Quantitative Texture Analysis

D. Chateigner
CRISMAT-ENSICAEN; IUT-UCBN
6 bd. M. Juin 14050 Caen

« classical texture analysis » (Google)

Journée Inauguration plateforme RX
Université Paris-Diderot, 3rd April 2014

Crystallographic texture

Diffraction Measurements
S space, $I(\chi,\varphi,\omega,\eta,2\theta)$

Corrections
(defocusing, localization,
Volume-absorption)

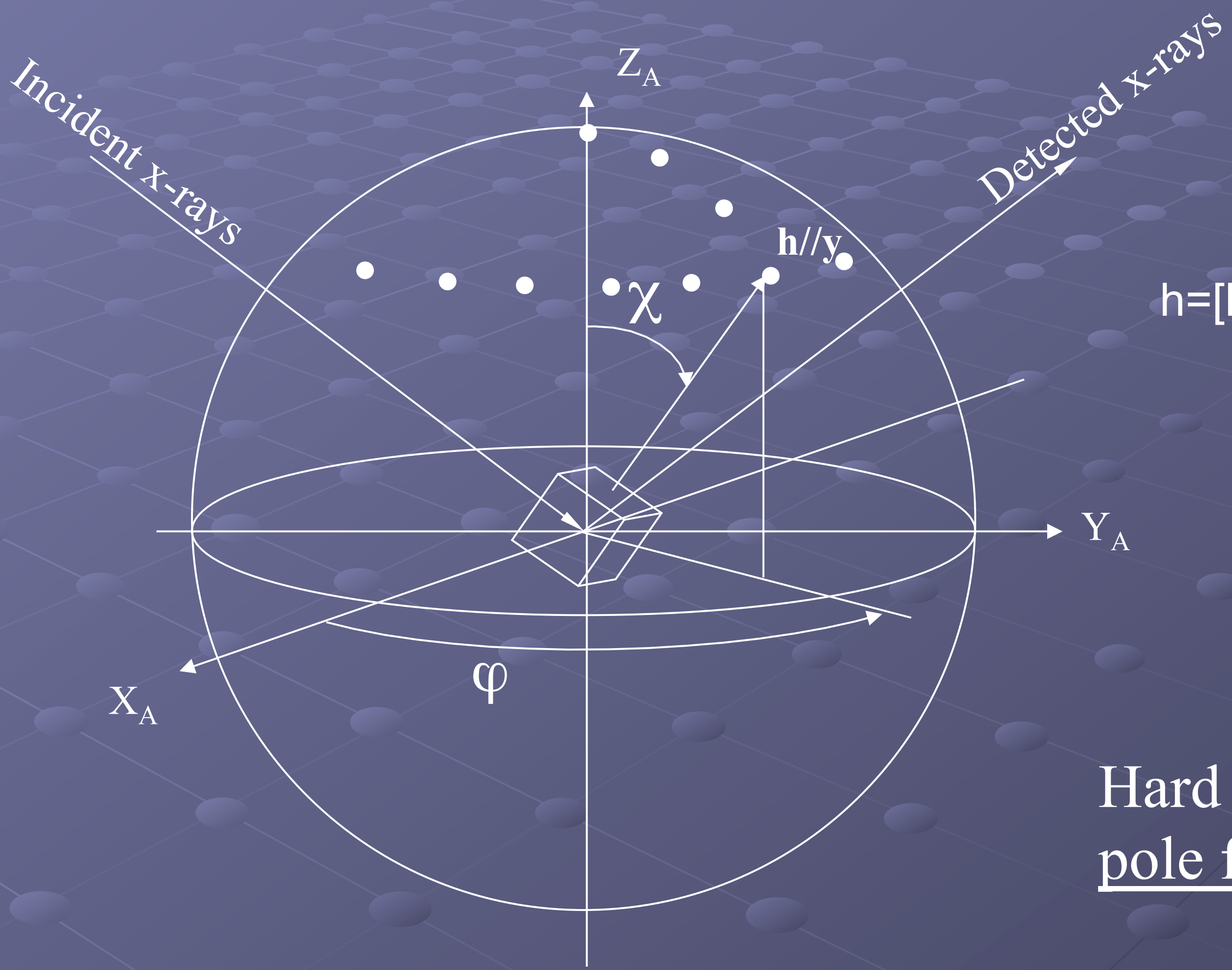
Normalization
$P_h(y)$

Pole figures
Y space, $I_h(y)$

Orientation Distribution Function
G space, $f(g)$

Macroscopic anisotropic properties $(C_{ijkl}, \sigma_{ij}, d_{ijk} \ldots)_M$
Elastic wave velocities (geophysics)
Anisotropic spectroscopies (P-EXAFS, ESR …)
Character analyses (phylogeny, palaeontology)
Art and Cultural Heritage

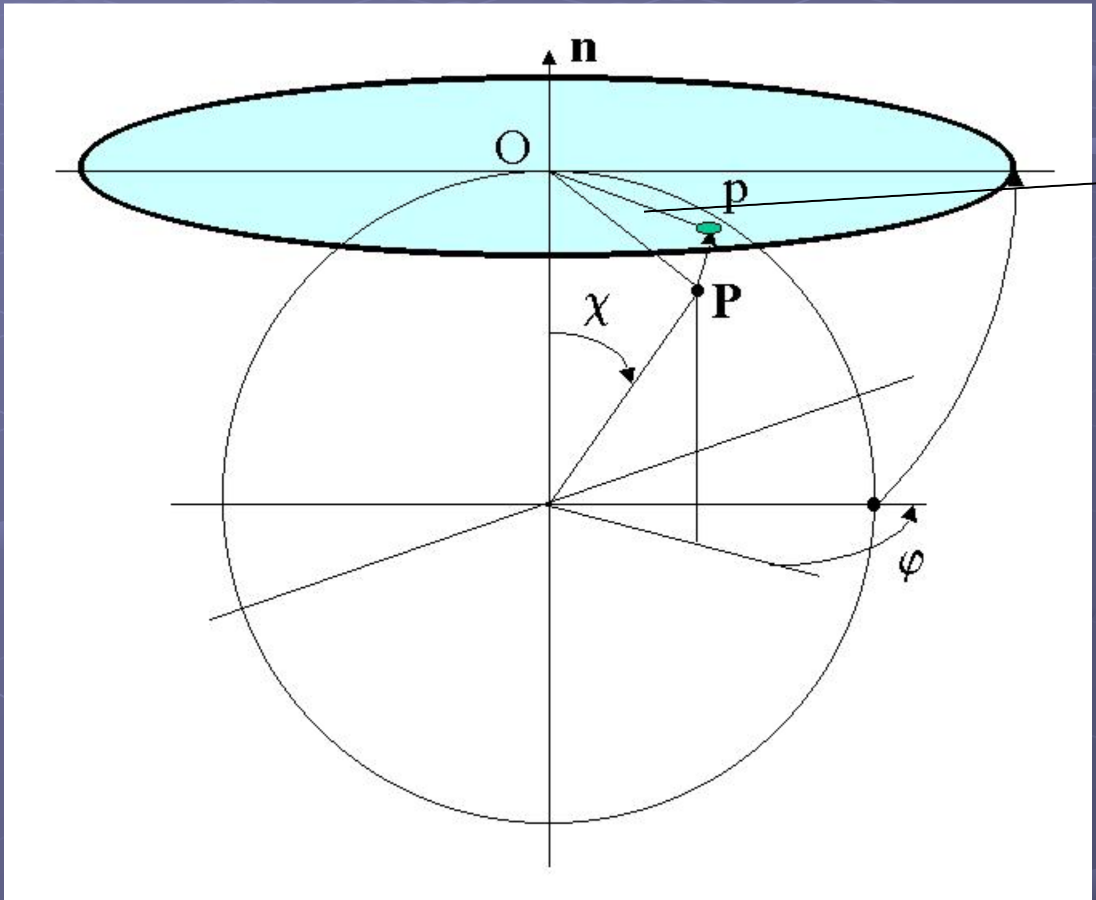
One crystallite oriented in the Pole sphere:



h=[hkl]* // y=(χ,φ,2θ,η,ω)

