ANISOTROPIC CRYSTALLITE SIZE ANALYSIS OF TEXTURED NANOCRYSTALLINE SILICON THIN FILMS PROBED BY X-RAY DIFFRACTION

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Introduction

➯ **The new "combined" x-ray technique is used, which is able to characterise quantitatively:**

- texture
- structure (cell parameters)
- anisotropic crystallite shapes
- film thickness

➯ **Samples are nanocrystalline silicon films, grown by reactive magnetron sputtering**

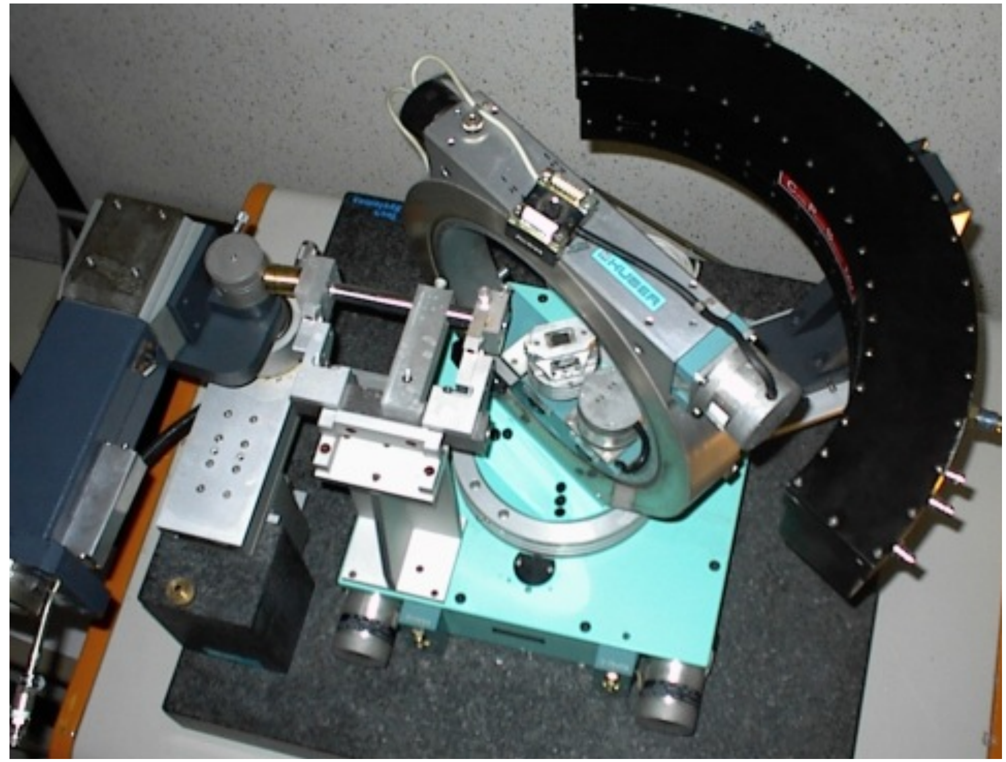
➯ **Crystallographic results are correlated to refraction indexes and optical pseudo-gap**

