Characterization of crystalline materials by X-Ray topography.

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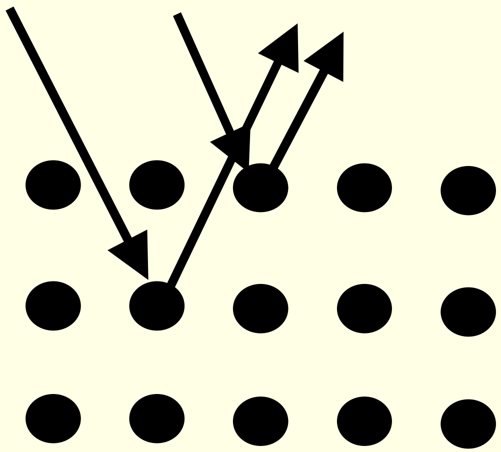
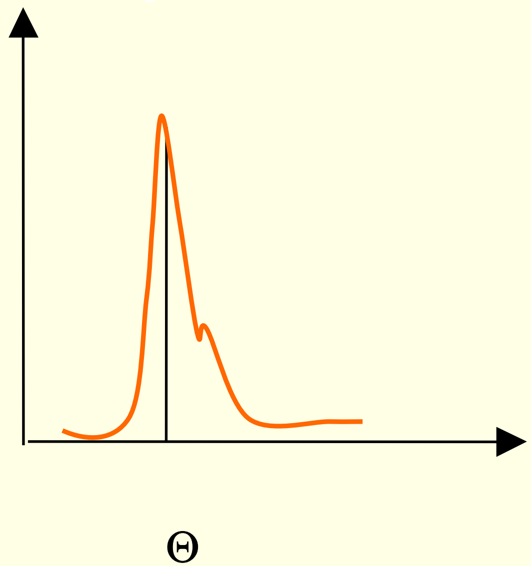
Agenda

- X-Ray Dynamical theory
  - Kinematical and Dynamical theory
  - Propagation of wave-fields
  - X-Ray topography
- Propagation of ultra-acoustic waves in piezoelectric devices
- Characterization of biological materials
- Heating of monochromators

Kinematical theory

- Limitations of the kinematical theory:



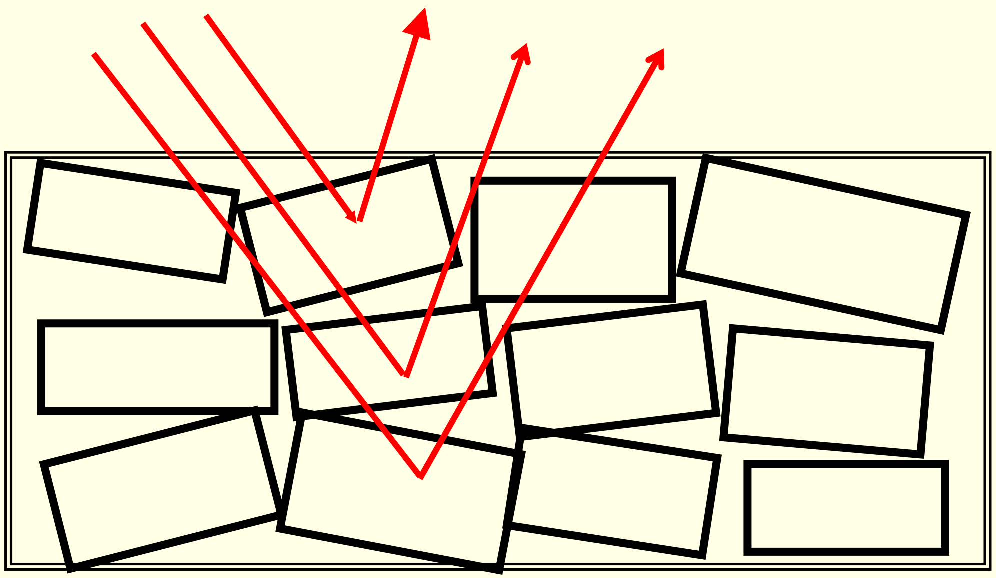


- Multiple diffraction is neglected, i.e. the intensity of the diffracted beam is small
- The index of refraction is 1, i.e. vacuum

The Bragg peak should be a Dirac!

Kinematical theory

This assumption is acceptable for imperfect crystals which may be described as a mosaic



Each bloc diffracts incoherently so that multiple diffraction does not occur.

Dynamical theory

The dynamical theory takes into account:

- The importance of the reflected amplitude i.e. multiple reflection: no need of a mosaic spread
- The refractive index of the materials (less than 1).