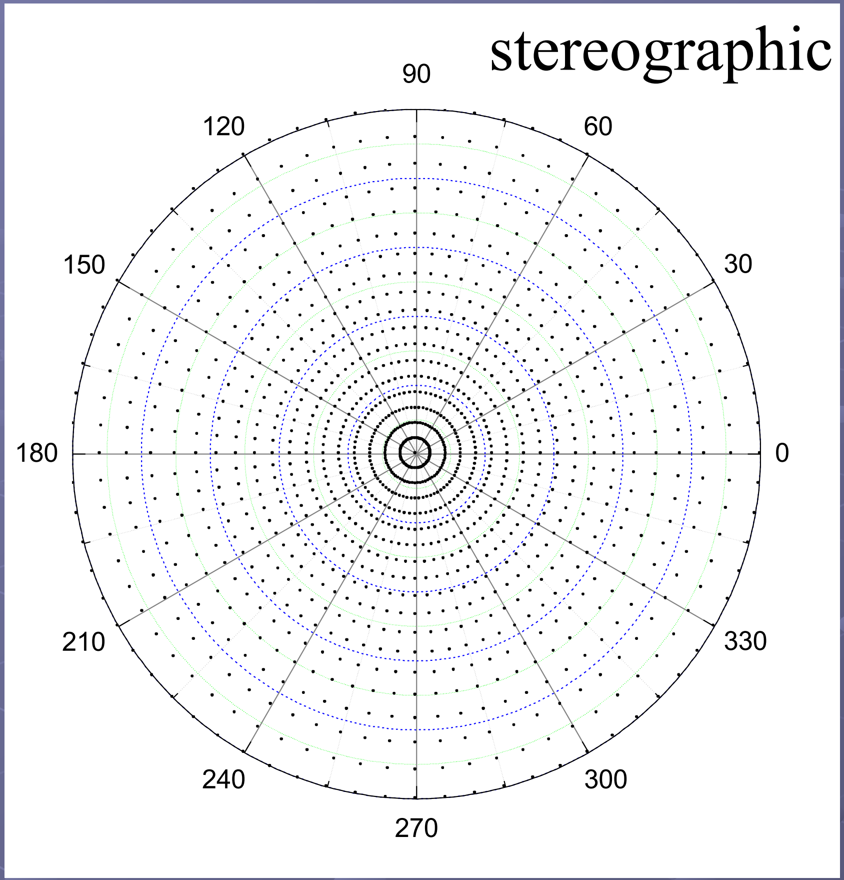
Hard to visualise: needs pole figures

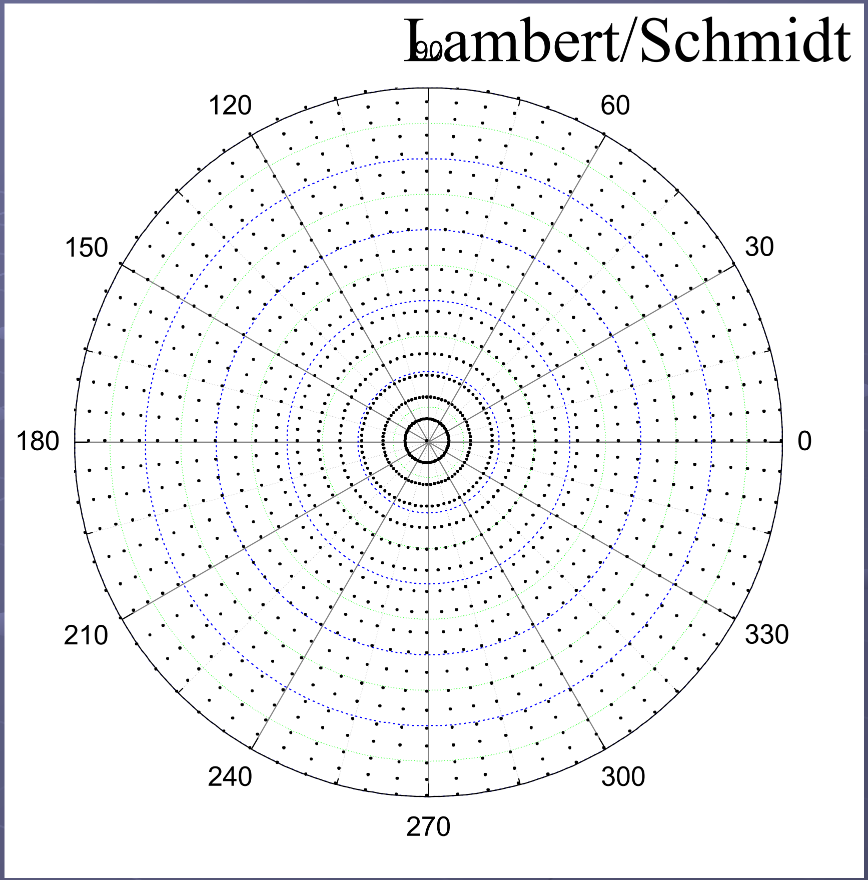
Lambert projections (equal area)



Poles: p(r',φ):

$r' = 2R \sin(\chi/2)$



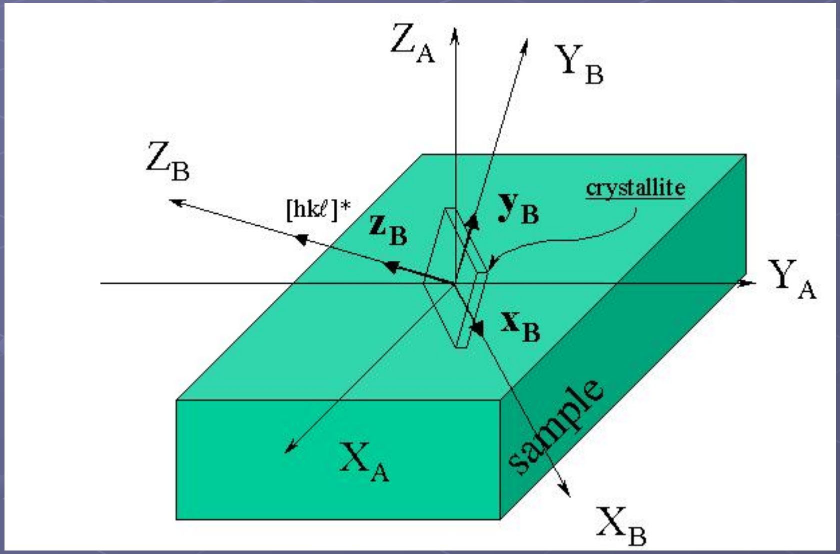


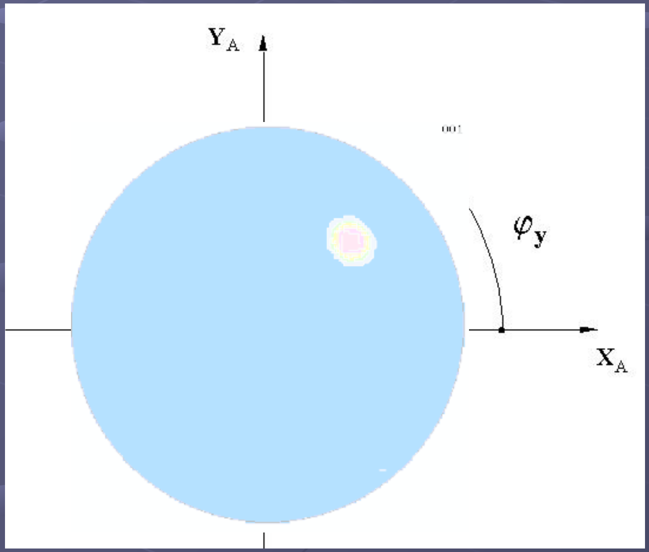
5° x 5° grid: 1368 points

Pole figures

{hkℓ}-Pole figure: location of distribution densities, for the {hkℓ} diffracting plane, defined in the crystallite frame K_B, relative to the sample frame K_A.

Pole figures space: , with **y** = (ϑ_y,φ_y) = [hkℓ]*

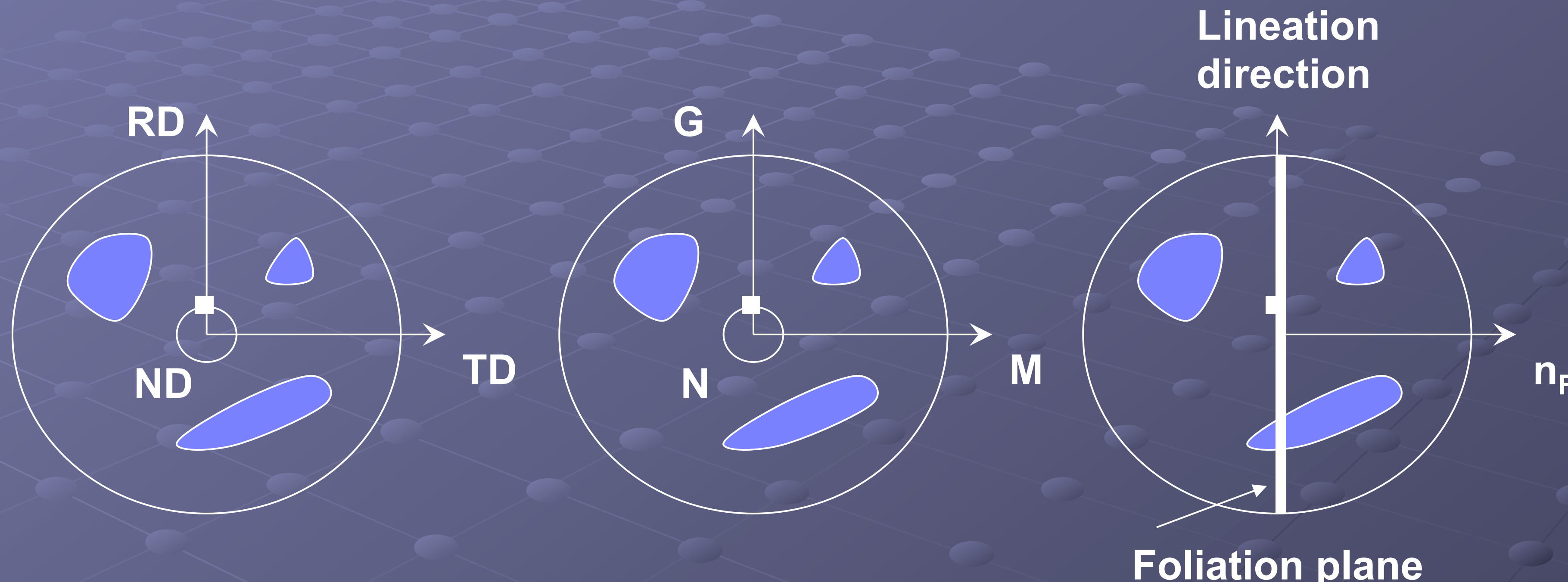




Direct Pole Figure: built on diffracted intensities I_h(**y**), **h** = <hkℓ>*
Normalised Pole Figure: built on distribution densities P_h(**y**)

Density unit: the "multiple of a random distribution", or "m.r.d."