X-rays experiments

4 - circles diffractometer

+

Curved Position Sensitive Detector



Cu K_{α} radiation, Graphite monochromator, calibration: LaB_6

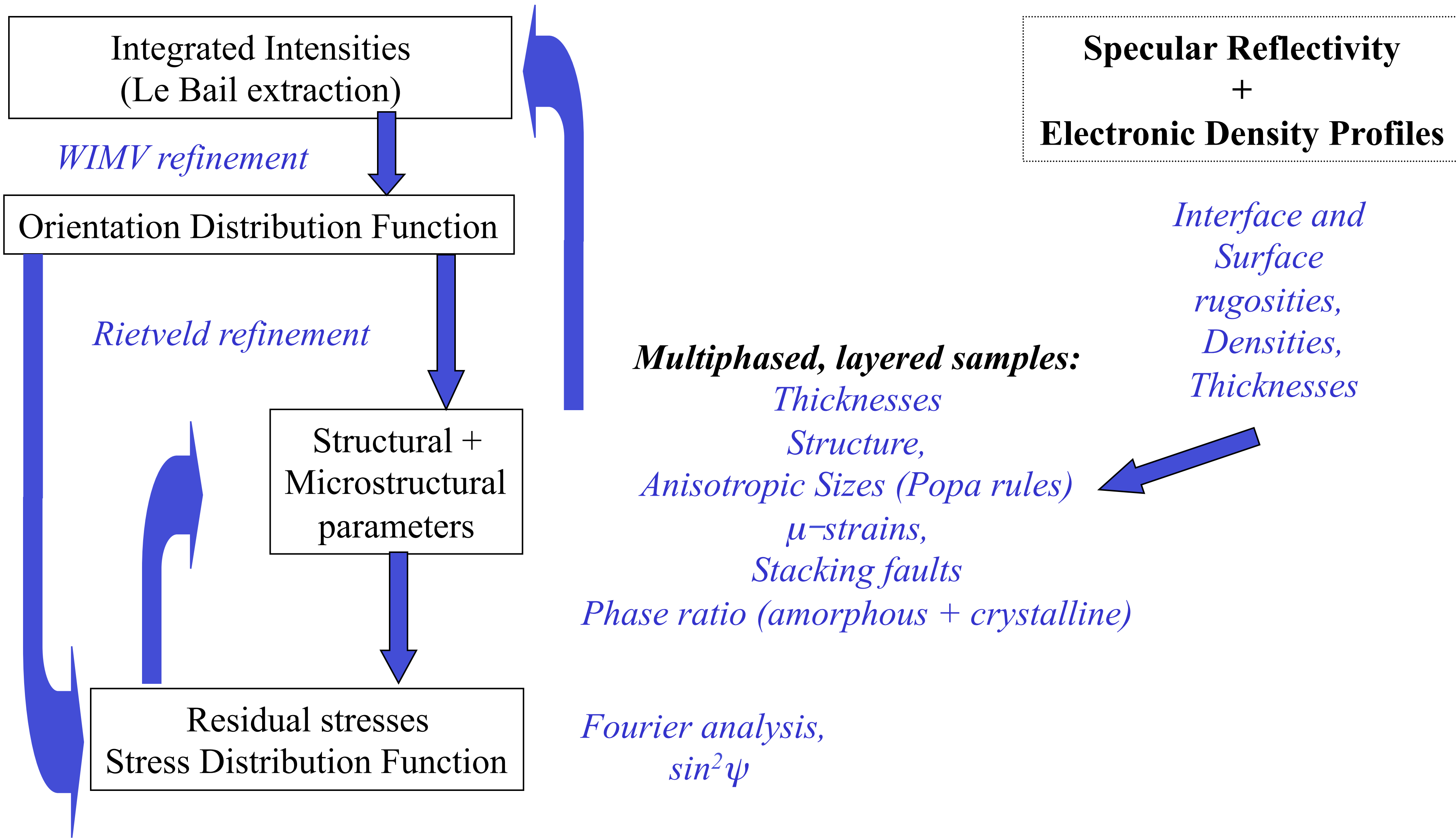
scans: $\omega = 14.25°$ (111 Si reflection), $0 \leq \chi \leq 35°$, $\Delta\chi = 5°$