Today formalism is based on von Laue description:

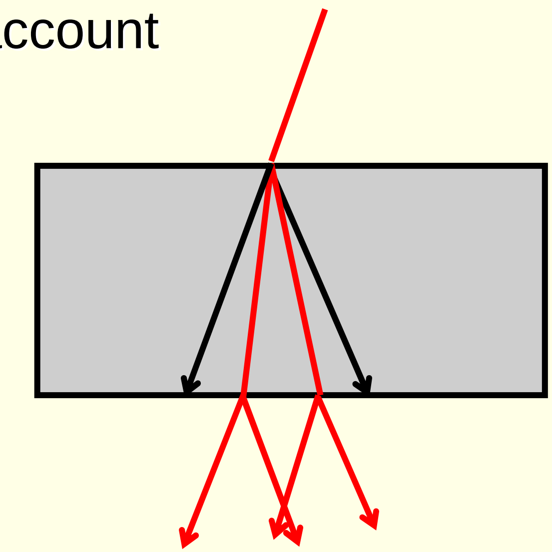
- Maxwell equations
- Fourier expansion of the polarizability

$$D(r) = \varepsilon E(r) = \varepsilon_0 E(r) + P(r) = \varepsilon_0 \left[1 + \chi(r)\right] E(r)$$

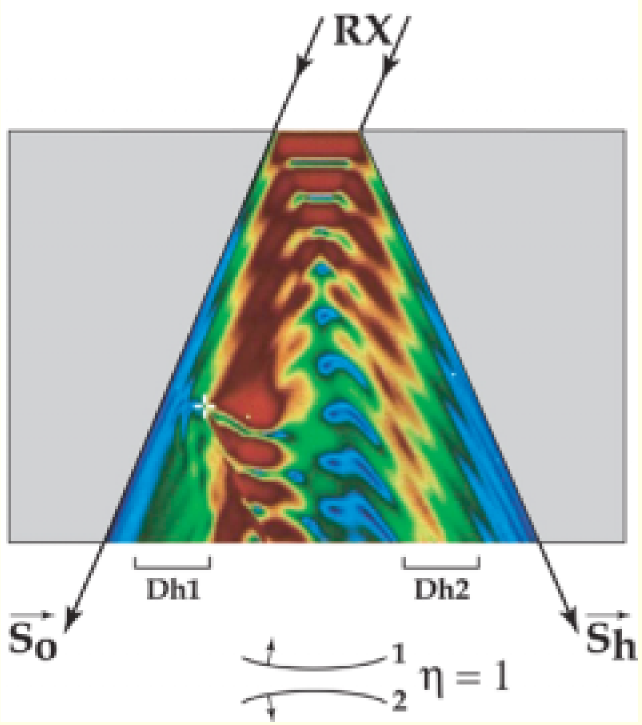
$$\chi(r) = \sum_h \chi_h(r) \exp\left(2\pi i h.r\right)$$

Dynamical theory

- The energy propagates in the crystal as wave-fields
- The nature of the incident wave has to be taken into account



Laue case

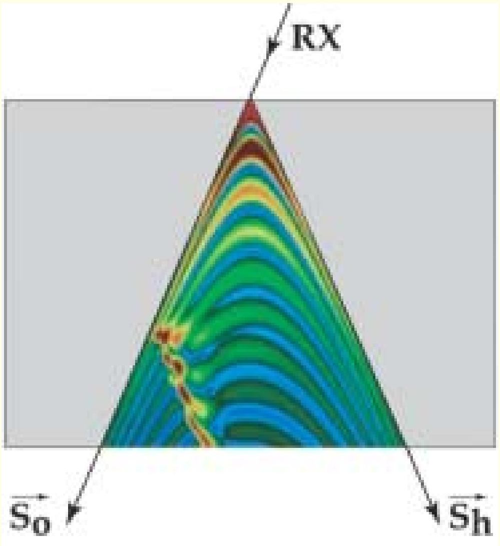


Incident plane wave (monochromator)

Dynamical theory

Kato approximation of a wave:
incident point source:

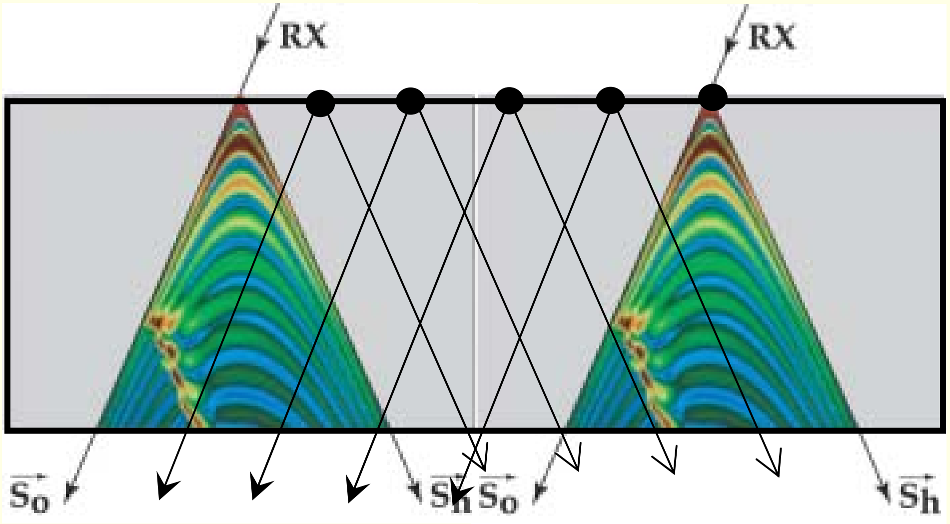
- Wave-fields interfere
- They are sensitive to the local deformation, thus to any local stress.