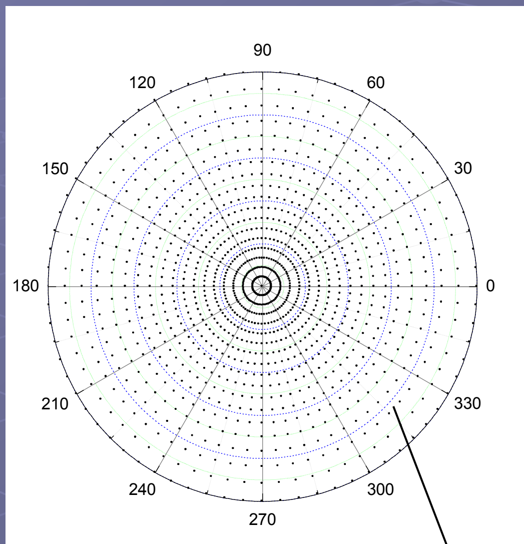
Usual pole figure frames K_A



metallurgy   malacology   geophysics

Thin films: substrate directions … X_A, Y_A, Z_A

Normalisation



$$I_{\mathbf{h}}(\vartheta_y, \varphi_y)$$

$$I_{\mathbf{h}}^{\text{total}} = \int_{\varphi_y=0}^{2\pi} \int_{\vartheta_y=0}^{\pi/2} I_{\mathbf{h}}(\vartheta_y, \varphi_y) \sin\vartheta_y \, d\vartheta_y d\varphi_y$$

$$I_{\mathbf{h}}^{\text{random}} = I_{\mathbf{h}}^{\text{total}} / \int_{\varphi_y=0}^{2\pi} \int_{\vartheta_y=0}^{\pi/2} \sin\vartheta_y \, d\vartheta_y d\varphi_y$$

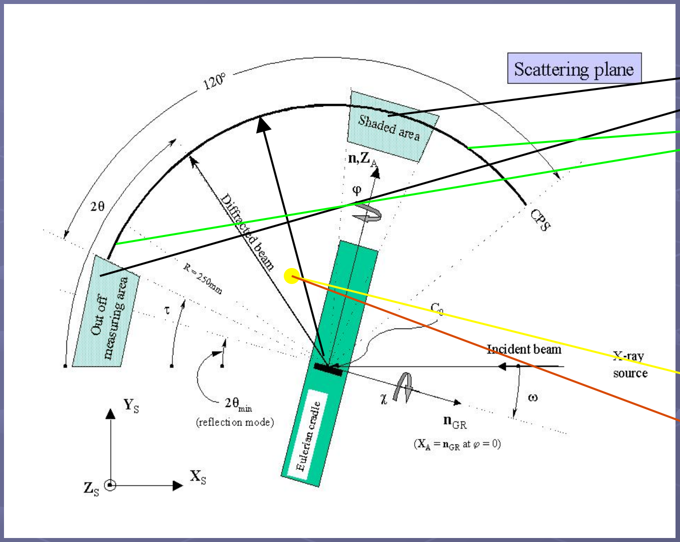
$$P_{\mathbf{h}}(\mathbf{y}) = \frac{I_{\mathbf{h}}(\mathbf{y})}{I_{\mathbf{h}}^{\text{random}}}$$

- Only valid for complete pole figures:

neutrons in symmetric geometry

- Needs a refinement strategy to get I^{random} for all **h**'s

Incompleteness and corrections of pole figures

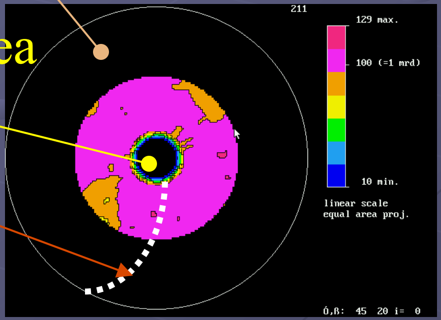


Missing Bragg peaks

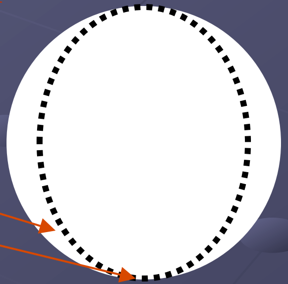
Absorption + volume

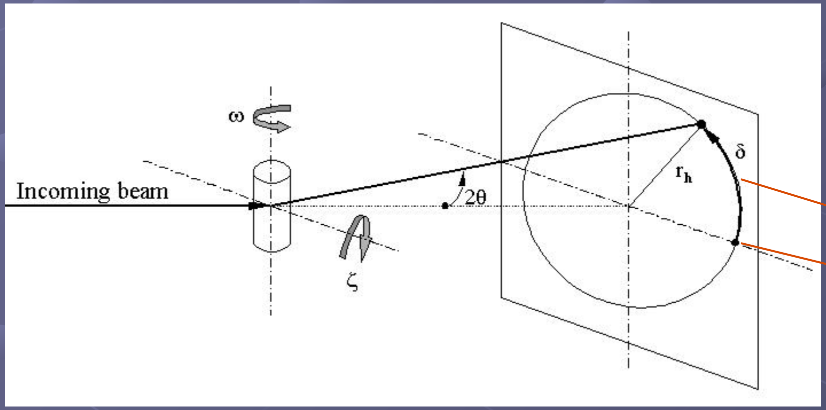
Defocusing (x-rays)

Blind area

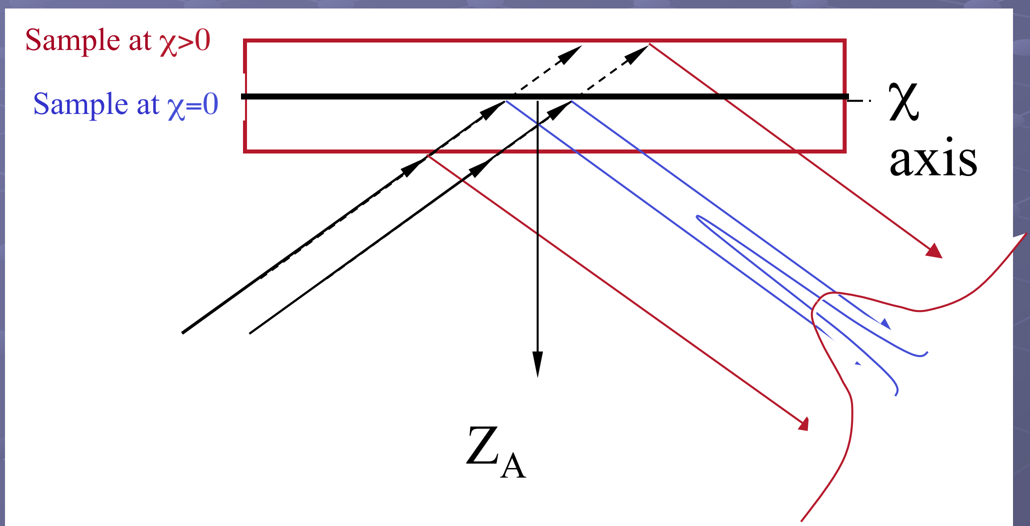


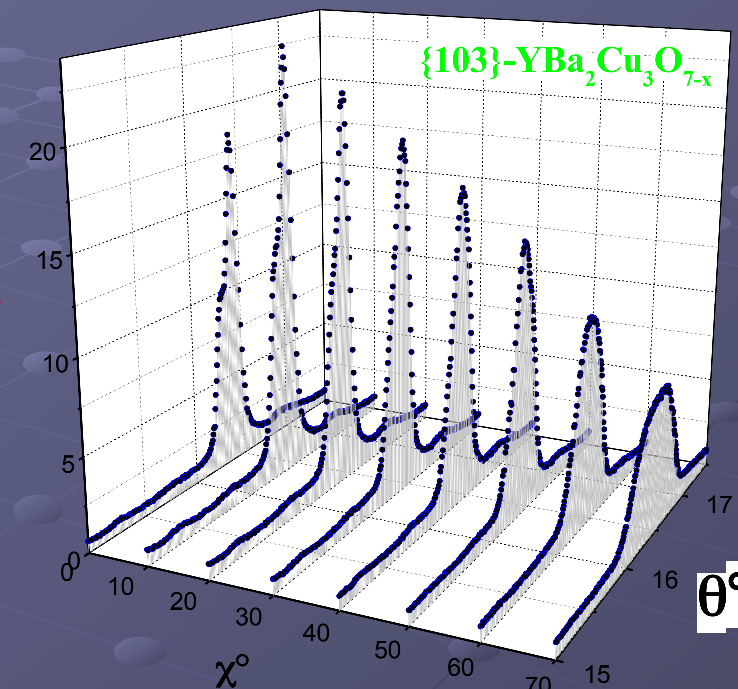
Localisation



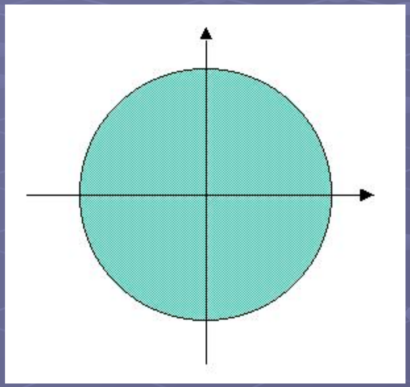


Defocusing (χ)



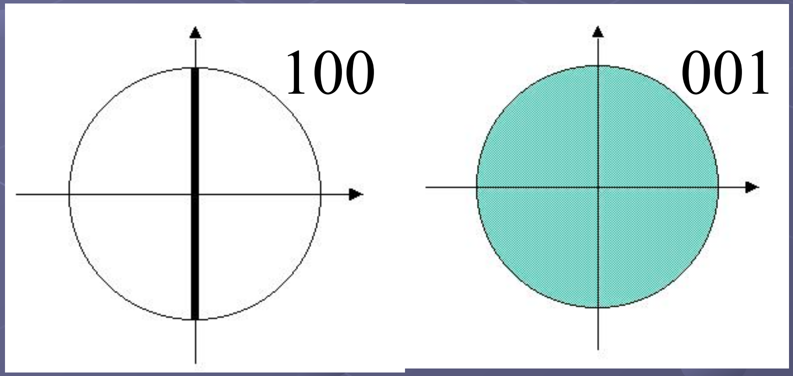


Texture types



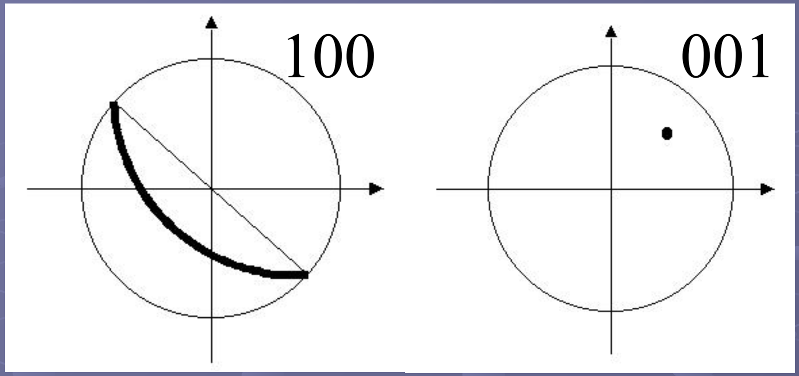
Random texture

3 degrees of freedom
All $P_h(\mathbf{y})$ homogeneous
1 m.r.d. density whatever $\mathbf{y}$



