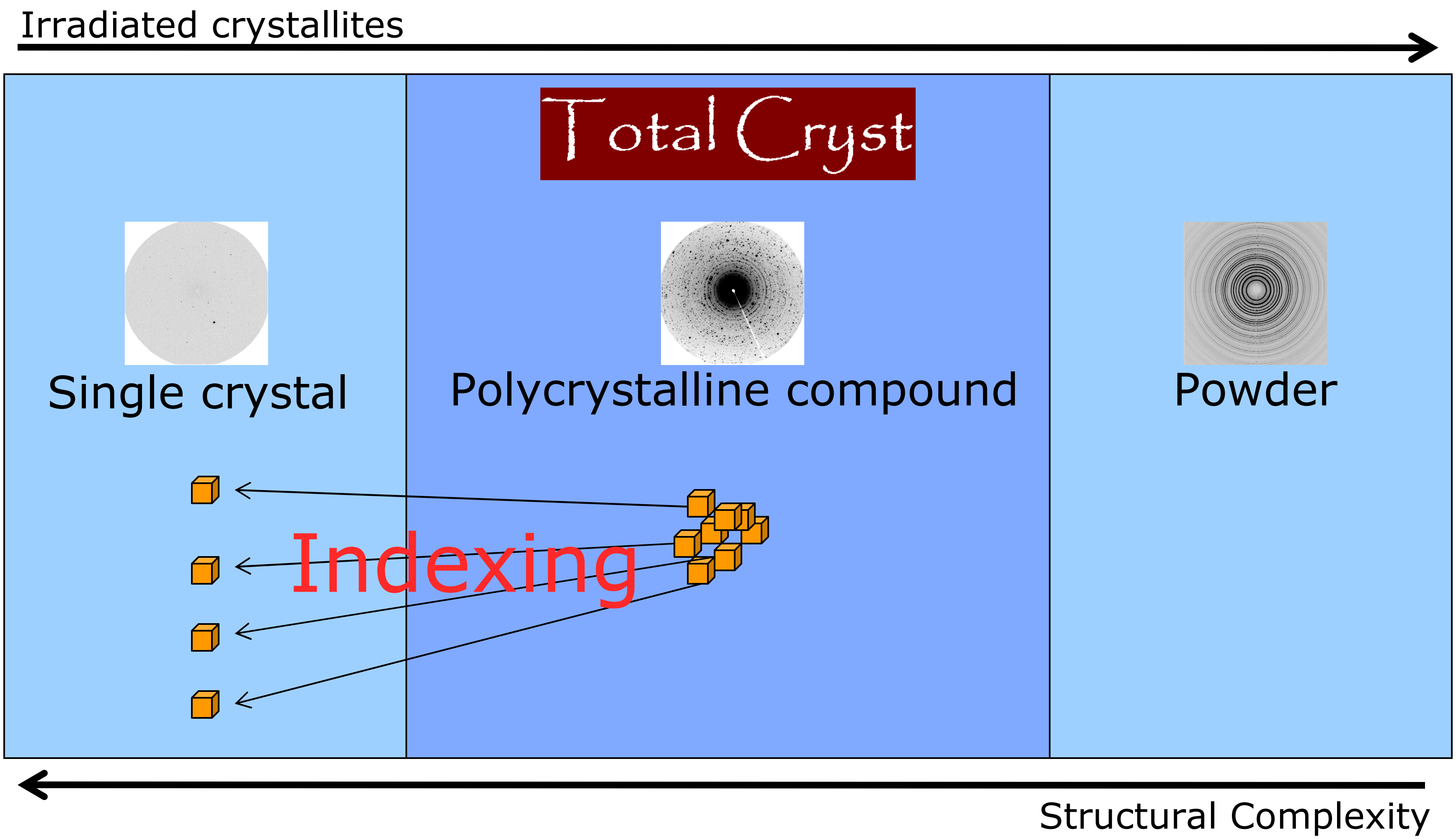
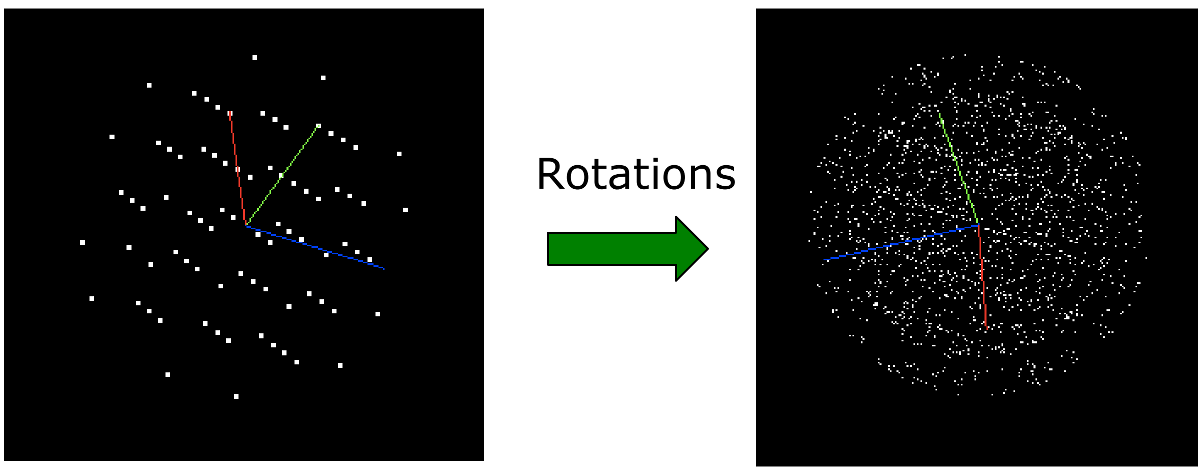
Introduction



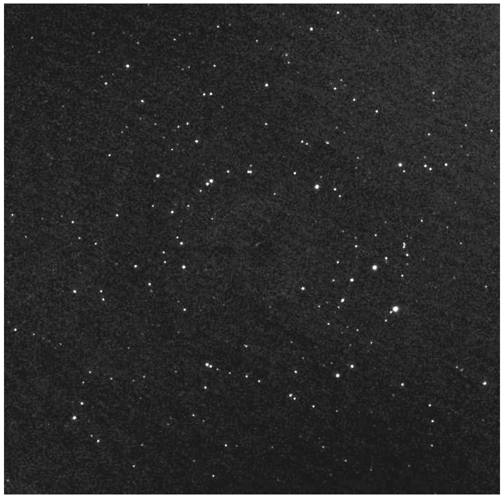
Indexing polycrystalline compound with known crystallography (extracting orientations)

- Bravias Lattice and Unit cell parameters are known a priori from, typically,
  - Powders, radial spectra (Powder Indexing Programs)
  - Single crystal or few crystallites (single crystal indexing program)

Indexing with known crystallography: Identifying copies *of the set of theoretical reflections* in the polycrystalline dataset.



Treatment of data



Reciprocal space

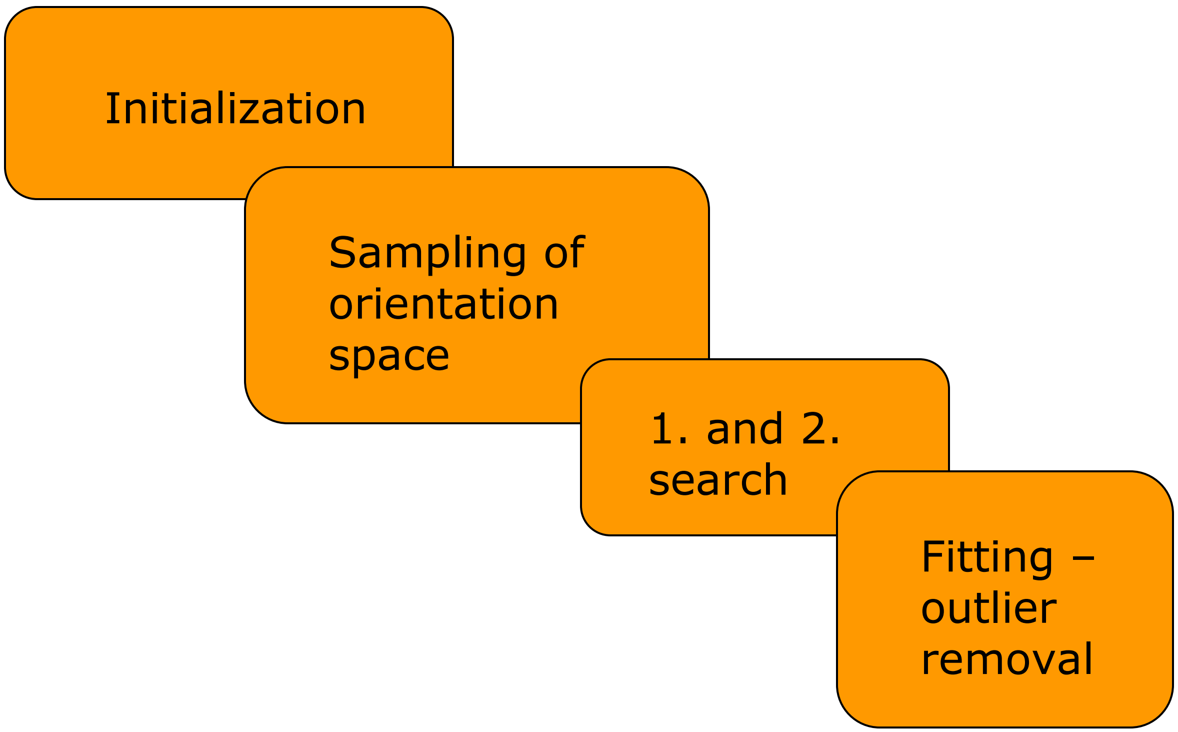
(g-vectors)