Laue case

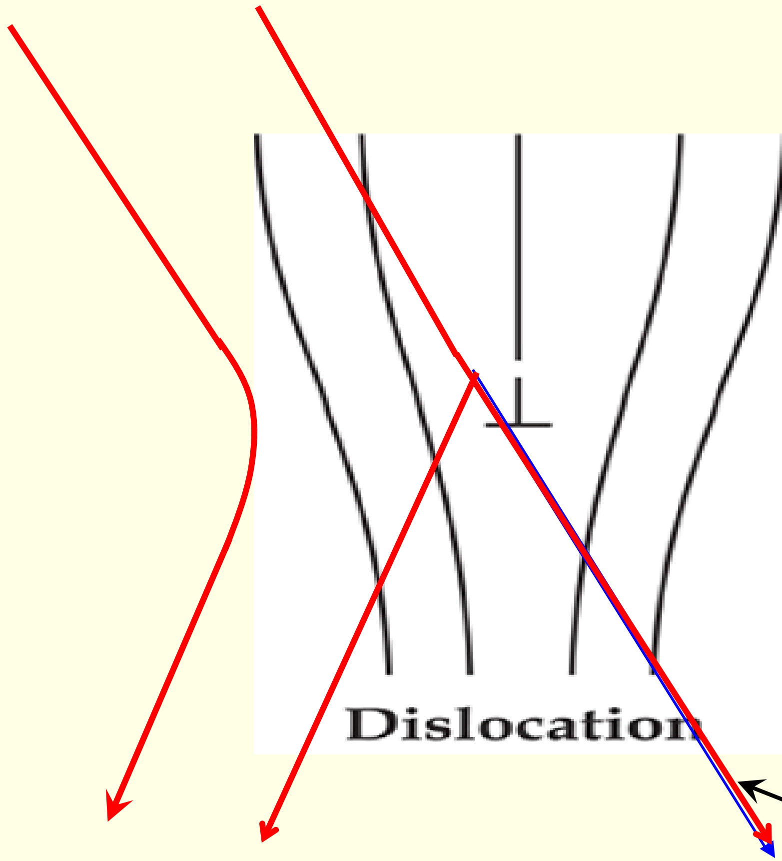
Dynamical theory

- In a white beam topograph the contrast arises from the contribution of incoherent point sources distributed along the surface.



Dynamical theory

Wave-fields are sensitive to the local deformation



They may be diffracted if the deformation is large enough

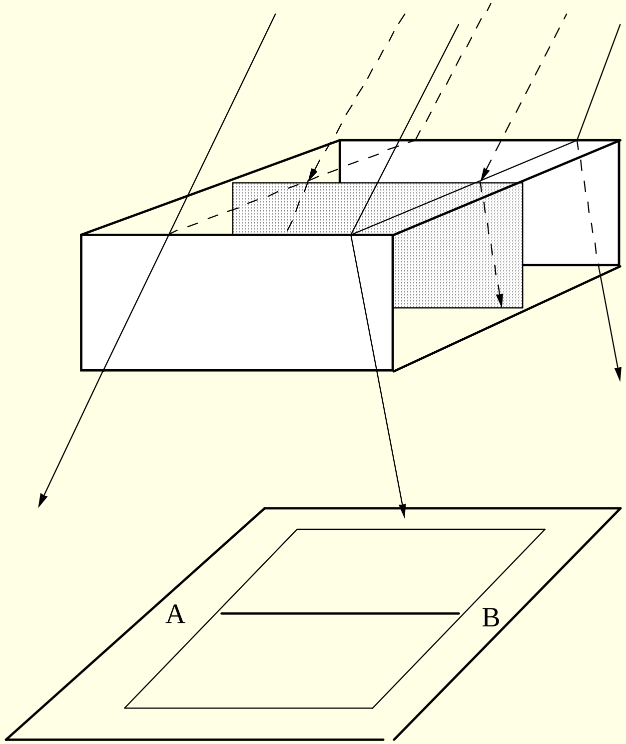
They may be curved if the deformation is small enough