Planar texture

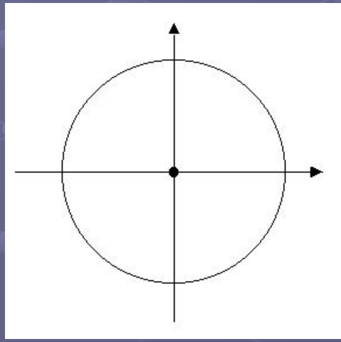
2 degrees of freedom
1 [hkℓ] at random in a plane



Fibre texture

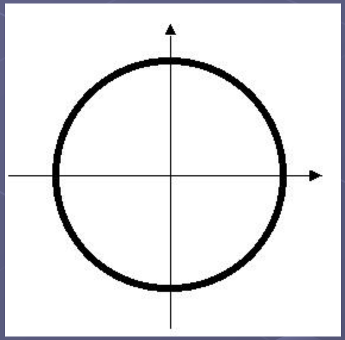
1 degree of freedom

1 [hkℓ] along 1 **y** direction



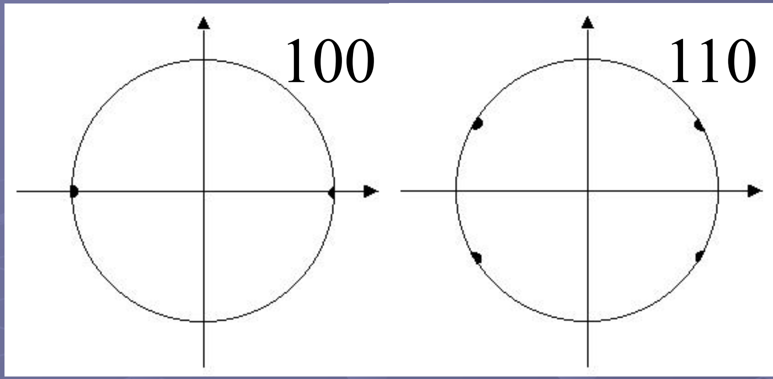
Cyclic-Fibre texture

c // Z_A



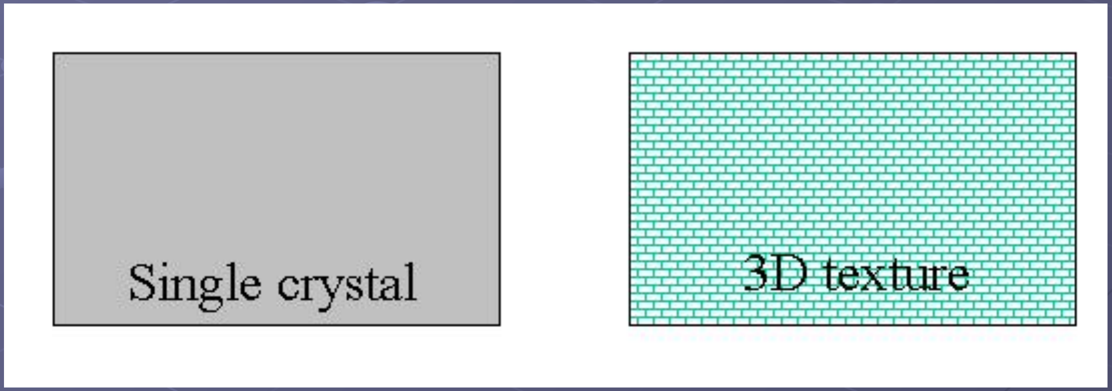
Cyclic-Planar texture

c // (X_A, Y_A)



Single crystal-like texture

0 degree of freedom
2 [hkℓ]'s along 2 **y** directions



Single-crystal and perfect 3D orientation not distinguished

Pole figure and Orientation spaces

Pole figure expression:

$$\frac{dV(\mathbf{y})}{V} = \frac{1}{4\pi} P_{\mathbf{h}}(\mathbf{y})\,\mathbf{dy}$$

$\mathbf{dy} = \sin\vartheta_y \, d\vartheta_y \, d\varphi_y$

$$\int_{\varphi_y=0}^{2\pi} \int_{\vartheta_y=0}^{\pi/2} P_{\mathbf{h}}(\vartheta_y, \varphi_y) \sin\vartheta_y \, d\vartheta_y d\varphi_y = 4\pi$$

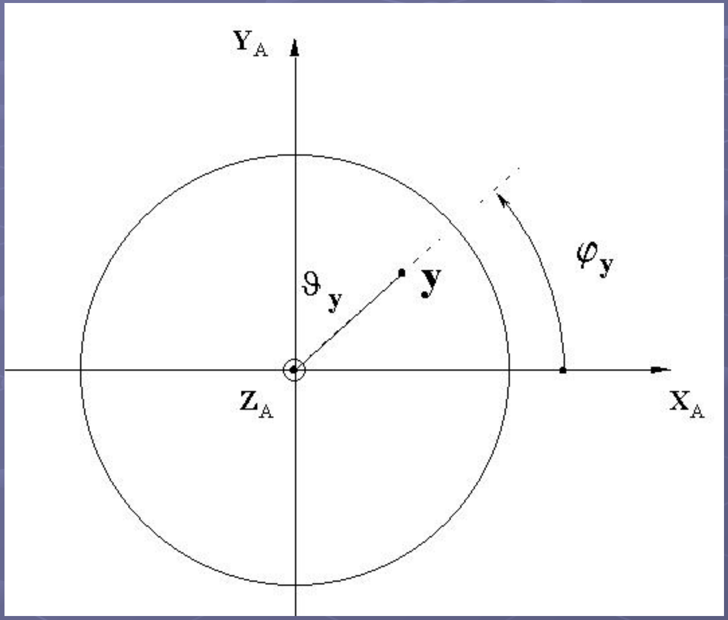
Orientation Distribution Function f(g):

$$\frac{dV(g)}{V} = \frac{1}{8\pi^2} f(g)\,dg$$

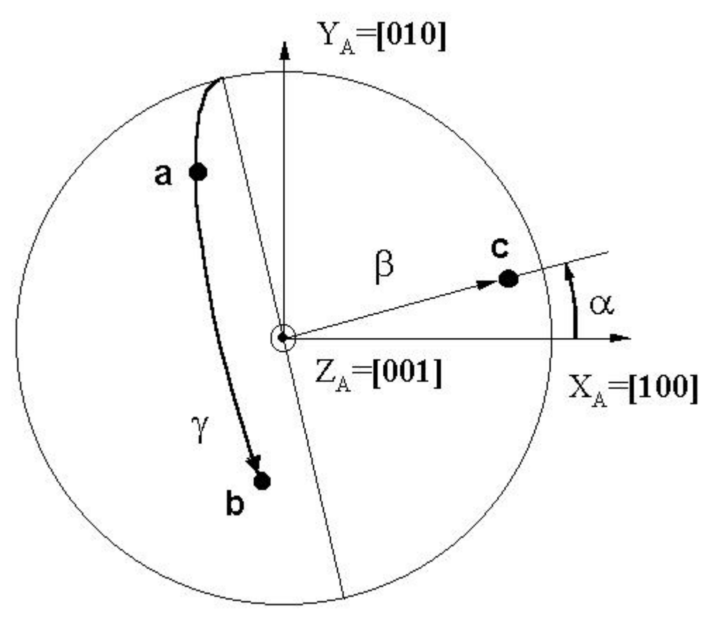
$dg = \sin(\beta) d\beta d\alpha d\gamma$

$$\int_{\alpha=0}^{2\pi} \int_{\beta=0}^{\pi/2} \int_{\gamma=0}^{2\pi} f(g)\,dg = 8\pi^2$$

From Pole figures to the ODF



Pole figure: one direction fixed in K_A



Orientation: two directions fixed in K_A

Fundamental Equation of QTA

$$P_{\mathbf{h}}(\mathbf{y}) = \frac{1}{2\pi} \int_{\mathbf{h}//\mathbf{y}} f(g)\, d\tilde{\varphi}$$

Needs several pole figures to construct f(g)

ODF refinement

One has to invert:

$$P_{\mathbf{h}}(\mathbf{y}) = \frac{1}{2\pi} \int_{\mathbf{h}//\mathbf{y}} f(g)\, d\tilde{\varphi}$$

- from Generalized Spherical Harmonics (Bunge):

$$f(g) = \sum_{l=0}^{\infty} \sum_{m,n=-l}^{l} C_l^{mn} T_l^{mn}(g)$$

$$P_{\mathbf{h}}(\mathbf{y}) = \sum_{l=0}^{\infty} \frac{1}{2l+1} \sum_{n=-l}^{l} k_l^n(\mathbf{y}) \sum_{m=-l}^{l} C_l^{mn} k_n^{*m}(\Theta_{\mathbf{h}} \phi_{\mathbf{h}})$$

Least-squares Refinement procedure

$$\sum_{\mathbf{h}} \sum_{\mathbf{y}} \left[I_{\mathbf{h}}(\mathbf{y}) - N_{\mathbf{h}} P_{\mathbf{h}}(\mathbf{y}) \right]^2 \mathbf{dy}$$

But even orders are the only available parts:

$$f^e(g) = \sum_{\lambda=0(2)}^{\infty} \sum_{m,n=-\lambda}^{\lambda} C_\lambda^{mn} T_\lambda^{mn}(g)$$

- WIMV iterative process (Williams-Imhof-Matthies-Vinel):

$$f^{n+1}(g) = N_n \frac{f^n(g) f^0(g)}{\left(\prod_{\mathbf{h}=1}^{I} \prod_{m=1}^{M_{\mathbf{h}}} P_{\mathbf{h}}^n(\mathbf{y}) \right)^{\frac{1}{IM_{\mathbf{h}}}}}$$

and

$$f^0(g) = N_0 \left(\prod_{\mathbf{h}=1}^{I} \prod_{m=1}^{M_{\mathbf{h}}} P_{\mathbf{h}}^{\exp}(\mathbf{y}) \right)^{\frac{1}{IM_{\mathbf{h}}}}$$

E-WIMV (Rietveld only):

$$f^{n+1}(g) = f^n(g) \prod_{m=1}^{M_{\mathbf{h}}} \left(\frac{P_{\mathbf{h}}(\mathbf{y})}{P_{\mathbf{h}}^n(\mathbf{y})} \right)^{r_n \frac{w_{\mathbf{h}}}{M_{\mathbf{h}}}}$$

with $0 < r_n < 1$, relaxation parameter,
M_h number of division points of the integral around k,
w_h reflection weight