ImageD11 (incl. indexing)

Structural solution,
Refinement

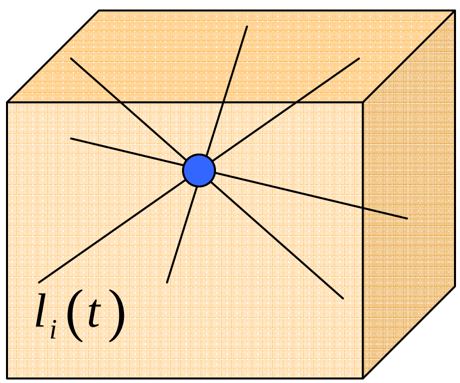
GrainSpotter

GrainSpotter, overview

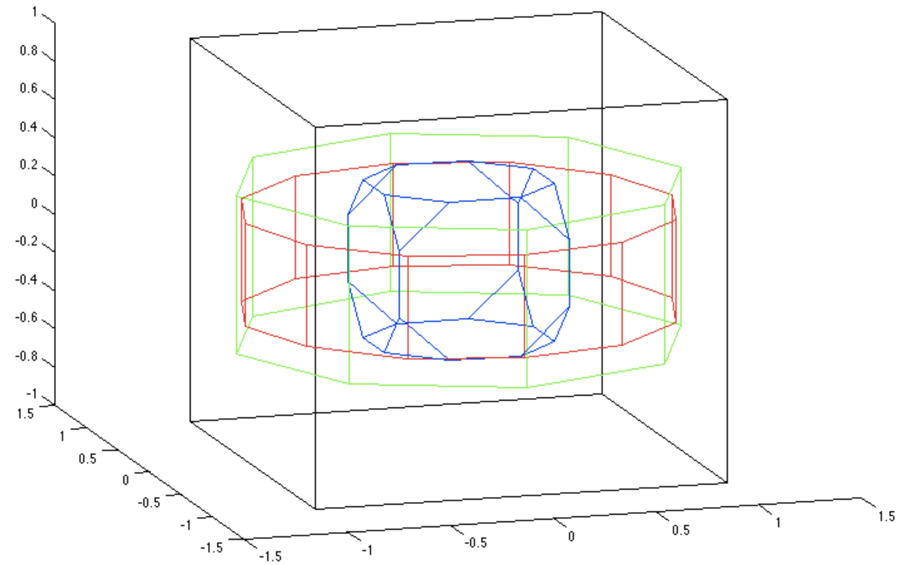


Rodrigues space

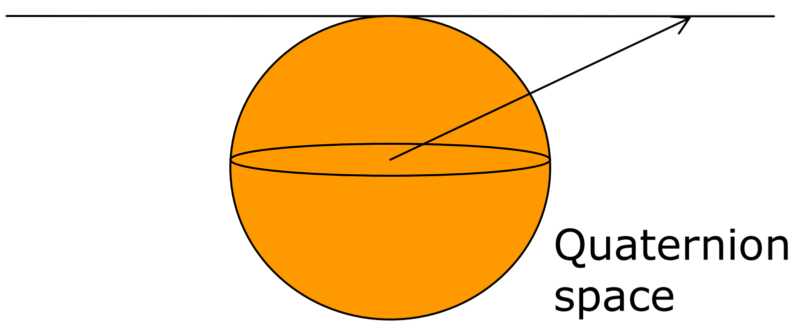
$$\bar{g}_i = U\bar{h}_i \qquad l_i(t) = \frac{\bar{g}_i \times \bar{h}_i}{1+\bar{g}_i \cdot \bar{h}_i} + t\,\frac{\bar{g}_i + \bar{h}_i}{1+\bar{g}_i \cdot \bar{h}_i}$$



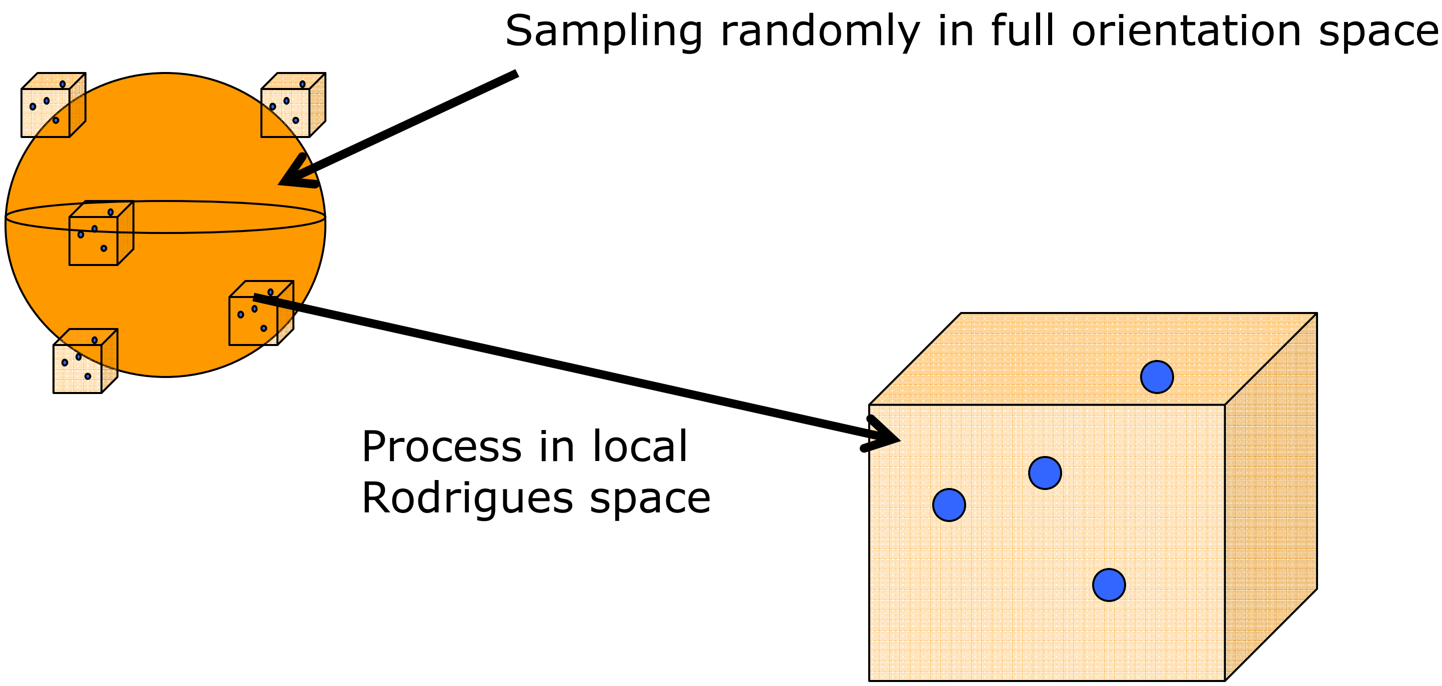
Rodrigues space



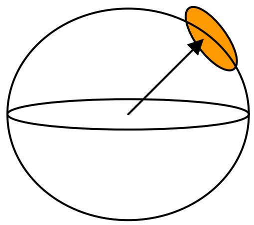
Limitation in orientation:



Searching for all orientations in the local Rodrigues space



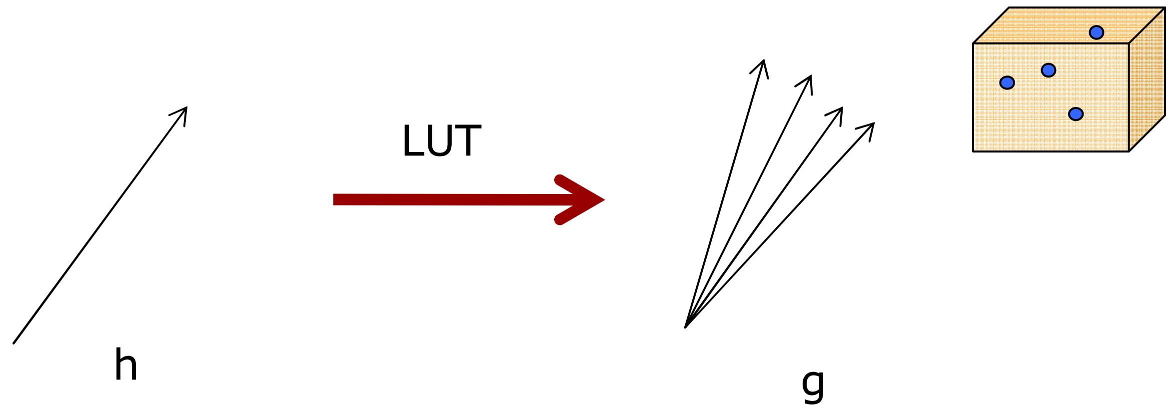
G-vector selection, Lookup table



Each hkl family has a lookup table of 64 by 64 by 64 entries.