Samples

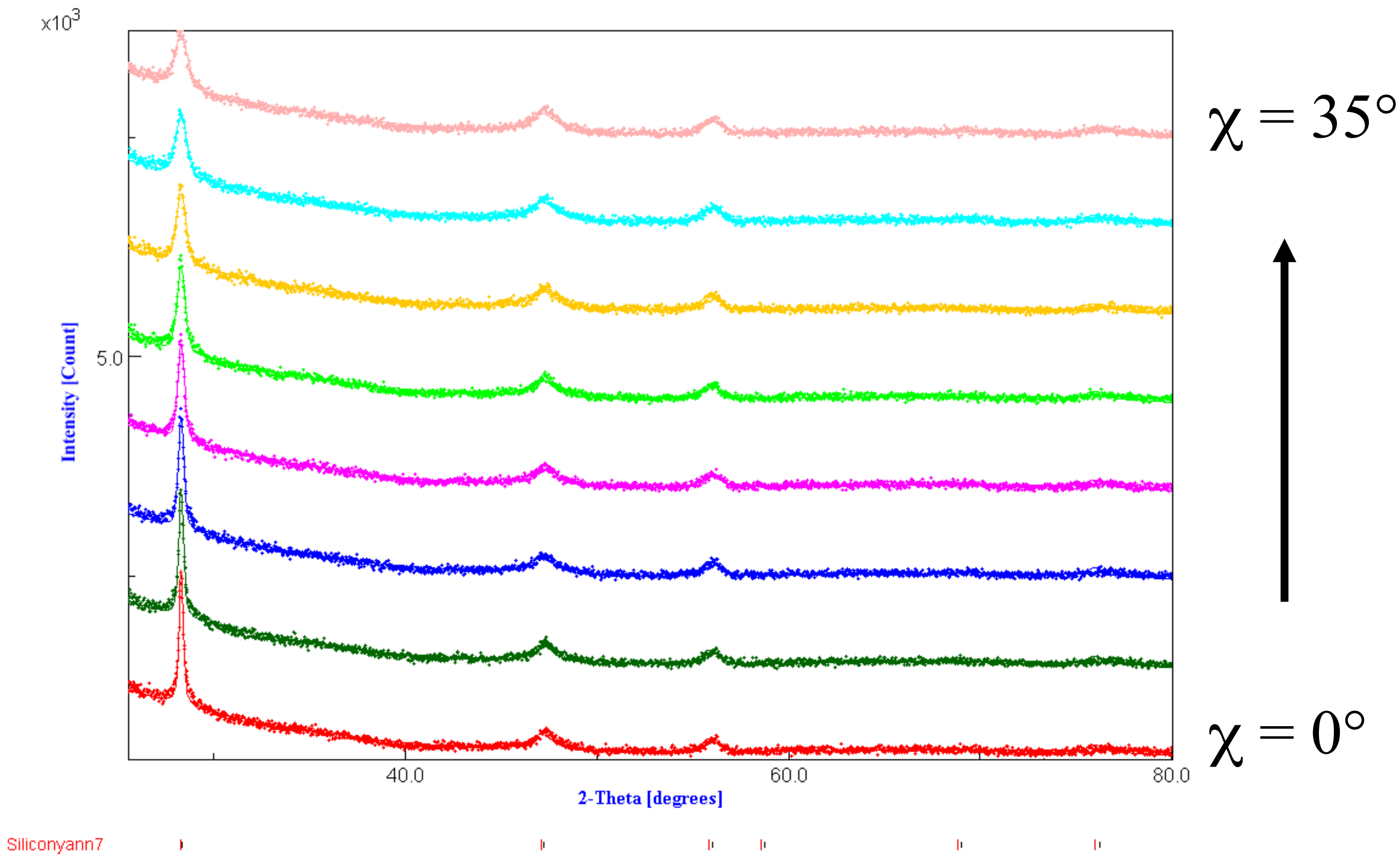
Silicon thin films deposition by reactive magnetron sputtering:

- power density 2W/cm^2
- total pressure: $p_{total} = 10^{-1}$ Torr
- plasma mixture: H_2 / Ar, pH_2 / p_{total} = 80 %
- temperature: 200°C
- substrates: amorphous SiO_2 (a-SiO_2)
  (100)-Si single-crystals
- target-substrate distance (d)
  - a-SiO_2 substrates: d = 4, 6, 7, 8, 10, 12 cm
    films A, B, C, D, E, F
  - (100)-Si: d = 6, 12 cm
    films G, H