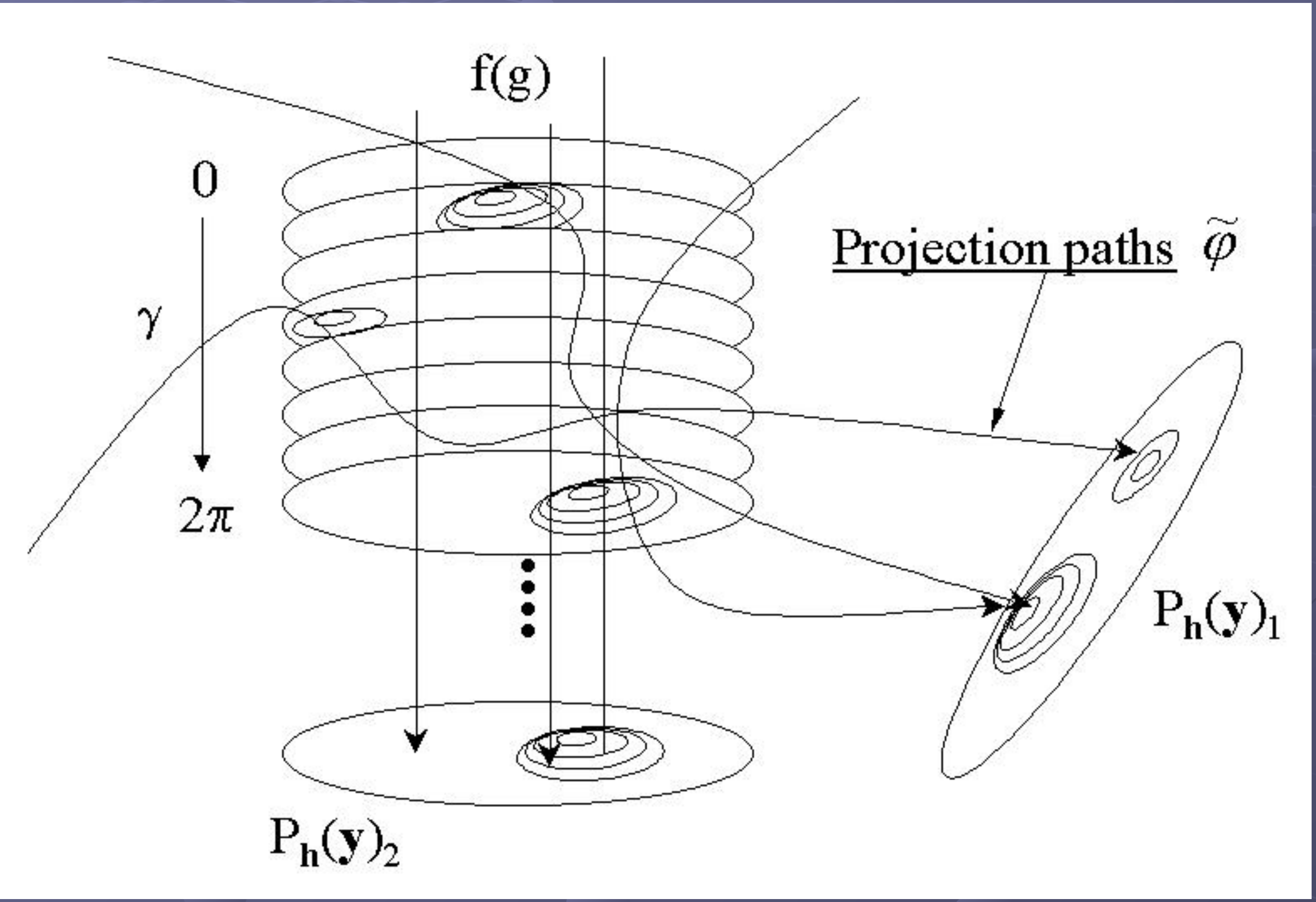
- Entropy maximisation (Schaeben) and exponential harmonics (van Houtte):

$$f^{n+1}(g) = f^n(g) \prod_{m=1}^{M_{\mathbf{h}}} \left(\frac{P_{\mathbf{h}}(\mathbf{y})}{P_{\mathbf{h}}^n(\mathbf{y})} \right)^{\frac{r_{\mathbf{n}}}{M_{\mathbf{h}}}}$$

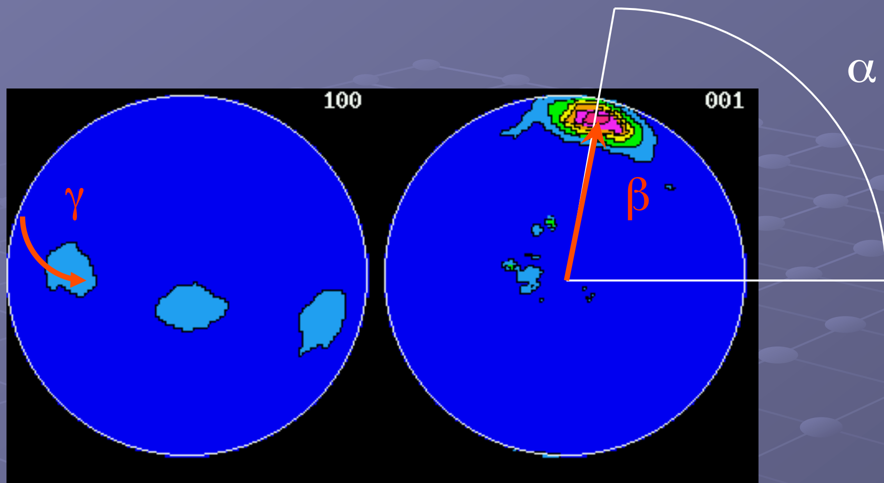
$$f_s(g) = e^{h(g)} \geq 0$$

$$C_{s\lambda}^{mn} = (2\lambda + 1) \int e^{h(g)} T_\lambda^{mn}(g) dg$$

From f(g) to the pole figures

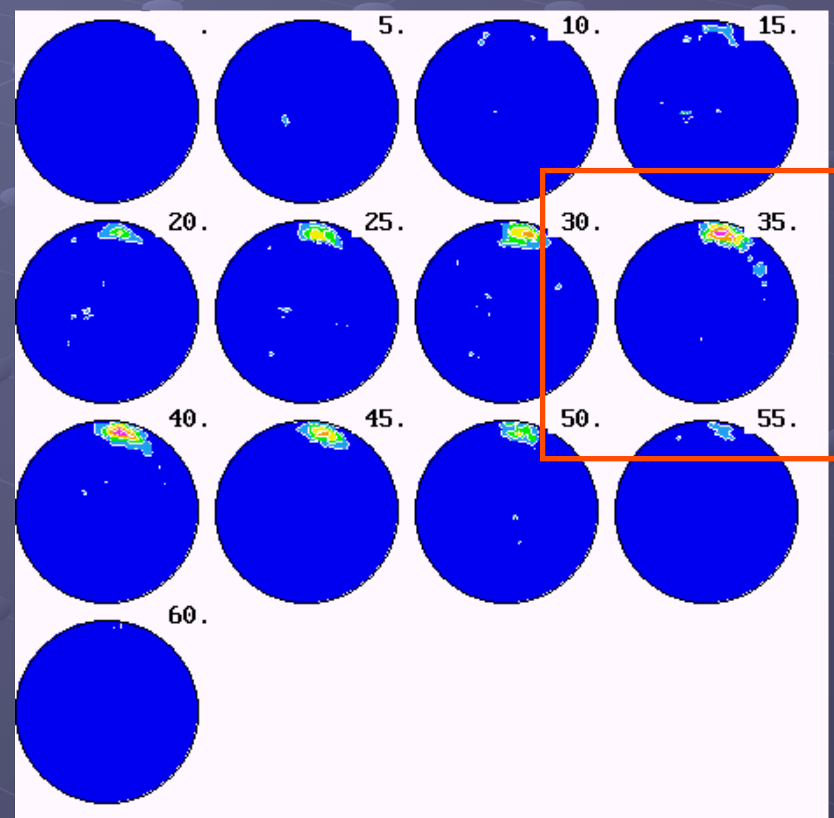


Deal with components in the ODF space



Pole figures

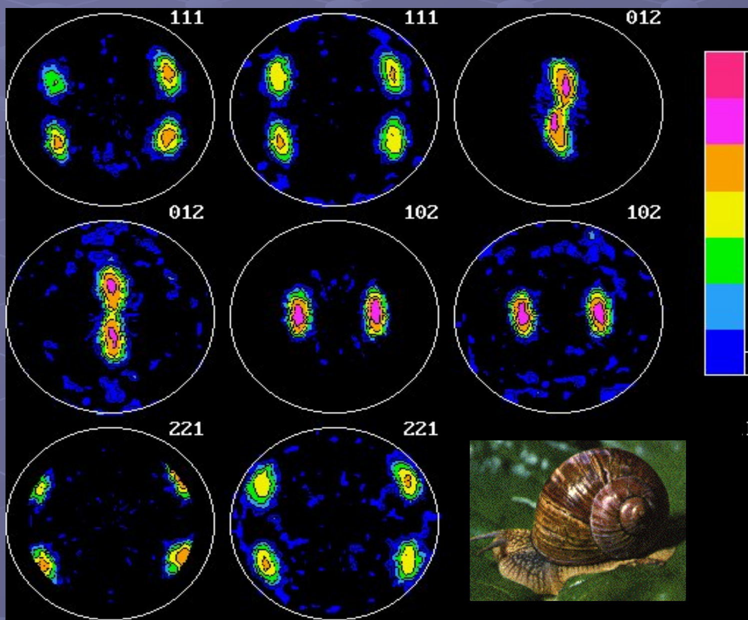
ODF γ-sections



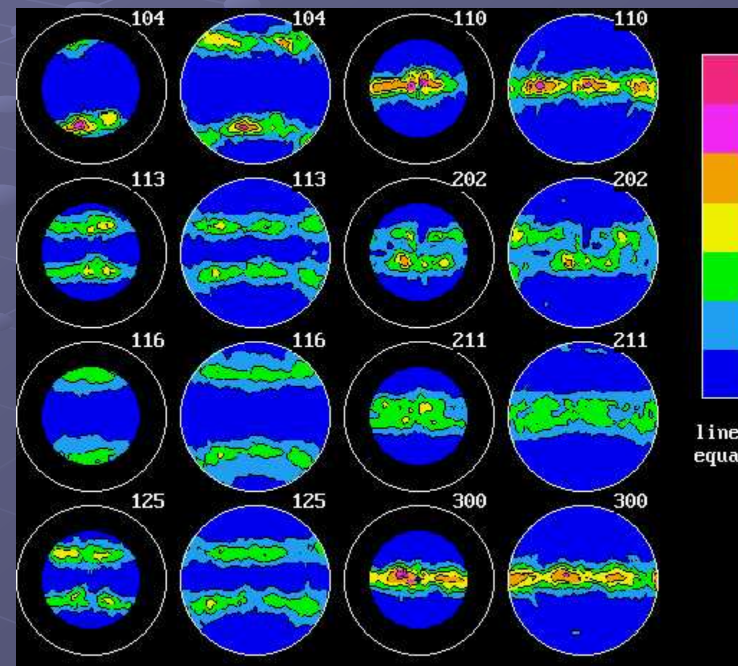
Component:
(Hexagonal system)
g = {85,80,35}