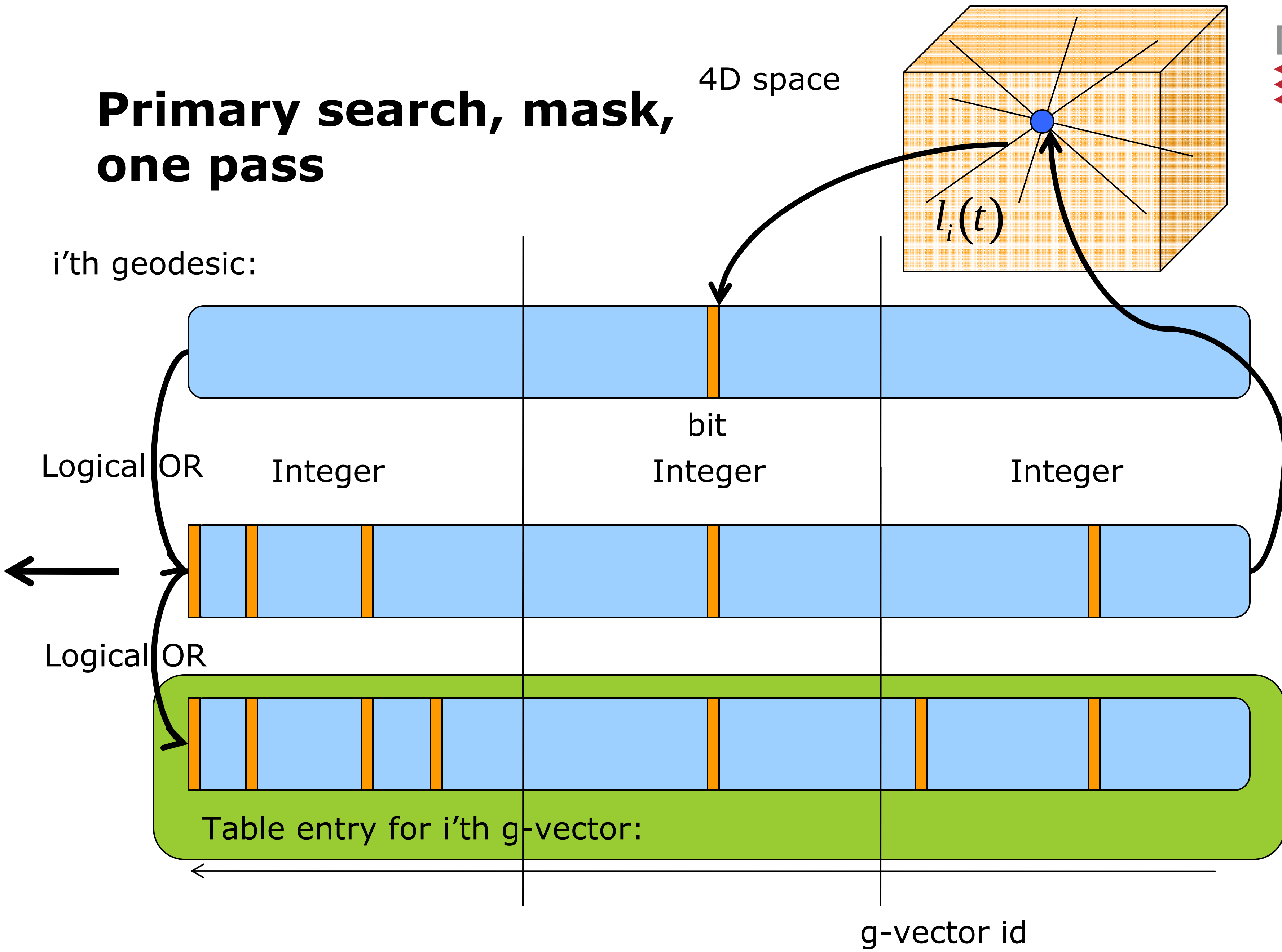
Only the entries near the surface of the sphere are filled with labels linking back to the g-vector list.

Based on the conditions each g-vector is stored in a larger region on the surface of the sphere.

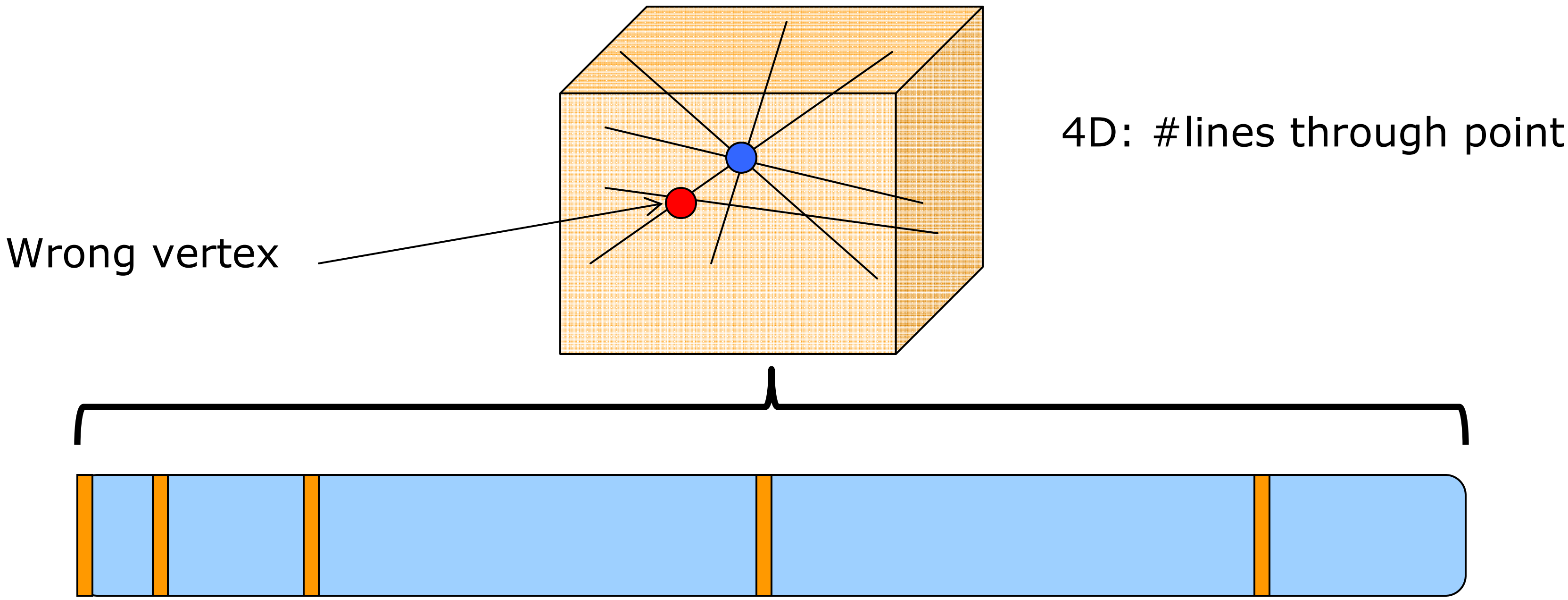


Advantage: Matrix-vector integer multiplication points directly to the right location in the LUT.

Primary search, mask, one pass



Secondary search, integration, one pass



One table entry
per g-vector

Finding real vertices.
Keeping vertices with
hits larger than user
specified minimum

Fitting orientation and position

$$\chi^2(\bar{x}_0) = \sum_i \left[|\overline{\Delta x_i}|^2 - \left(\bar{L}_i \cdot \overline{\Delta x_i} \right)^2 \right], \qquad (11)$$

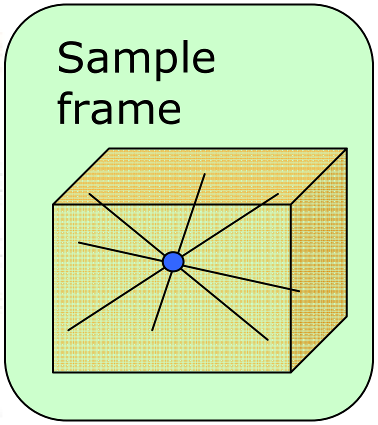
where $\overline{\Delta x_i} = \bar{x}_{l_{rot},i} - \bar{x}_0$ is the difference vector to the diffraction spot $\bar{x}_{l_{rot},i}$ *in the sample reference system*, i.e. $\bar{x}_{rot,i} = \bar{x}_{l,i}\Gamma_i^{-1}$, and the direction of the ray, $\bar{L}_i$, is given by Eq(6). More specifically,