Combined XRD analysis: MAUD



Typical refinement



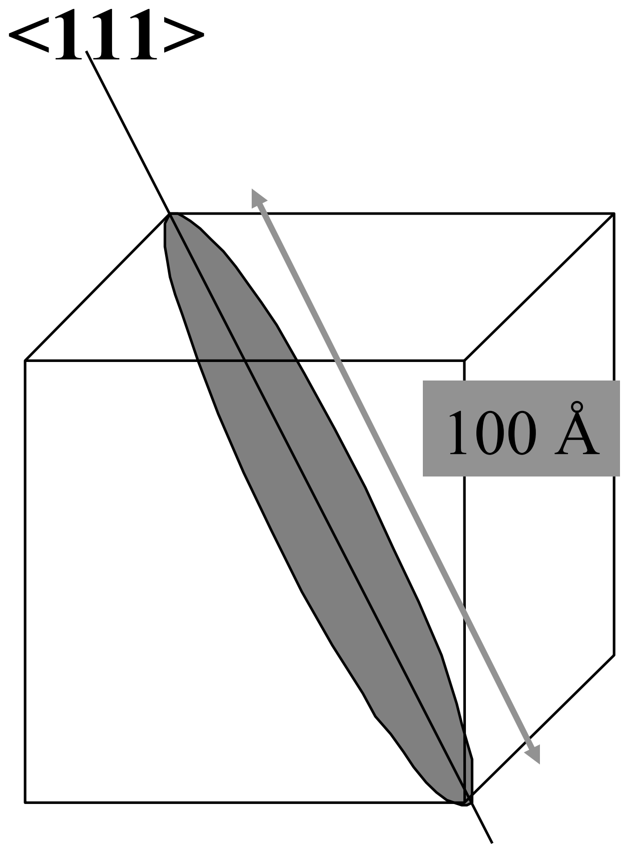
Measured: dots, simulated: lines

broad, anisotropic diffracted lines, textured samples

Refinement Results

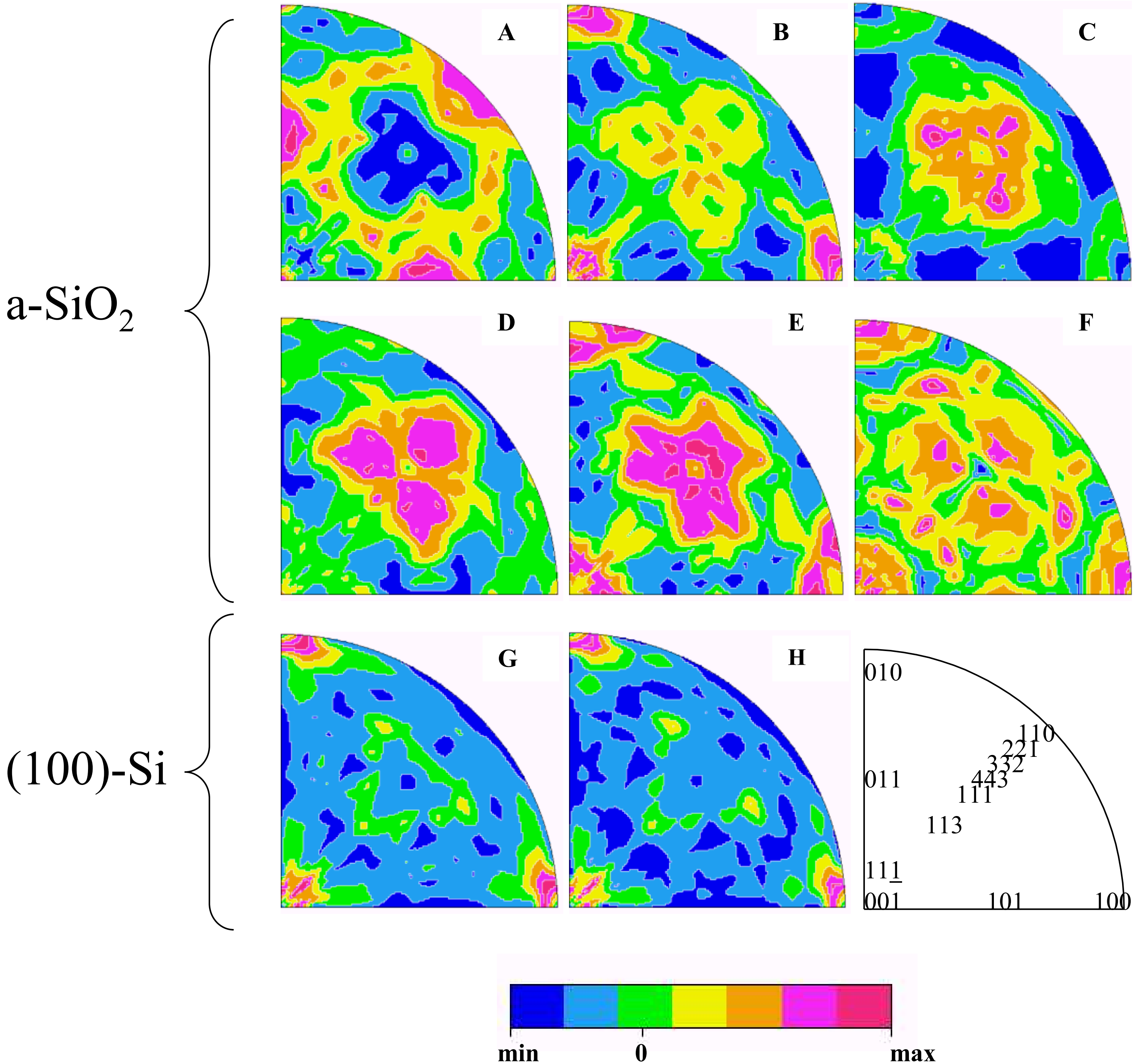
Sample	d (cm)	a (Å)	RX thickness (nm)	Anisotropic sizes (Å)			Texture parameters			Reliability factors (%)			
				<111>	<220>	<311>	Maximum (m.r.d.)	minimum (m.r.d.)	Texture index F^2 (m.r.d^2)	RP_0	R_w	R_B	R_{exp}
A	4	5.4466 (3)	---	94	20	27	1.95	0.4	1.12	1.72	4.0	3.7	3.5
B	6	5.4439 (2)	711 (50)	101	20	22	1.39	0.79	1.01	0.71	4.9	4.3	4.2
C	7	5.4346 (4)	519 (60)	99	40	52	1.72	0.66	1.05	0.78	4.3	4.0	3.9
D	8	5.4461 (2)	1447 (66)	100	22	33	1.57	0.63	1.04	0.90	5.5	4.6	4.5
E	10	5.4462 (2)	1360 (80)	98	20	25	1.22	0.82	1.01	0.56	5.0	3.9	4.0
F	12	5.4452 (3)	1110 (57)	85	22	26	1.59	0.45	1.05	1.08	4.2	3.5	3.7
G	6	5.4387 (3)	1307 (50)	89	22	28	1.84	0.71	1.01	1.57	5.2	4.7	4.2
H	12	5.4434 (2)	1214 (18)	88	22	24	2.77	0.50	1.12	2.97	5.0	4.5	4.3