Estimators of Refinement Quality

Visual assessment



Helix pomatia (Burgundy land snail: Outer com. crossed lamellar layer)

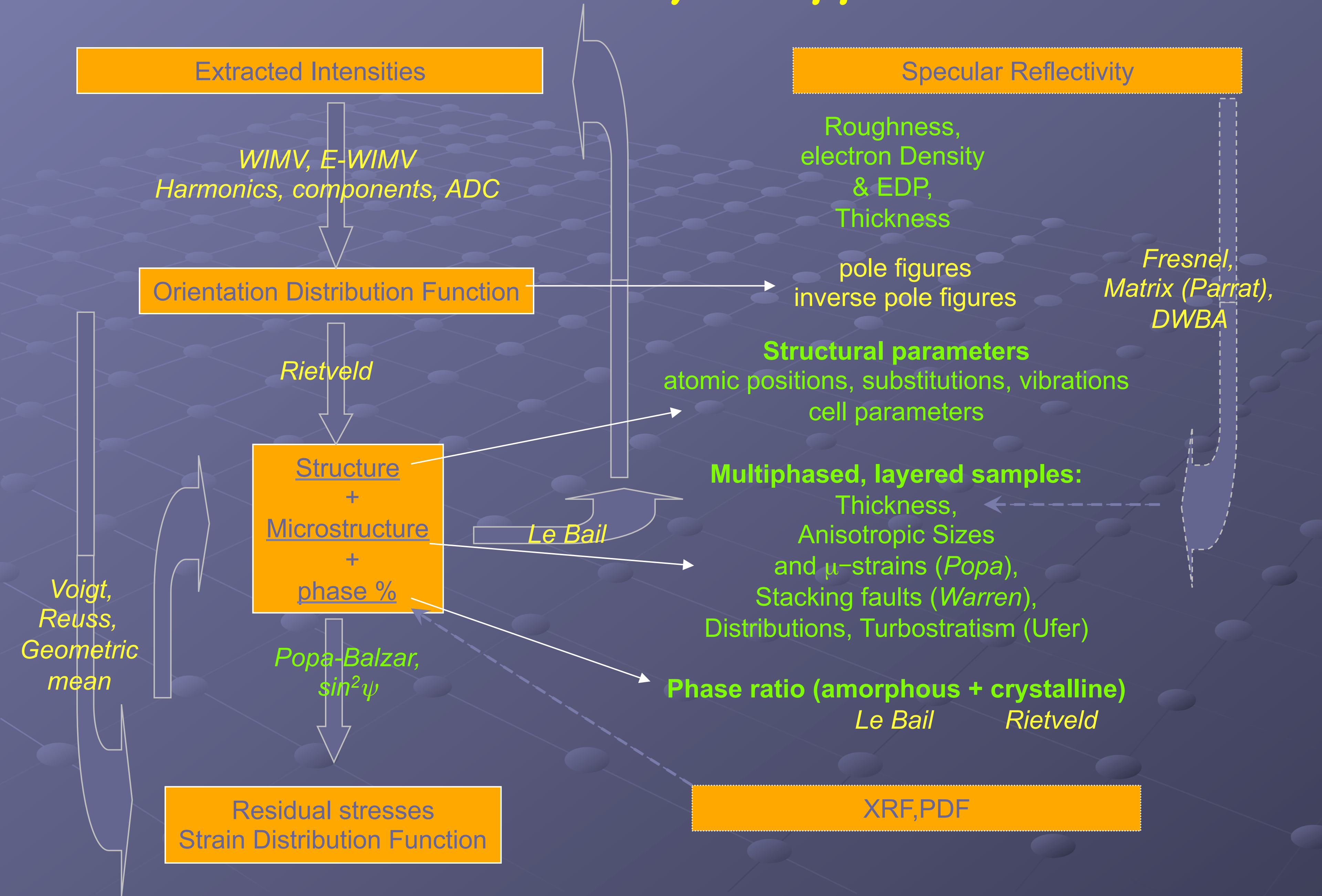


Bathymodiolus thermophilus (deep ocean mussel: Outer Prismatic layer)

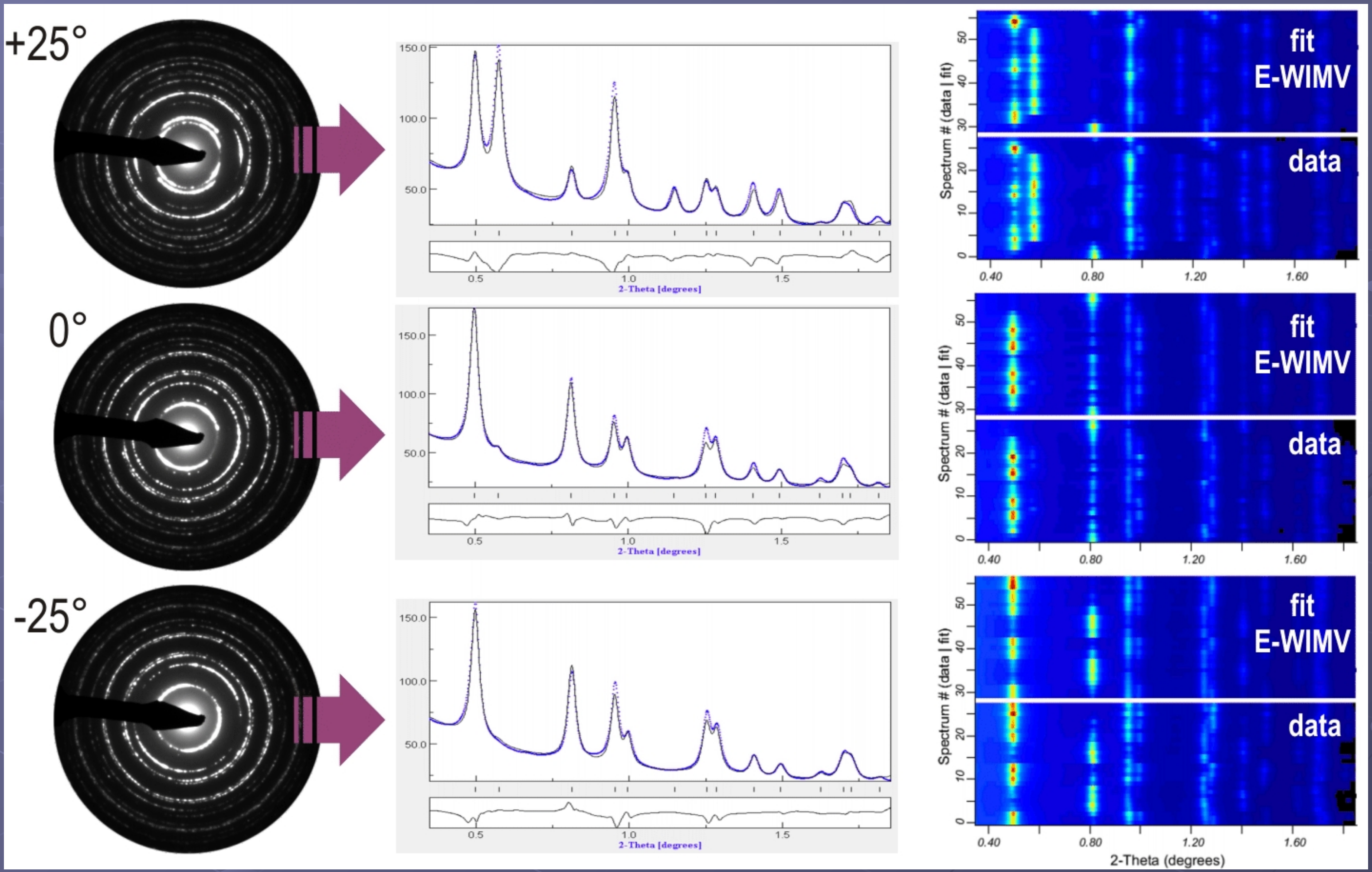
Rietveld – Texture and more: Combined Analysis

$$I_i^{calc}(\mathbf{y}) = \sum_{n=1}^{Nphases} S_n \sum_k L_k \left|F_{k;n}\right|^2 S\left(2\theta_i - 2\theta_{k;n}\right) P_{k;n}(\mathbf{y}) A + bkg_i(\mathbf{y})$$