Less intensity

Dynamical theory

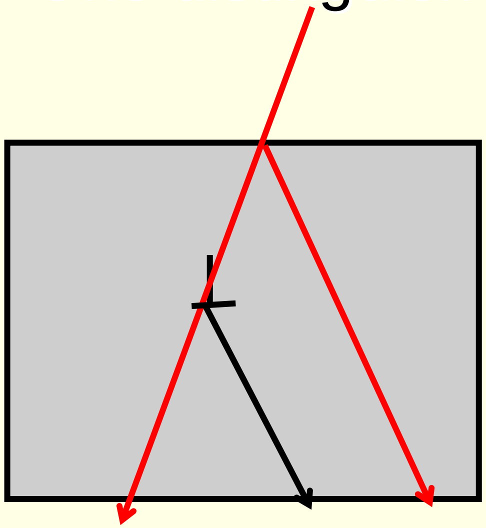
All the wave-fields interfere and create a complex contrast related to the deformation.

The image is made of the contribution of the wave-fields in each plane of incidence.



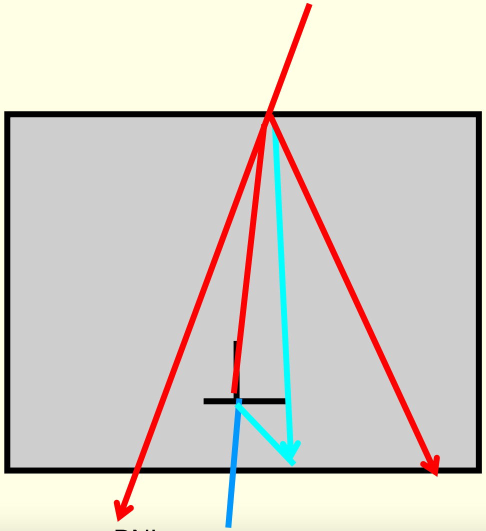
Dynamical theory

One distinguished three parts in the image of a defect:



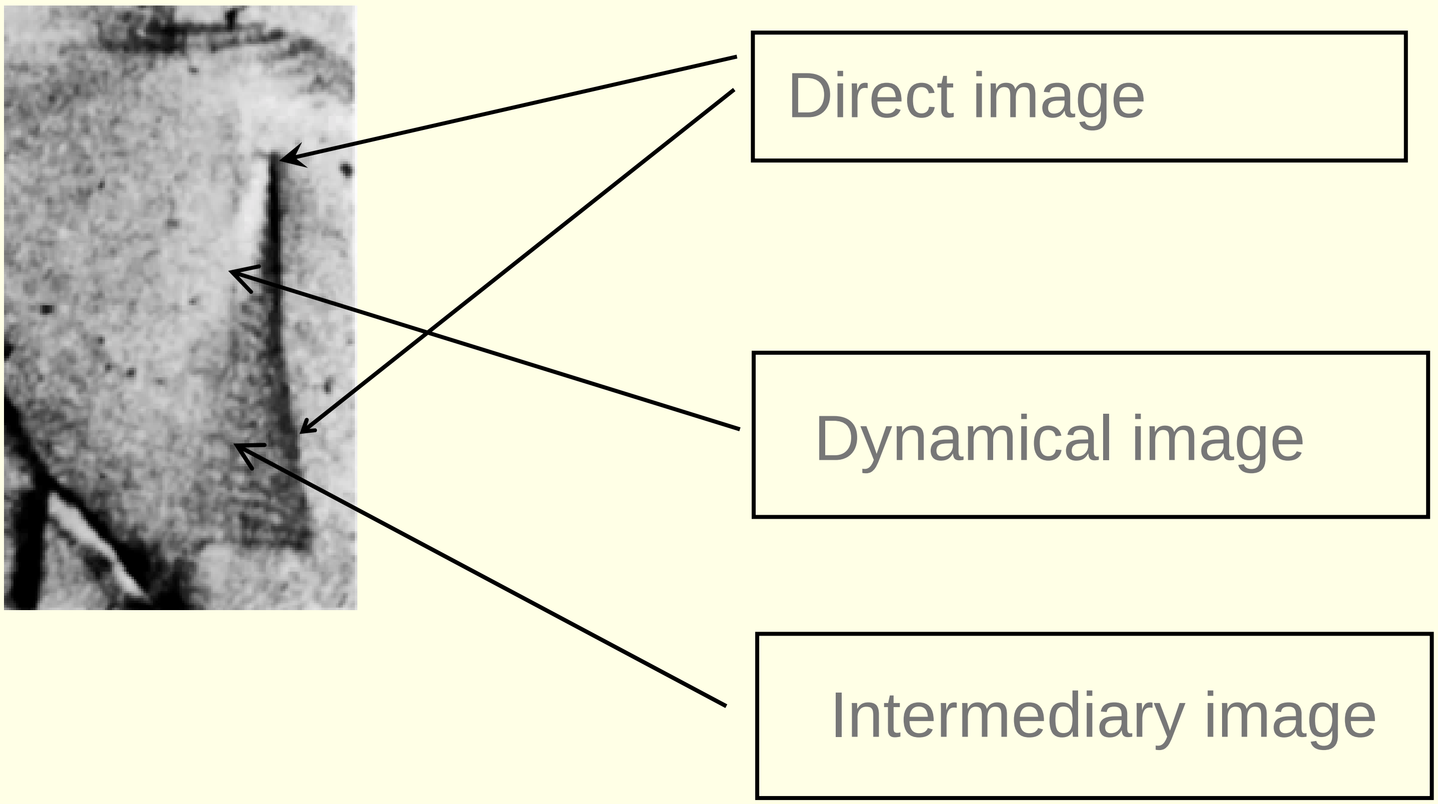
The direct image when the defect intersects the direct beam