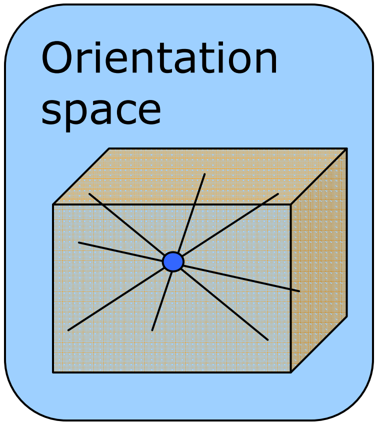
$$\bar{x}_0 = S^{-1}\bar{b}, \qquad (12)$$

where

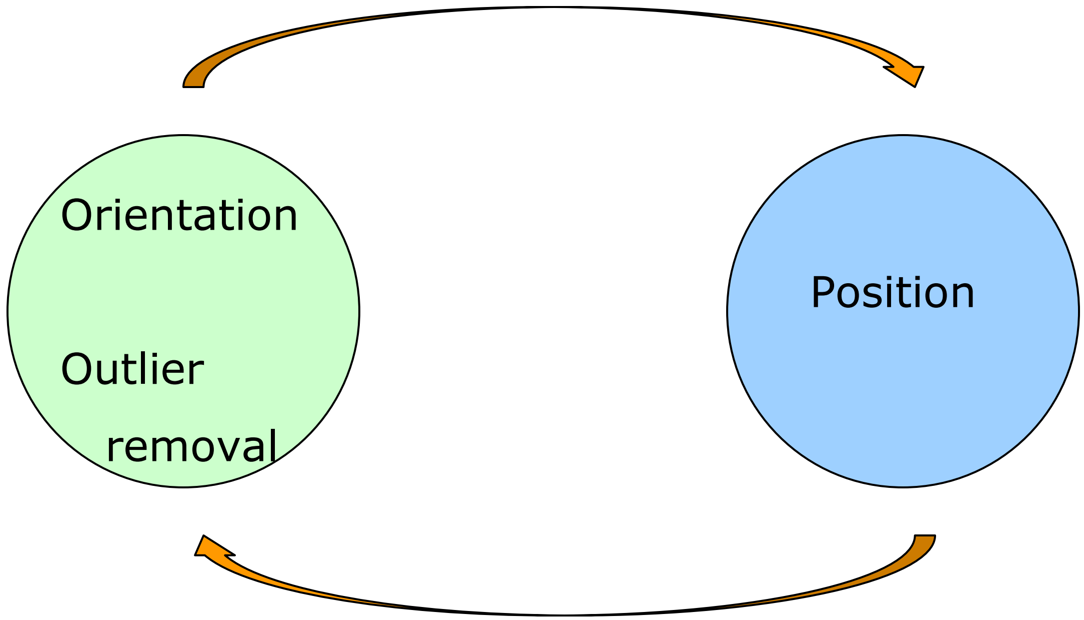
$$S_{pq} = \sum_i \left[\bar{L}_{p,i}\bar{L}_{q,i} - \delta_{pq} \right], \quad \bar{b} = \sum_i \left[\bar{L}_i \left(\bar{L}_i \cdot \bar{x}_{l_{rot},i} \right) - \bar{x}_{l_{rot},i} \right]. \qquad (13)$$

Likewise, the orientation $\bar{r}_0$ is fitted by substituting $\bar{x}_{l_{rot},i}$ with the origin of the geodesics, $\bar{r}_o^g$, and $\bar{L}_i$ with the direction of the geodesic, $\bar{r}_s^g$,





Fitting



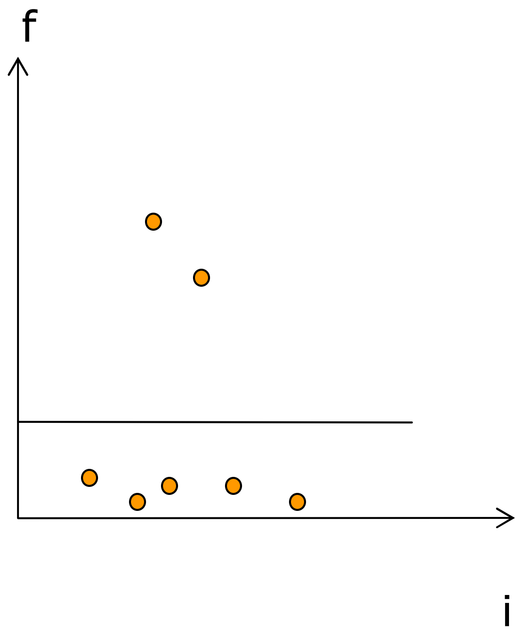
Stop when stable solution is found or if number of measurements goes below a user specified minimum

Outlier removal

$$f_i = \frac{\chi_i^2}{\psi_{max}^2}\frac{\chi_i^2 N}{\chi^2},$$

Absolute   Relative

$f_i > f_{cutoff}$: Discarded



Partial symmetry analysis

- Busing-Levy: $U = MT$

Pair of theoretical g-vectors

Pair of measured g-vectors

- Each space group has a set of equivalent matrices: E_i

$$\tilde{U} = UE_i$$

g-vectors: permutated

When calculating U from:

t1 → h1 and t2 → h2

is this the right orientation giving the highest completeness?

Generally not, since we sometimes have multiple solutions on miller indices for h1 and h2 with **same internal angle**.

$$U_1 = MT_1$$
$$U_2 = MT_2$$

Theoretical g-vector pairs forming T1 and T2 have **same internal angels** (and come from the same hkl families):

$\exists i : T_1 = T_2 E_i$ Not a pseudo twin

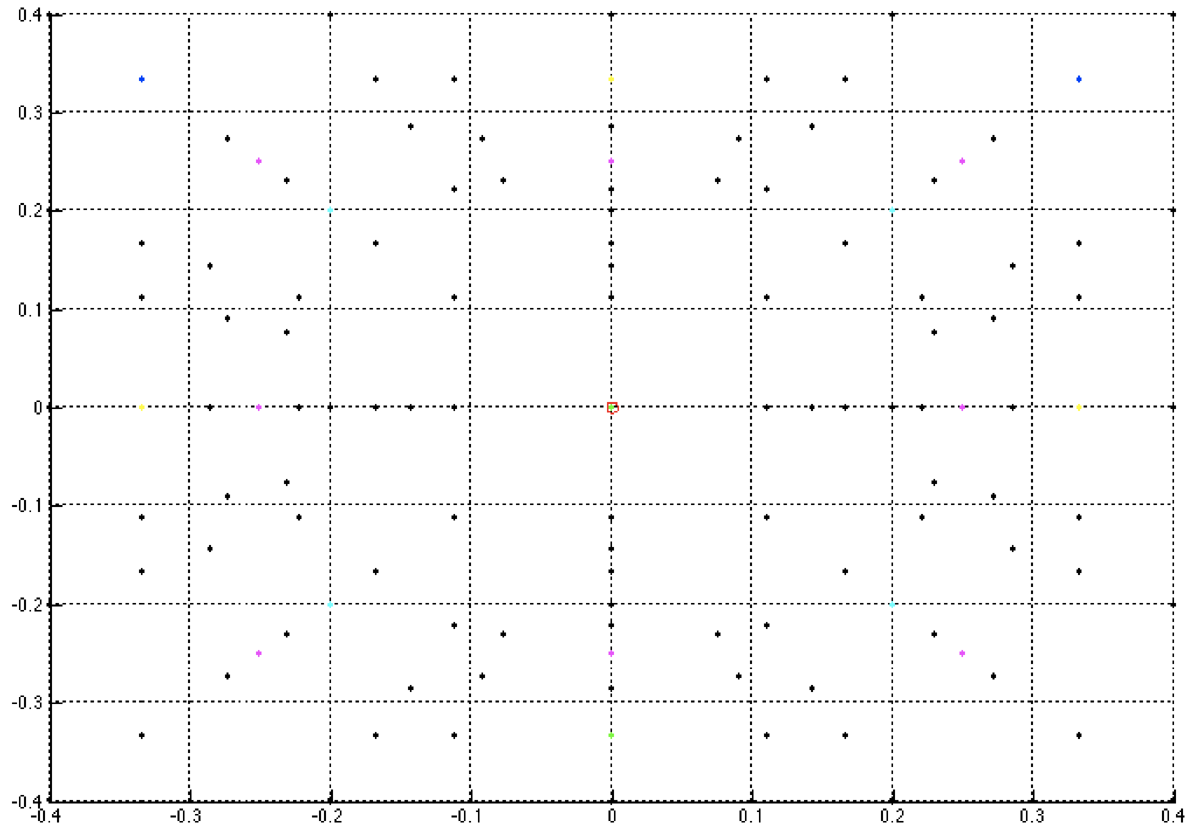
$\forall i : T_1 \neq T_2 E_i$ Pseudo twin

$U = MT_1$ then all $\hat{U} = UT_1^{-1} T_{2i}, \quad \forall i : T_1 \neq T_2 E_i = T_{2i}$ must be tested