Combined Analysis approach

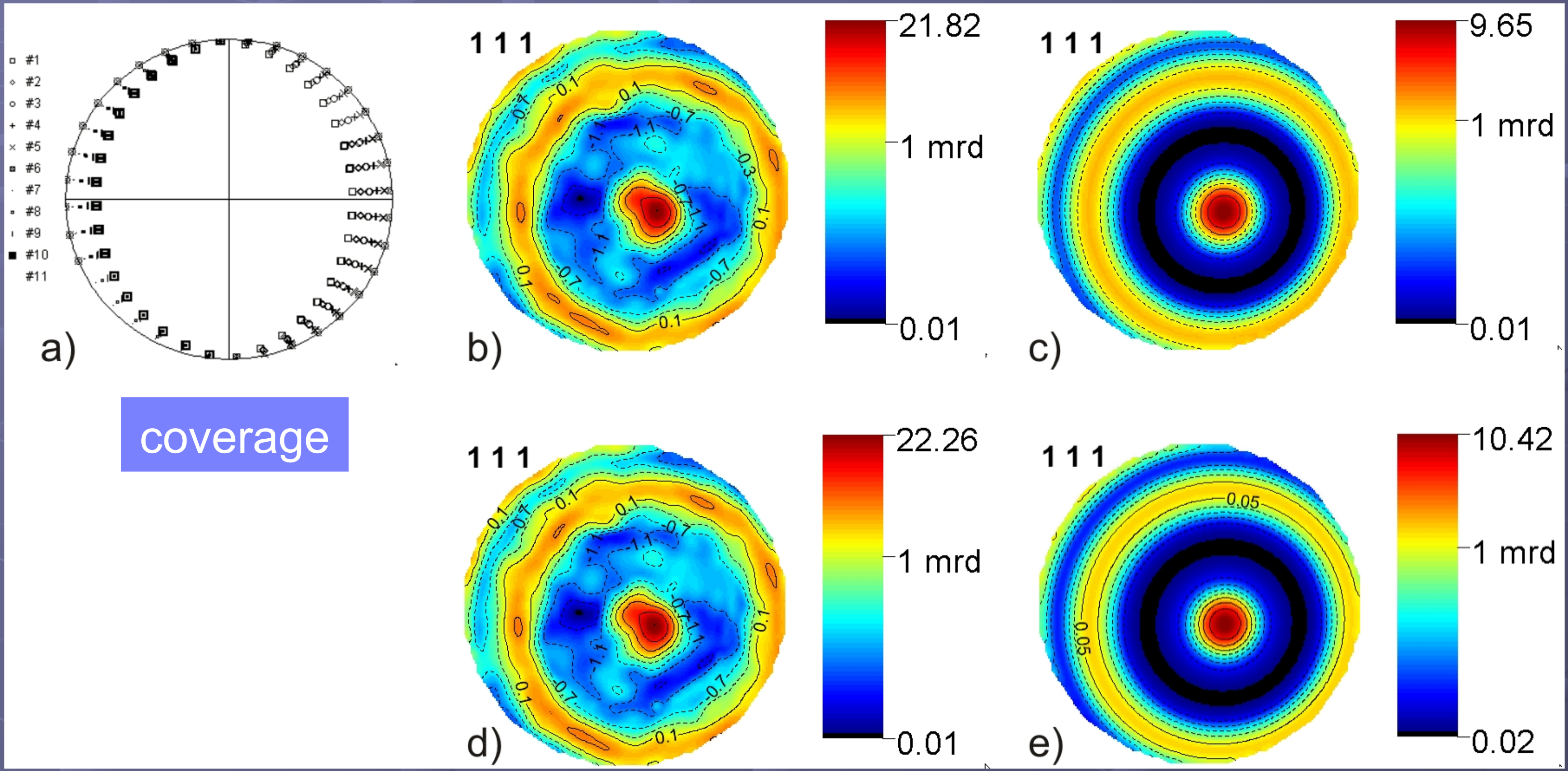


Paterns taken from +25° to -25° (step 5°) tilts: thin film prepared for TEM plan view



3 out of 11 EPD, 1D and 2D plots. Pattern matching (Pawley)

Pawley pattern matching
EWIMV Fiber component



coverage

EWIMV Fiber component
2-beams dynamical (Blackman)

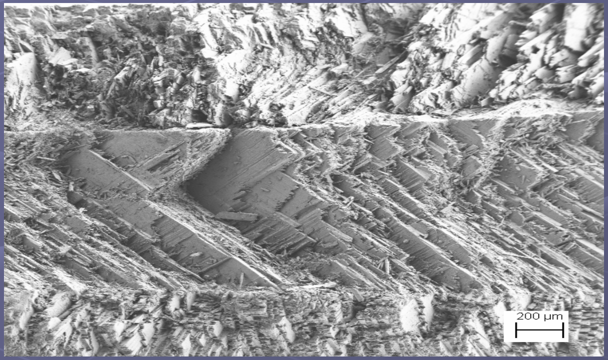
Why needing QTA

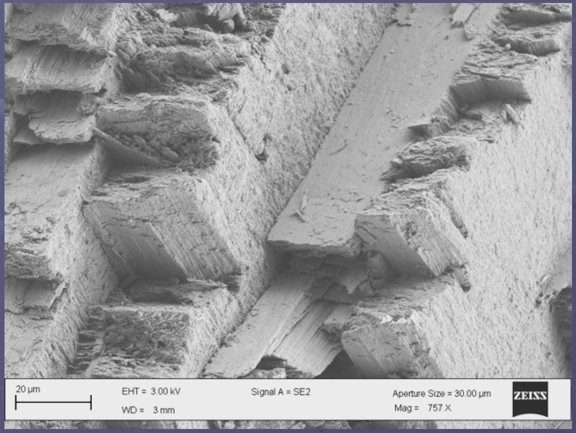
- Correct for QTA effects in XRD: structure analysis

QTA and structure correlations: yes, but

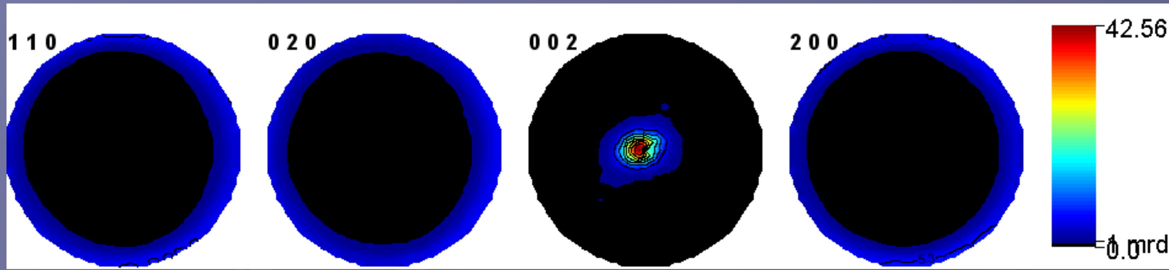
f(g) and $|F_{\mathbf{h}}|^2$ are different !

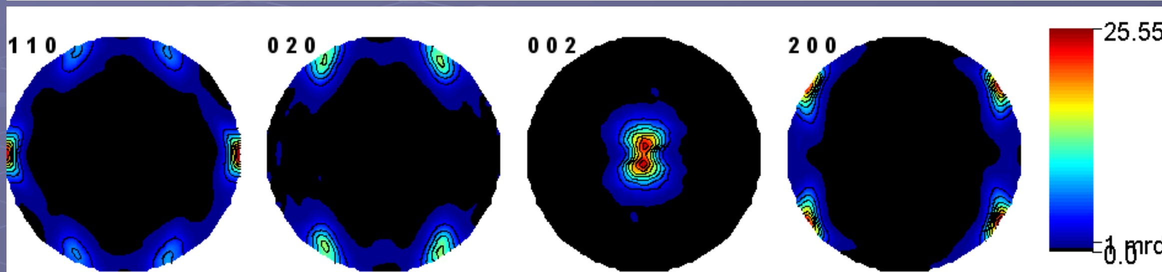
Charonia lampas lampas

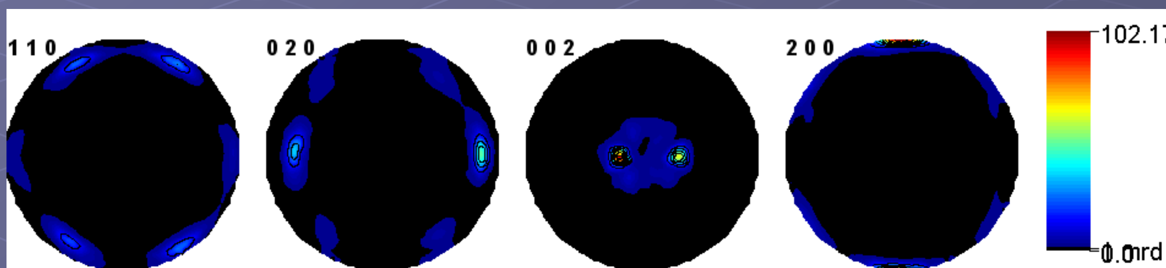


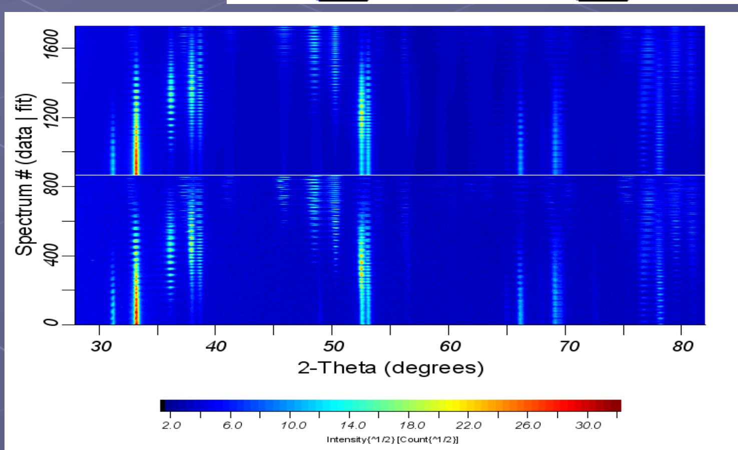


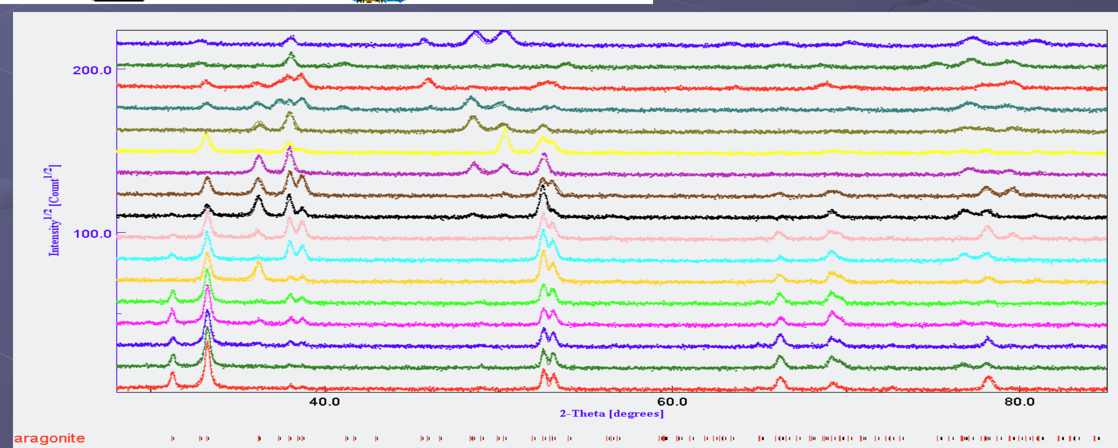
OD maximum (m.r.d.) **OD minimum (m.r.d.)**		299 0	196 0	2816 0
Texture index (m.r.d.²)		42.6	47	721
Texture reliability factors	**R_w (%)**	14.3	11.2	32.5
	R_B (%)	15.6	12.7	47.8
Rietveld reliability factors	**GoF (%)**	1.72	1.72	3.05
	R_w (%)	29.2	28.0	57.3
	R_B (%)	22.9	21.7	47.2
	R_{exp}(%)	22.2	21.3	32.8