The dynamical image (less intensity)



The intermediary image

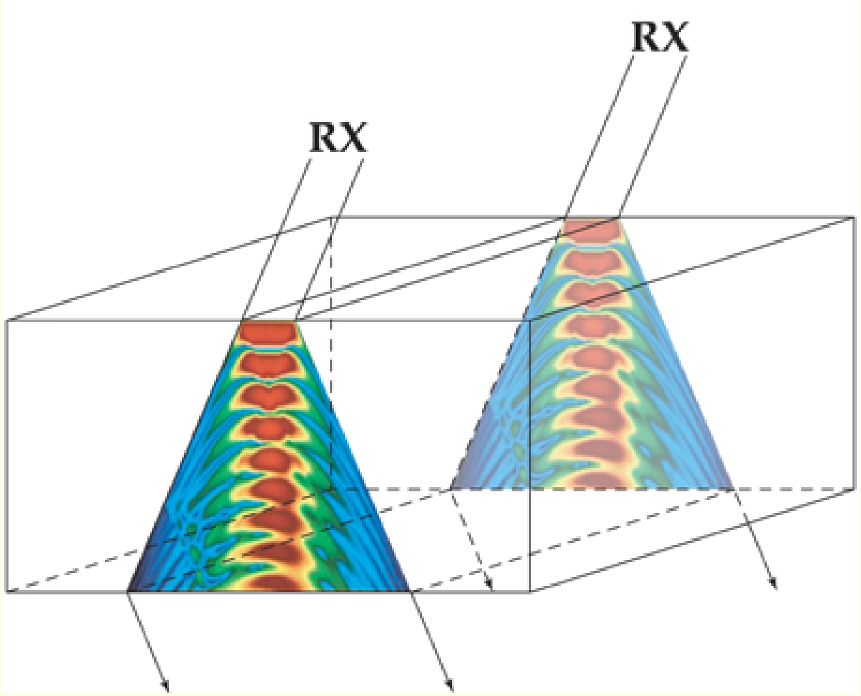
X-Ray topography



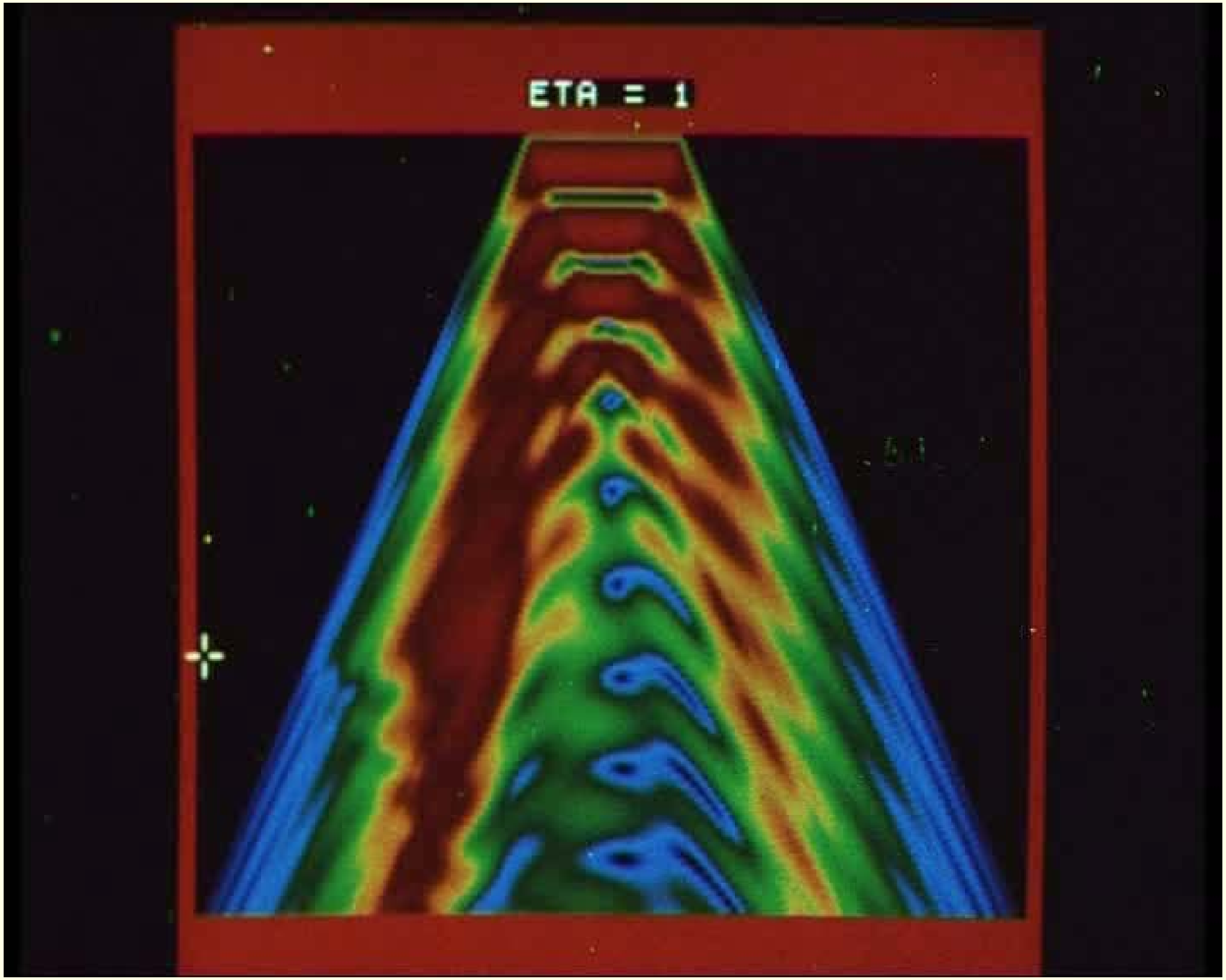
Simulation

Paths of wave - fields:

- In one incidence plane
- Dislocation moves from left to right
- Hot color means high intensity
- Cold color (blue) means low intensity