Pseudotwins, example, FCC

First 8 hkl-families

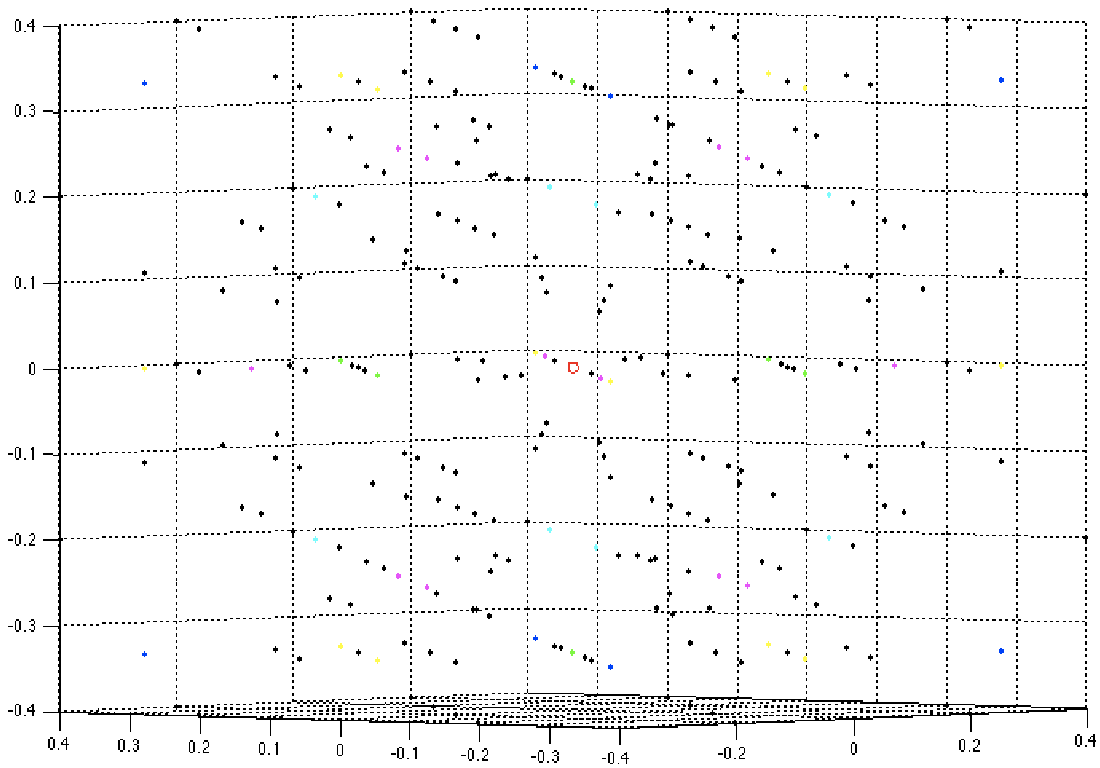
r=0



Red: 112, true
Blue: 34
Green: 24
Cyan: 16
Magenta: 10
Yellow: 8
Black: 2, 4 and 6

Pseudotwins, example, FCC, r=0

$$r_{pseudo} = \frac{p}{q}$$, p and q integers



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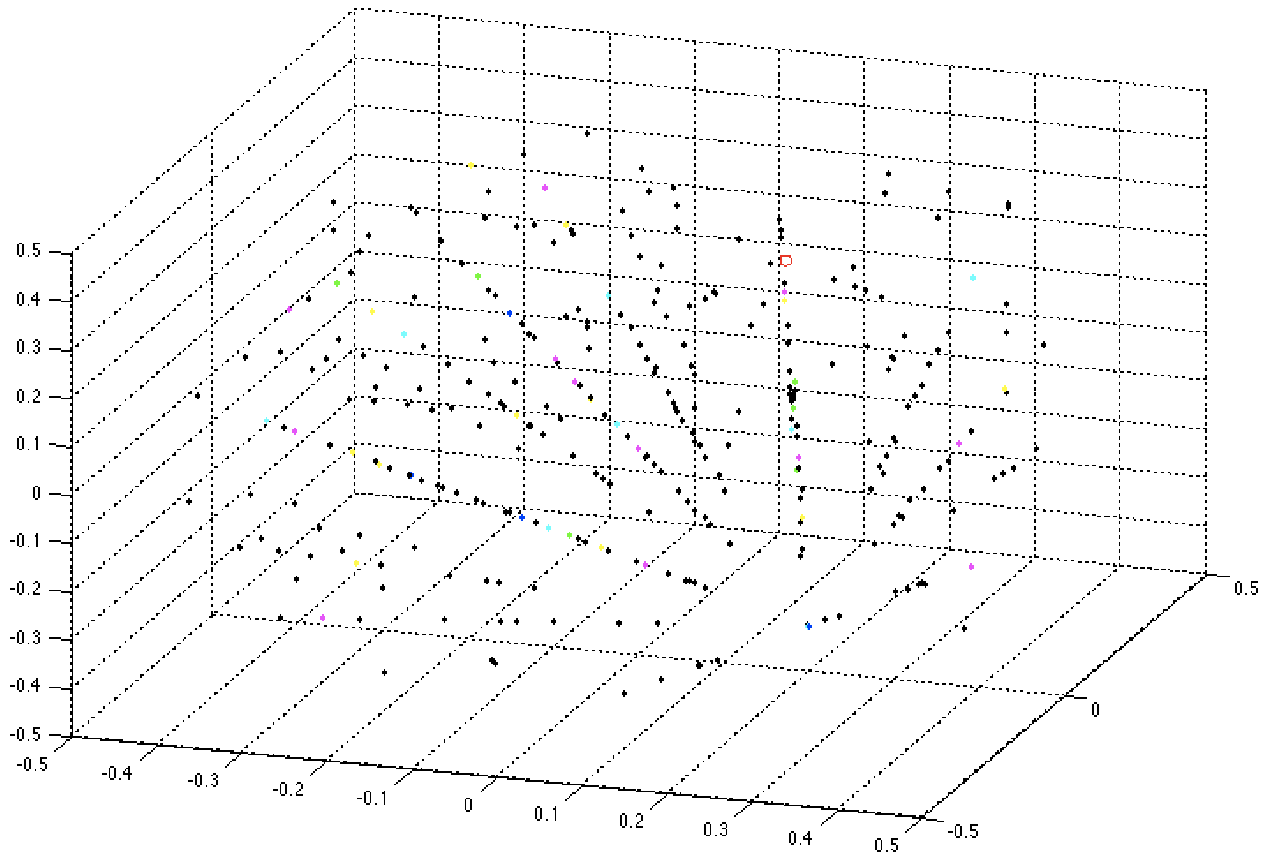
Pseudotwins, FCC, example r.0



Red: 112, true
Blue: 34
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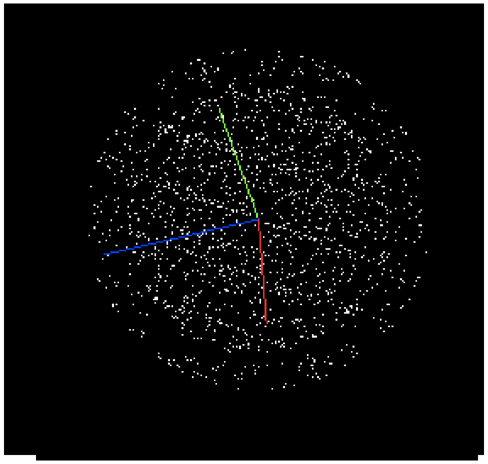
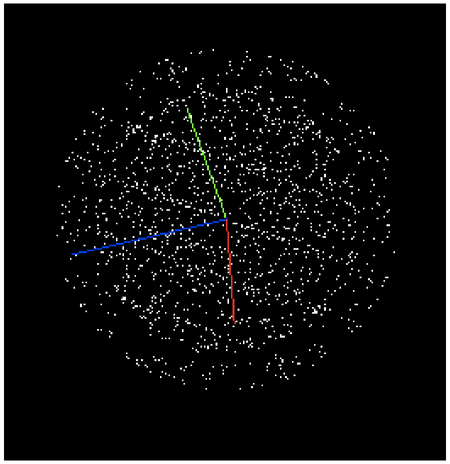
Indexing compounds with unknown crystallography