1 1 0
0 2 0
0 0 2
2 0 0
42.56
1 mrd
0.0


1 1 0
0 2 0
0 0 2
2 0 0
25.55
1 mrd
0.0


1 1 0
0 2 0
0 0 2
2 0 0
102.17
1 mrd
0.0


Spectrum # (data | fit)
0
400
800
1200
1600
30
40
50
60
70
80
2-Theta (degrees)
2.0
6.0
10.0
14.0
18.0
22.0
26.0
30.0
Intensity(^1/2) [Count(^1/2)]


200.0
100.0
Intensity^1/2 [Count^1/2]
40.0
60.0
80.0
2-Theta [degrees]
aragonite

		Geological reference	*Charonia lampas* OCL	*Charonia lampas* RCL	*Charonia lampas* ICCL	**Strombus decorus**
a (Å) b (Å) c (Å)		4.9623(3) 7.968(1) 5.7439(3)	4.98563(7) 8.0103(1) 5.74626(3)	4.97538(4) 7.98848(8) 5.74961(2)	4.9813(1) 7.9679(1) 5.76261(5)	4.9694(3) 7.9591(4) 5.7528(1)
Ca	y z	0.41500 0.75970	0.41418(5) 0.75939(3)	0.414071(4) 0.76057(2)	0.41276(9) 0.75818(8)	0.4135(7) 0.7601(8)
C	y z	0.76220 -0.08620	0.7628(2) -0.0920(1)	0.76341(2) -0.08702(9)	0.7356(4) -0.0833(2)	0.7607(4) -0.0851(7)
O1	y z	0.92250 -0.09620	0.9115(2) -0.09205(8)	0.9238(1) -0.09456(6)	0.8957(3) -0.1018(2)	0.9228(4) -0.0905(9)
O2	x y z	0.47360 0.68100 -0.08620	0.4768(1) 0.6826(1) -0.08368(6)	0.4754(1) 0.68332(9) -0.08473(5)	0.4864(3) 0.6834(2) -0.0926(1)	0.4763(6) 0.6833(3) -0.0863(7)
ΔZ_{C-O1} (Å)		0.05744	0.00029	0.04335	0.1066	0.031

Calcite: $\Delta Z = 0$

Biogenic intercrystalline effect

- Predict macroscopic anisotropic properties: Elastic

Arithmetic average

$$\langle \mathcal{T} \rangle = \int_g \mathcal{T}(g)\, f(g)\, dg$$

$$\langle (\mathcal{T})^{-1} \rangle \neq \langle \mathcal{T} \rangle^{-1}$$

Voigt average
Homogeneous strain

$$C_{ijk\ell}^{M} = \langle C_{ijk\ell} \rangle$$

Upper bound

Reuss average
Homogeneous stress

$$S_{ijk\ell}^{M} = \langle S_{ijk\ell} \rangle$$

Lower bound

Geometric average

$$\lceil b \rceil = \prod_{k=1}^{N} b_k^{\,w_k} = \exp\left(\langle \ln b \rangle\right)$$

scalar

$$\langle \ln b \rangle = \sum_{k=1}^{N} \ln b_k\, w_k$$

$$\lceil T \rceil_{ij} = \exp(\langle \ln T \rangle_{i'j'})$$ tensor

$$\lceil \lambda_I \rceil = 1/\lceil 1/\lambda_I \rceil = \lceil \lambda_I^{-1} \rceil^{-1}$$ Eigenvalues of T_{ij}

$$\left\langle (C_{ijk\ell})^{-1} \right\rangle = \left\langle C_{ijk\ell} \right\rangle^{-1}$$

- Predict macroscopic anisotropic properties: Electric polarisation

$$\langle \mathbf{p}_h \rangle = \frac{\iint_{\mathbf{y}} \mathbf{p}_h P_h(\mathbf{y})\, d\mathbf{y}}{\iint_{\mathbf{y}} P_h(\mathbf{y})\, d\mathbf{y}}$$

- Predict macroscopic anisotropic properties: BAW

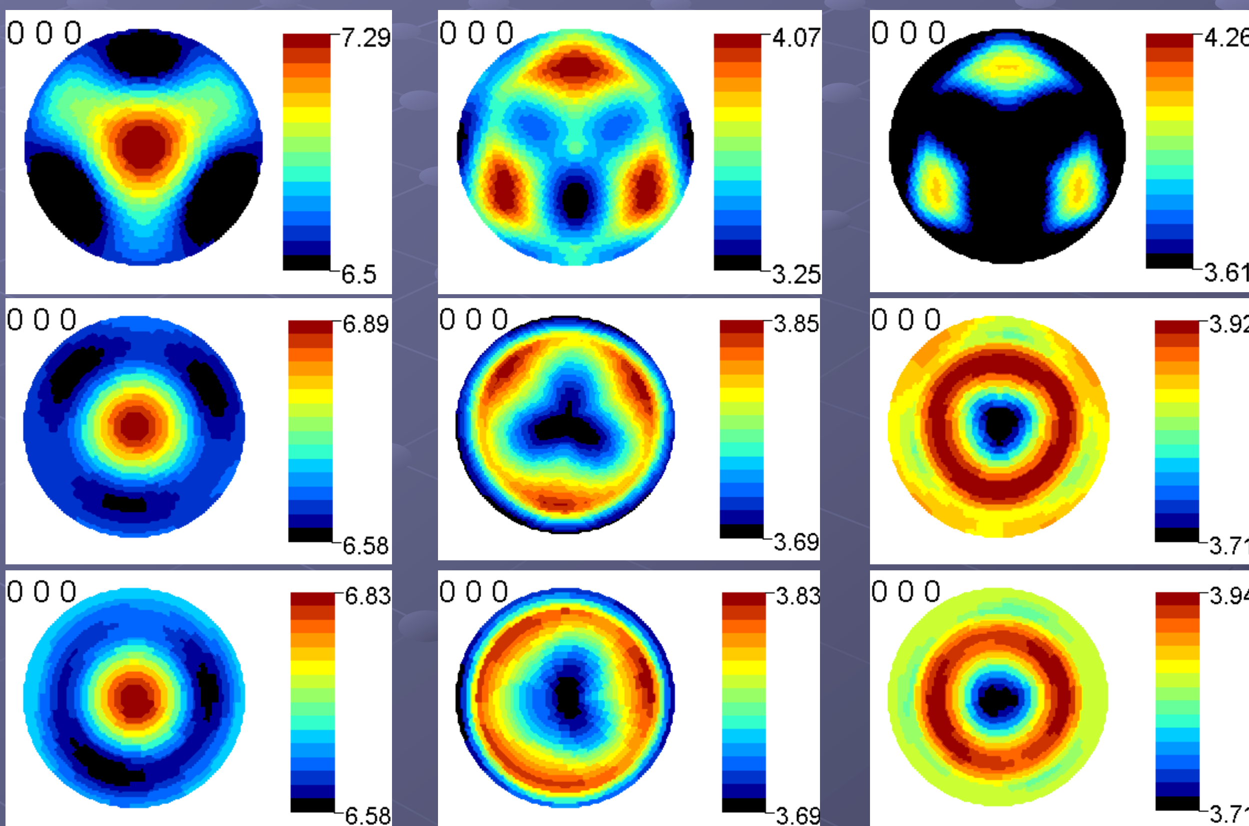
Propagation equation

$$\rho \frac{\partial^2 u^i}{\partial t^2} = \left[C^{i\ell mn}\right] \frac{\partial^2 u_n}{\partial x^m \partial x^\ell}$$

Propagation direction	V_P	V_{S1}	V_{S2}
[100]	$\sqrt{\frac{c^M{}_{11}}{\rho}}$	$\sqrt{\frac{c^M{}_{44}}{\rho}}$	$\sqrt{\frac{c^M{}_{44}}{\rho}}$
[110]	$\sqrt{\frac{c^M{}_{11} + 2c^M{}_{44} + c^M{}_{12}}{2\rho}}$	$\sqrt{\frac{c^M{}_{11} - c^M{}_{12}}{2\rho}}$	$\sqrt{\frac{c^M{}_{44}}{\rho}}$
[111]	$\sqrt{\frac{c^M{}_{11} + 4c^M{}_{44} + 2c^M{}_{12}}{3\rho}}$	$\sqrt{\frac{c^M{}_{11} + c^M{}_{44} - c^M{}_{12}}{3\rho}}$	$\sqrt{\frac{c^M{}_{11} + c^M{}_{44} - c^M{}_{12}}{3\rho}}$

Cubic crystal system

	c_{11} or c_{11}^M	c_{12} or c_{12}^M	c_{13} or c_{13}^M	c_{14} or c_{14}^M	c_{33} or c_{33}^M	c_{44} or c_{44}^M
Single crystal	201	54.52	71.43	8.4	246.5	60.55
$LiNbO_3$/Si	206.4	68.5	67.6	0.48	216.5	64
$LiNbO_3$/Al_2O_3	204	65.7	69.7	1.1	219.9	63.2



Single crystal

$LiNbO_3$/Si

$LiNbO_3$/Al_2O_3

THANKS !!!