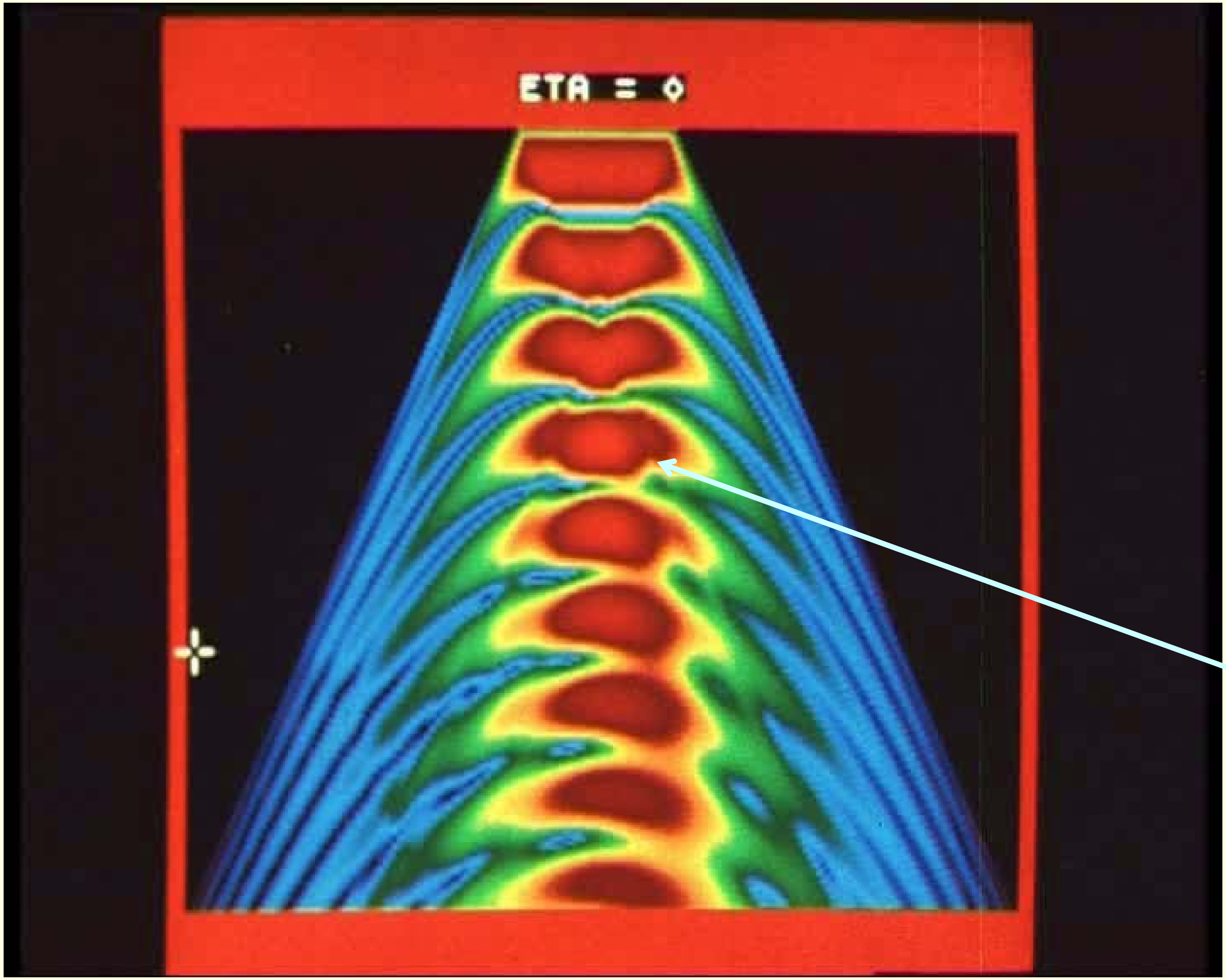
X-Ray Dynamical theory



Plane wave case at half maximum

X-Ray Dynamical theory

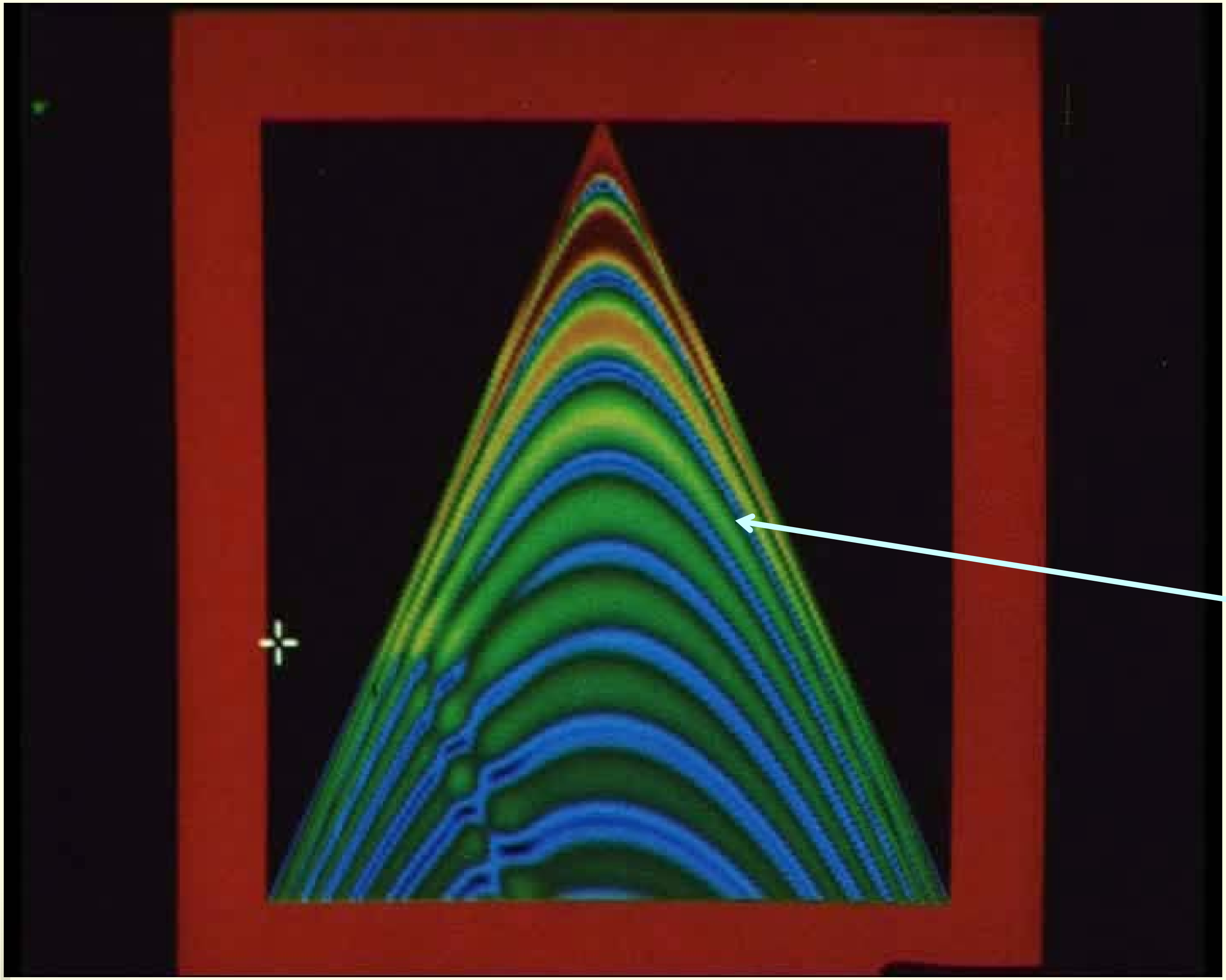
X-Ray Dynamical theory



Plane wave case at exact Bragg condition

Pendellösung fringes

X-Ray dynamical theory



Incident point source

Extinction fringes

X-Ray Dynamical theory

- See “Dynamical Theory of X-Ray Diffraction”
  - André Authier, Oxford Science Publications 2001