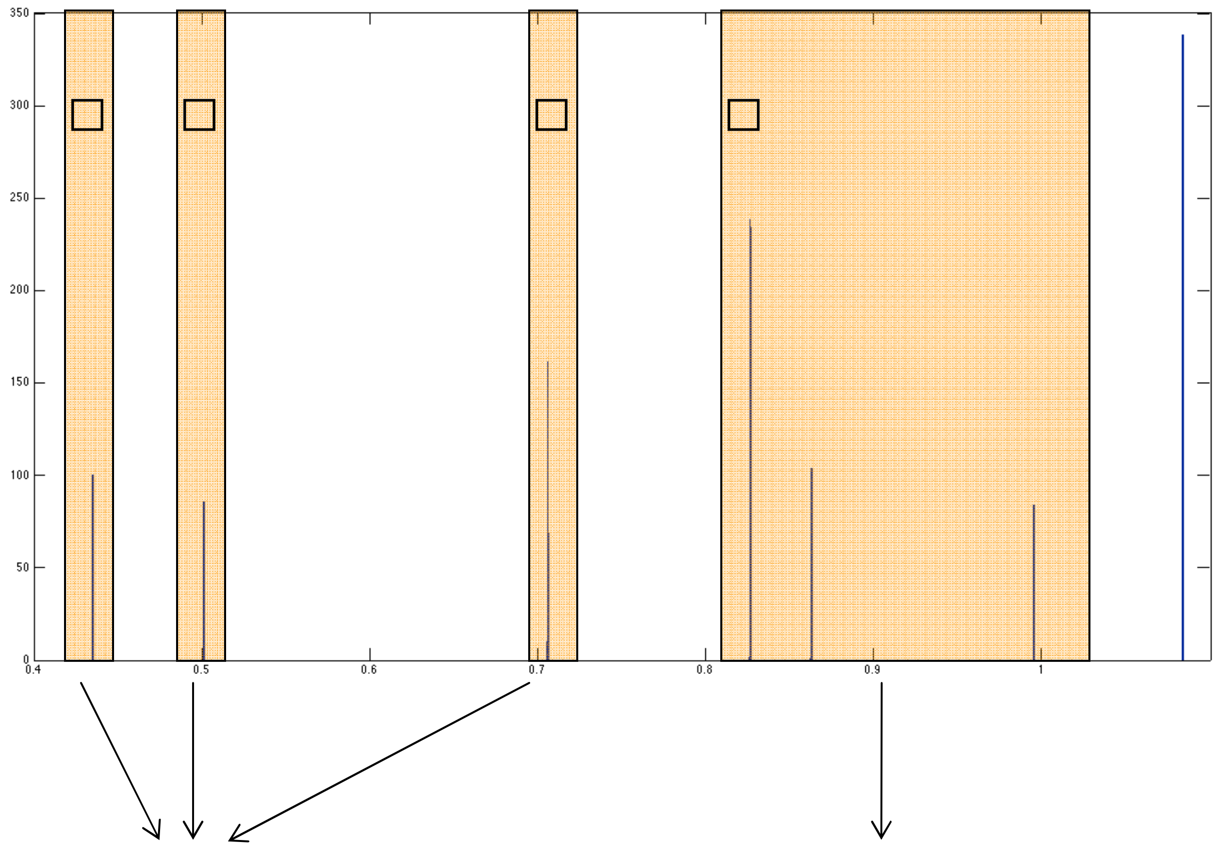


Rotations



Ideally N^2N_{sym} occurrences in orientation space + pseudo twins

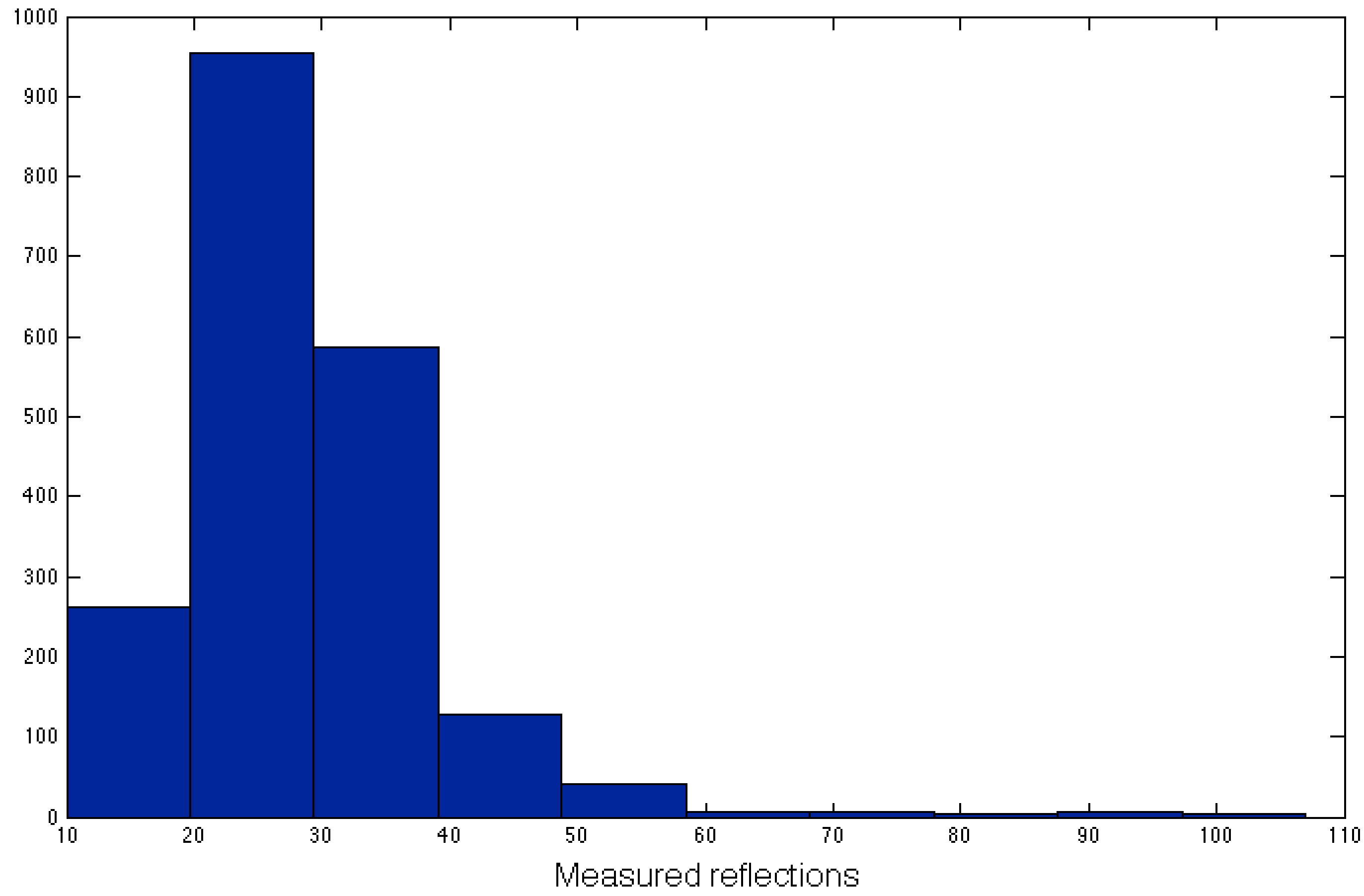
Indexing unknown, pseudo data

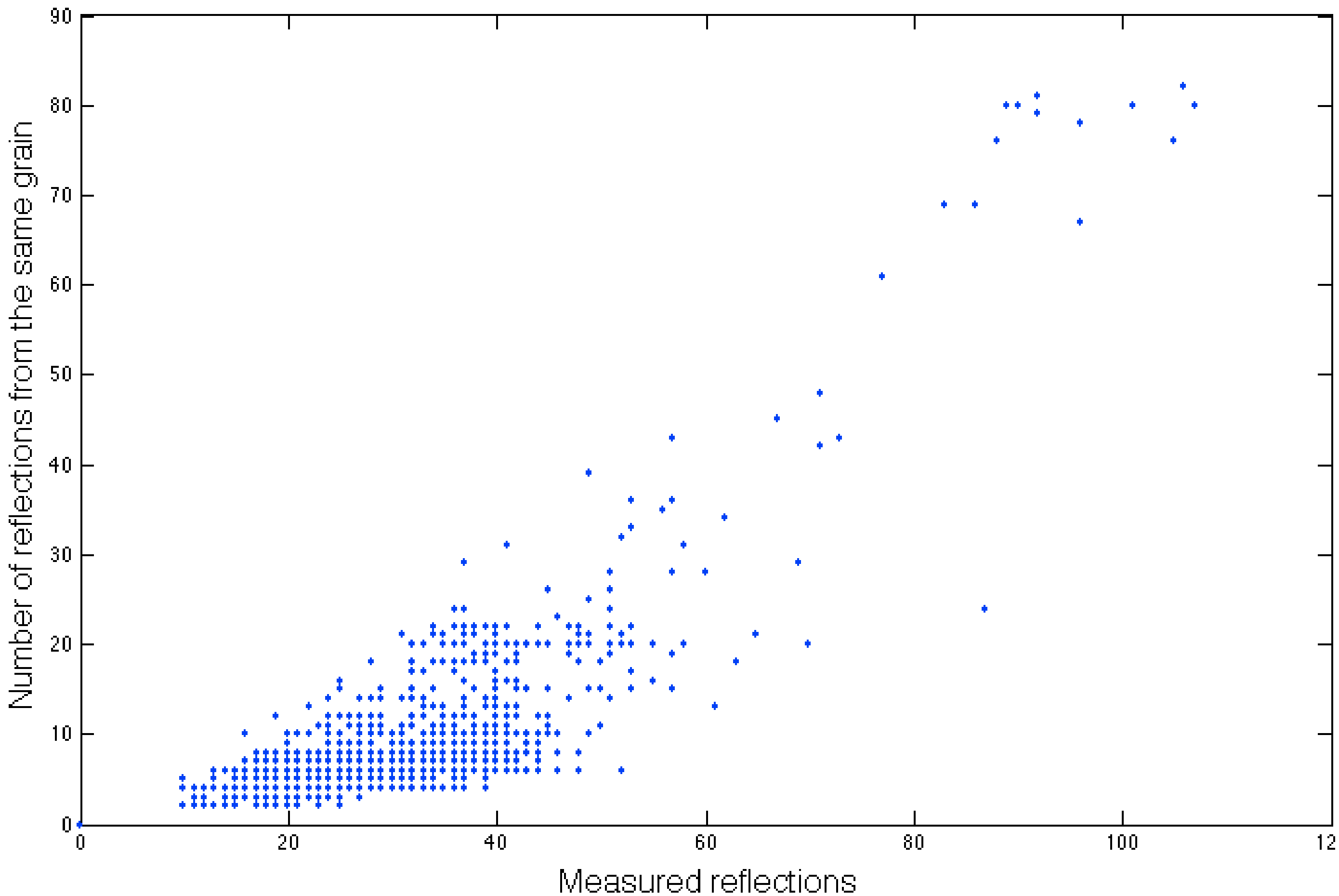


Initial search

Extended search

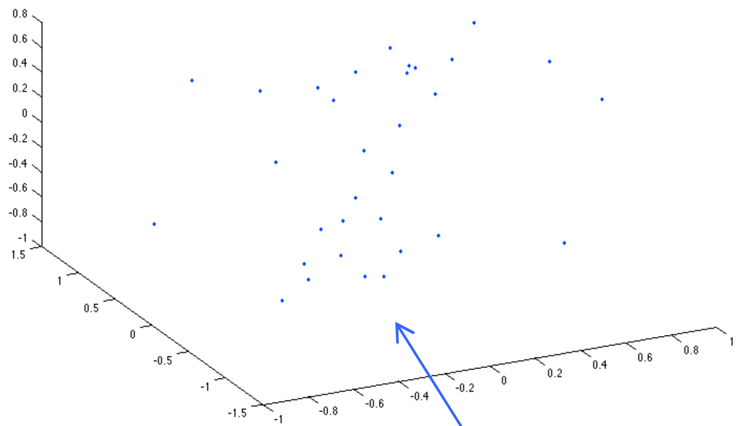
Trying 2000 random (mis)-orientations

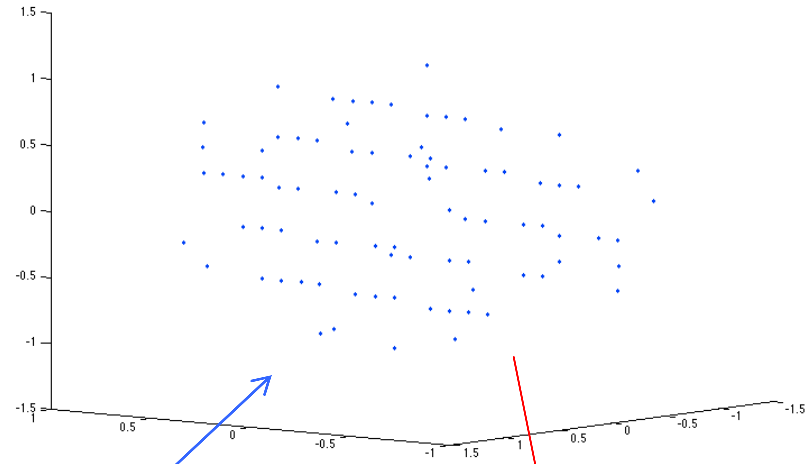


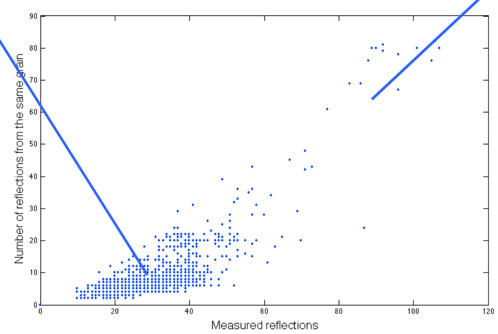


Number of reflections from the same grain
Measured reflections
0
10
20
30
40
50
60
70
80
90
0
20
40
60
80
100
120

Lattice matches





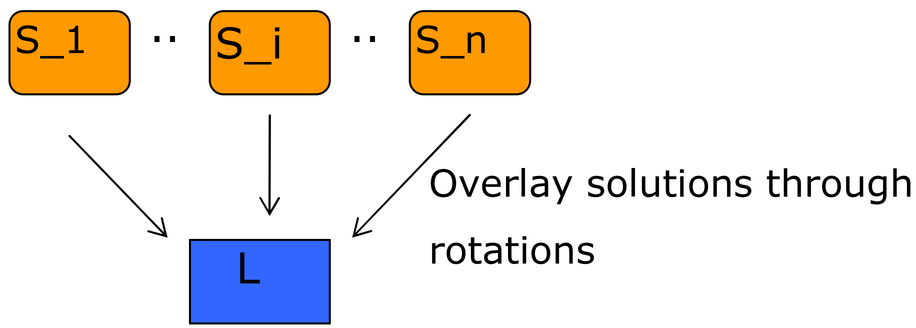


Single crystal indexing program

Additional noise filtering