Mean anisotropic shape



Schematic of the mean crystallite shape for Sample D represented in a cubic cell, as refined using the Popa approach and exhibiting a strong elongation along <111> (see Table).

001 Inverse Pole Figures



Texture evolution with d

Films on a-SiO_2 substrates:

• overall texture strength almost unaffected by d (F^2 around 1.2 m.r.d.^{2} at maximum), but texture components strongly influenced:

- smallest distances (Sample A) favours <110> orientation with minor <100> and <124> components
- <110> orientation is destabilised for larger d's

• <110> component removal accompanied by a slight tilt of <100> and the appearance of a large <221> like component

• progressive shift of <221> like component towards <111> for larger d's (Samples B to F)

• no pure <111> orientation is observed

Films on (100)-Si substrates:

- stabilisation of single <100> component for all d's

- heteroepitaxial growth:

  - [100]-film // [100]-substrate
  - native amorphous SiO_2 layer etched by hydrogen species of the plasma

- no <111> orientation is observed

- a_{Si} in films always larger than a_{Si} in bulk

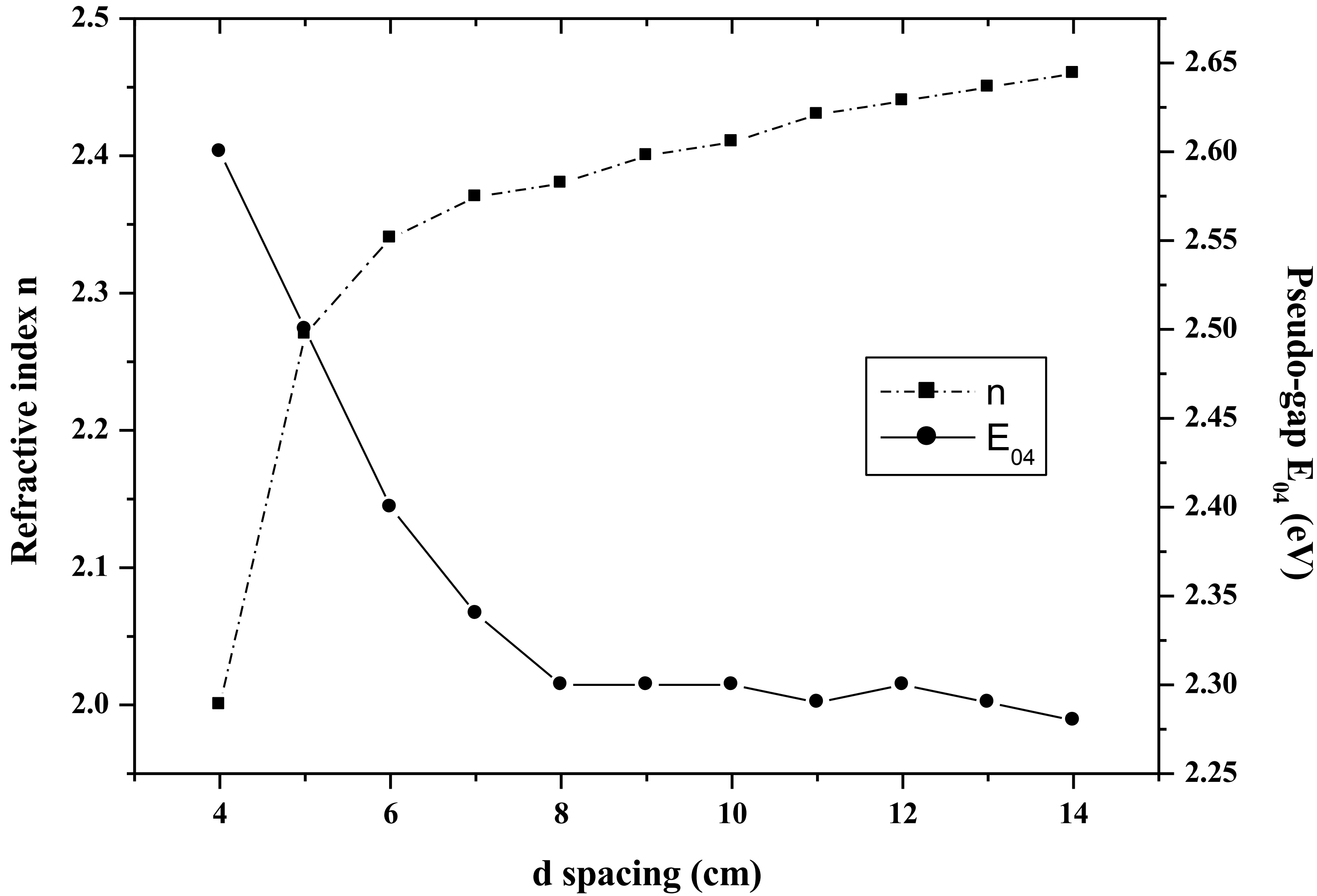
- ODF maxima larger using (100)-Si substrates

Profilometry versus XRD thickness

Samples	d (cm)	Profilometry Thickness (nm)	RX thickness (nm)
A	4	700	-----
B	6	1350	711 (50)
C	7	1530	519 (60)
D	8	1465	1447 (66)
E	10	1470	1360 (80)
F	12	1208	1110 (57)
G	6	1350	1307 (50)
H	12	1200	1214 (18)

} high porosity (samples B and C)

Optical measurements: refractive indexes and pseudo-gap



μ−structure versus optical properties

- large minimum ODF values: small anisotropy expected, 60 % to 20 % of textured volume

- abrupt increase of the refractive index (n) for small d's then saturation

⇓

reflects the film compactness

- opposite evolution of n and E_{04}

⇓

relatively high density of microcavities inherent to the film porosity

Conclusions

- Preferred orientations, cell parameters and anisotropic crystallite sizes of nanocrystalline silicon thin films deposited on a-SiO_2 and (100)-Si substrates have been quantitatively determined.

- Strong texture variations are observed when the electrode distance and/or the substrate is varied.

- Texture variations are correlated to the anisotropic crystal growth

- Porosities are correlated to refractive indices