Keeping the solutions with most points, however still noisy points occur.

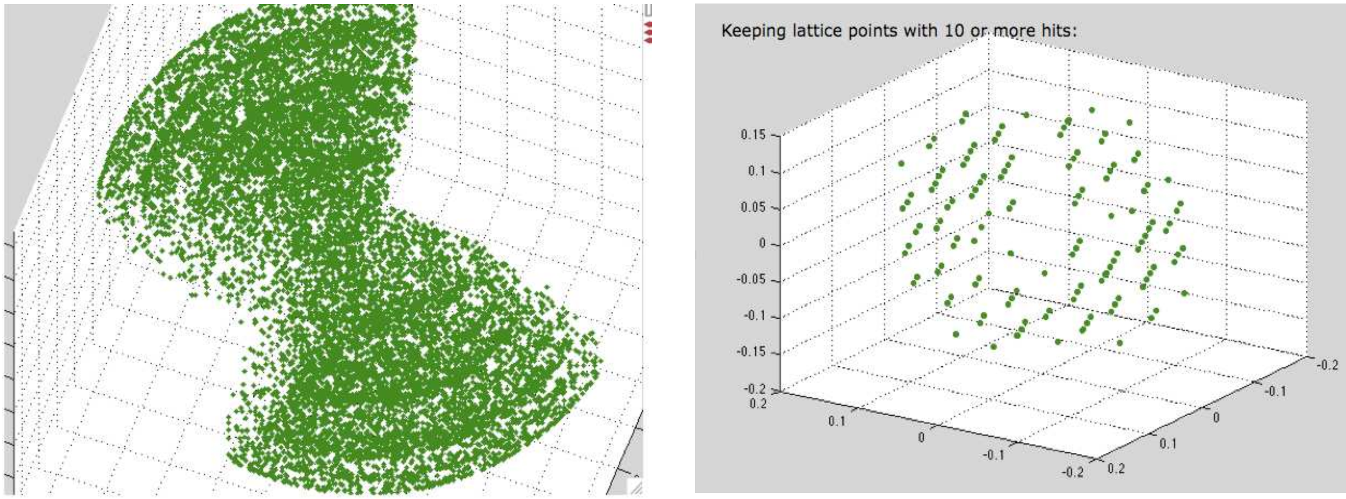
Combine solutions:



Keeping high frequency (lattice) points

Two approaches

A) All combinations of (h1->t1, h2->t2) – computationally heavy

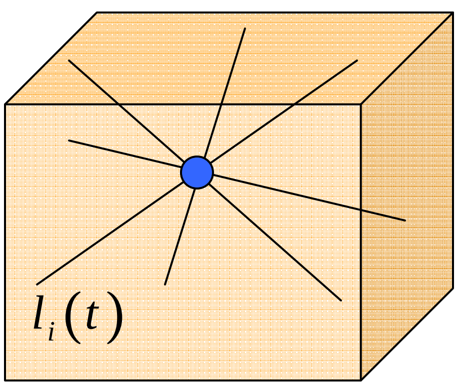


Pseudo data, fcc, 100 grains, Approx 12000 gvectors

Two approaches

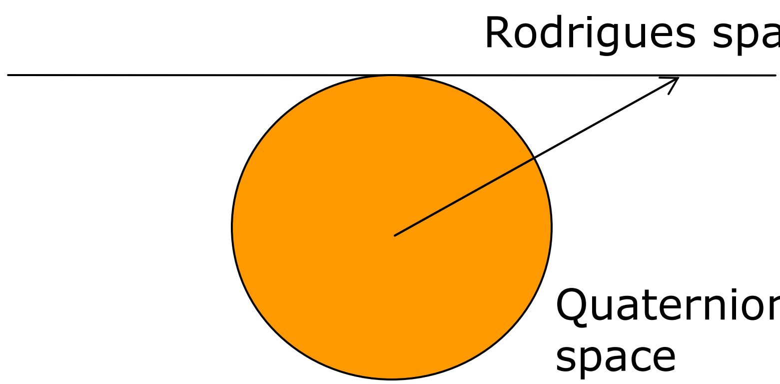
B) Yet again: Using geodesics in orientations space

$$\bar{g}_i = U\bar{h}_i \qquad l_i(t) = \frac{\bar{g}_i \times \bar{h}_i}{1 + \bar{g}_i \cdot \bar{h}_i} + t\,\frac{\bar{g}_i + \bar{h}_i}{1 + \bar{g}_i \cdot \bar{h}_i}$$

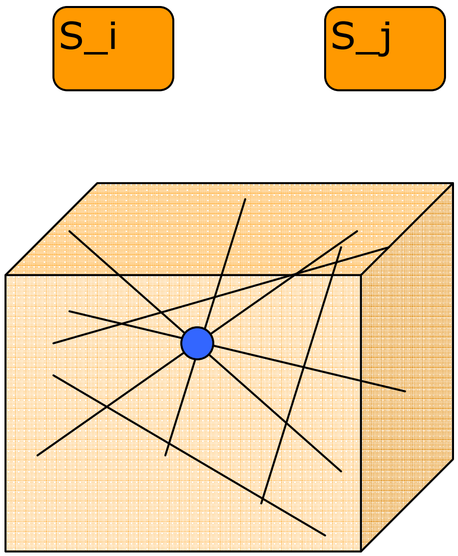


Rodrigues space

Limitation in mis-orientation:



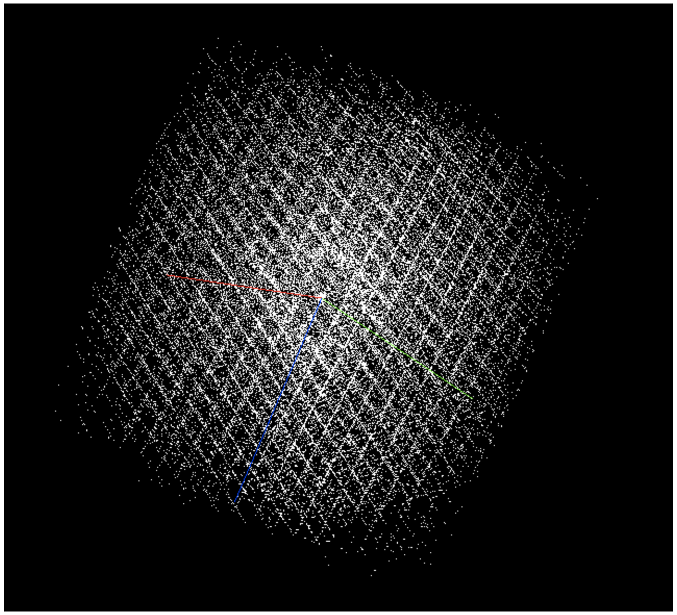
Two approaches

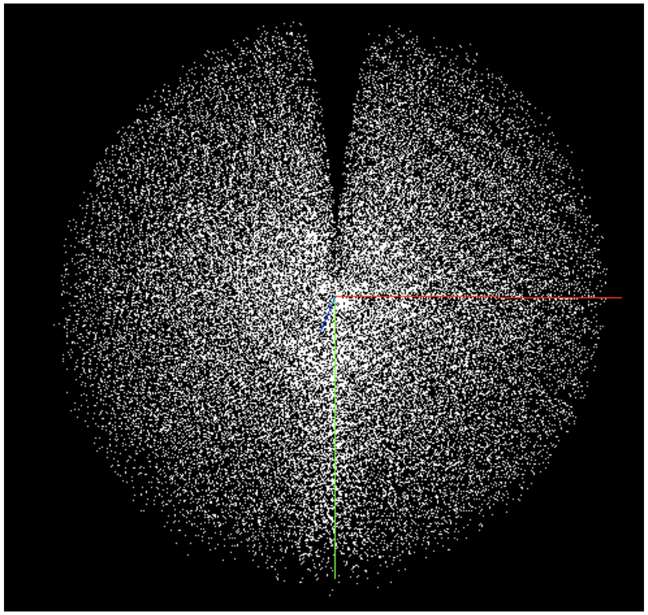


Again match lattice against lattice

Keep track of high frequency lattice points

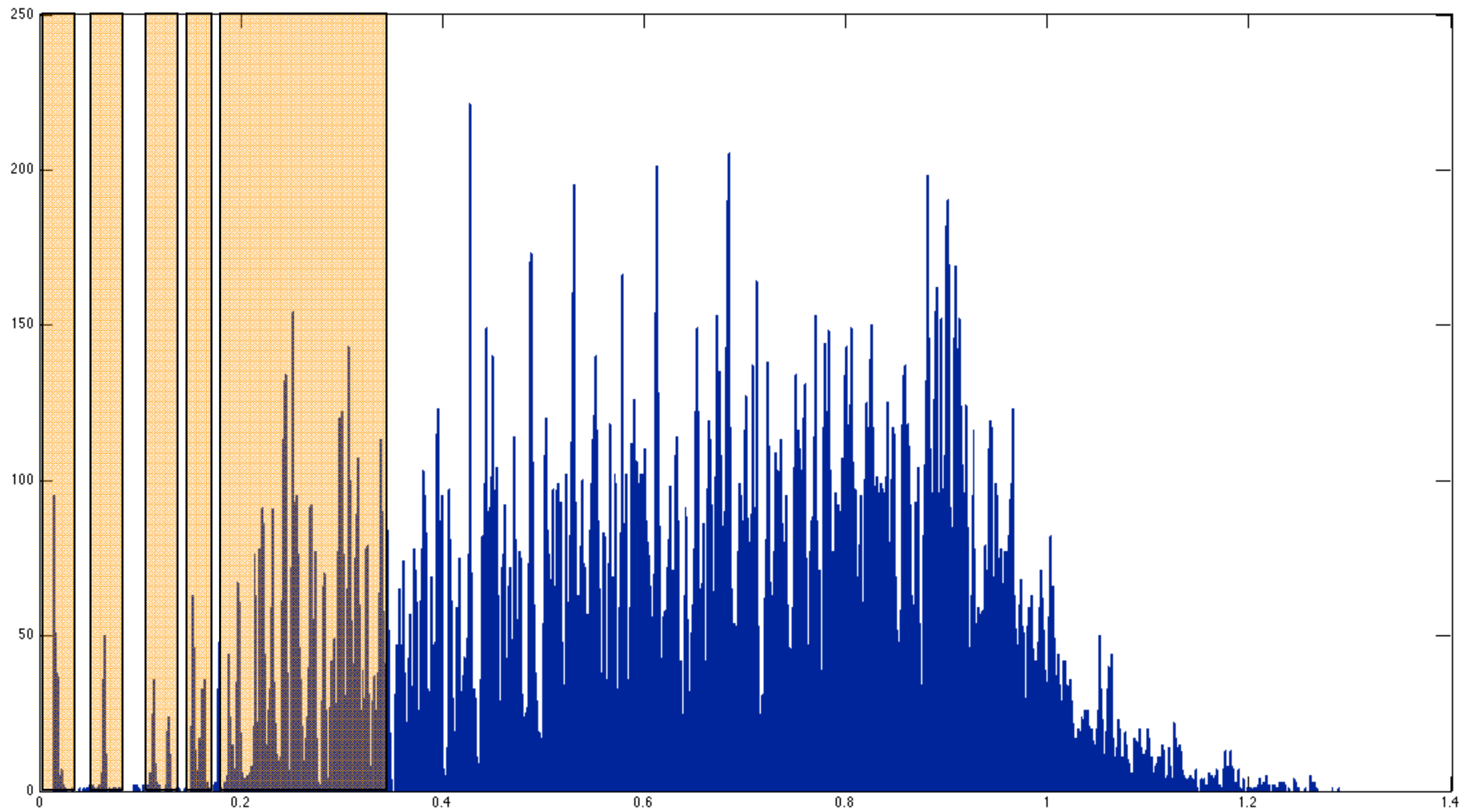
Example, real data





Few grains

Distribution of d-spacings

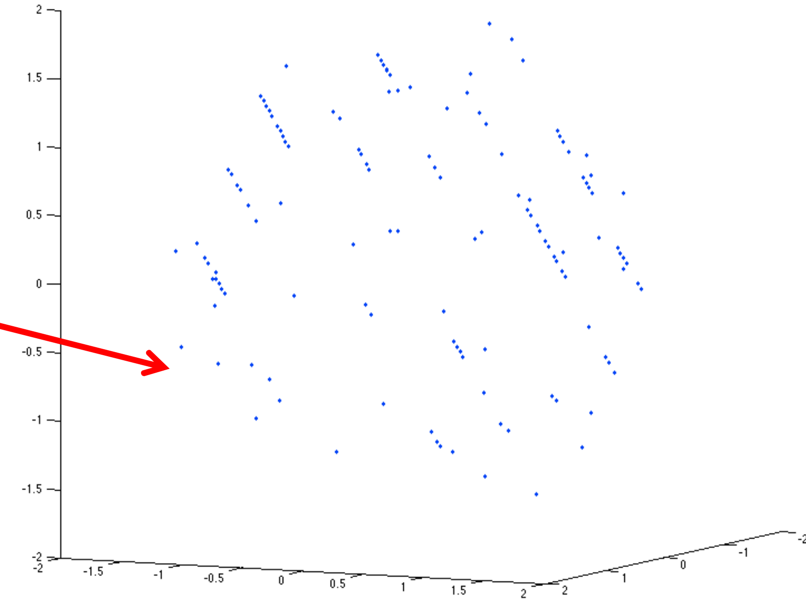


d-spacing ranges used in the search

Solution with most points

10000 random trials

133, 94, 92, 83, 82,…



Dirax: a=8.643 b=10.630 c=31.378 =90.01 =90.24 =90.14

Duisenberg, A.J.M.(1992). J. Appl. Cryst. 25, 92-96

Compound: (bbcp) 2-benzyl-5-benzylidene-cyclopentanone

Jav Davaasambuu et al 2005 J. Phys. D: Appl. Phys. 38 A204-A207.

Orthorhombic, Pbca, Sp gr. 61

12 grains in data

Next

- Noise filtering: Use integrated intensities from diffraction spots
- Simultaneous identification of multiple phases
- Also, index_unknown.py (ImageD11) on lattice candidates instead of Dirax

DTU

Acknowledgement

• J. Wright, H.O. Sørensen, G. Vaughan, C. Gundlach, S. Techert, J. Davaasambuu

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