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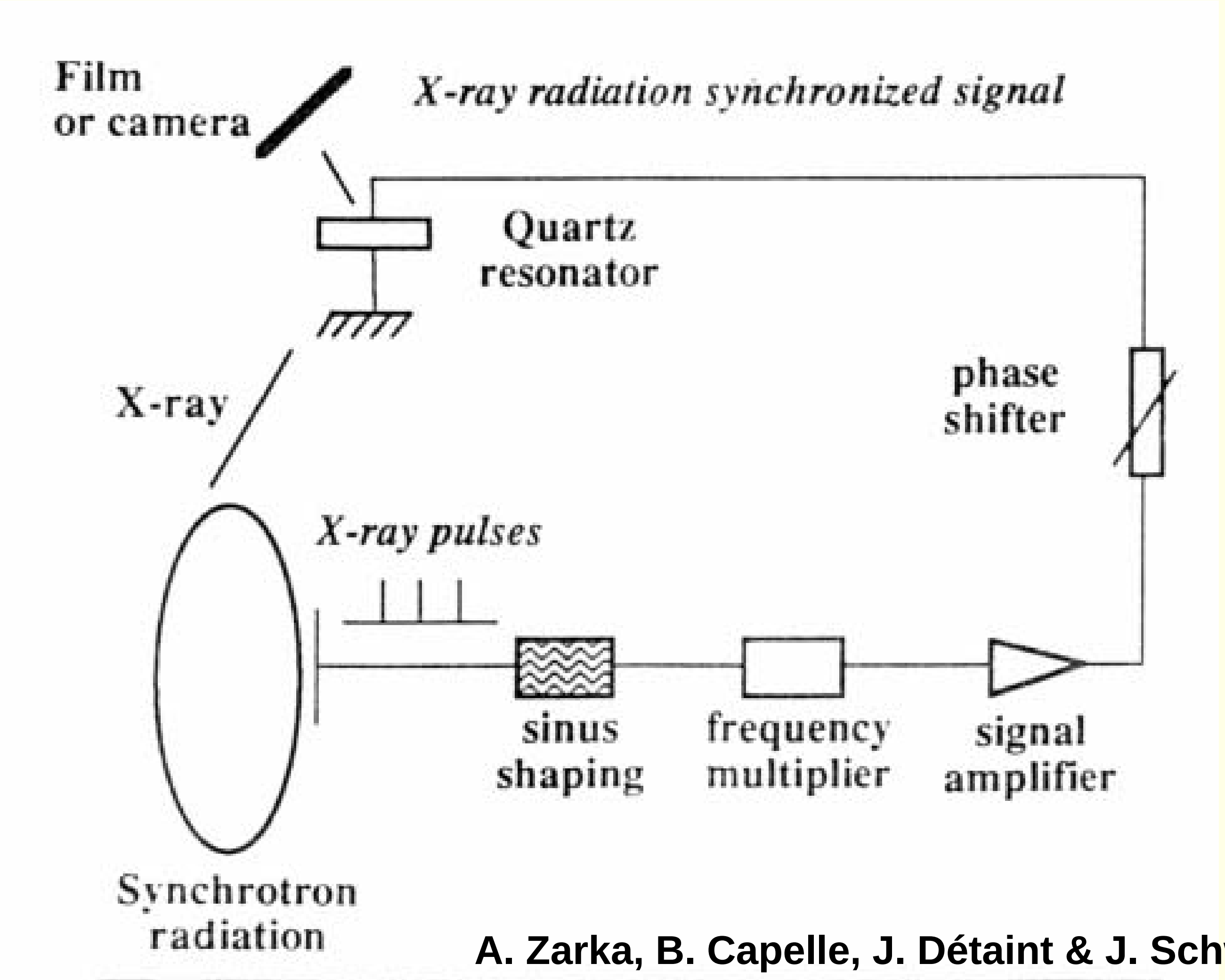
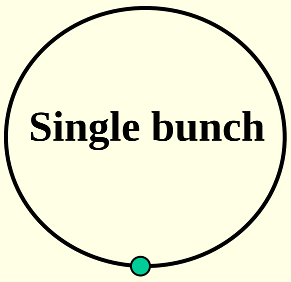
Stroboscopic topography

- Work performed at LURE and ESRF

  B. Capelle, J. Détaint & Y. Epelboin

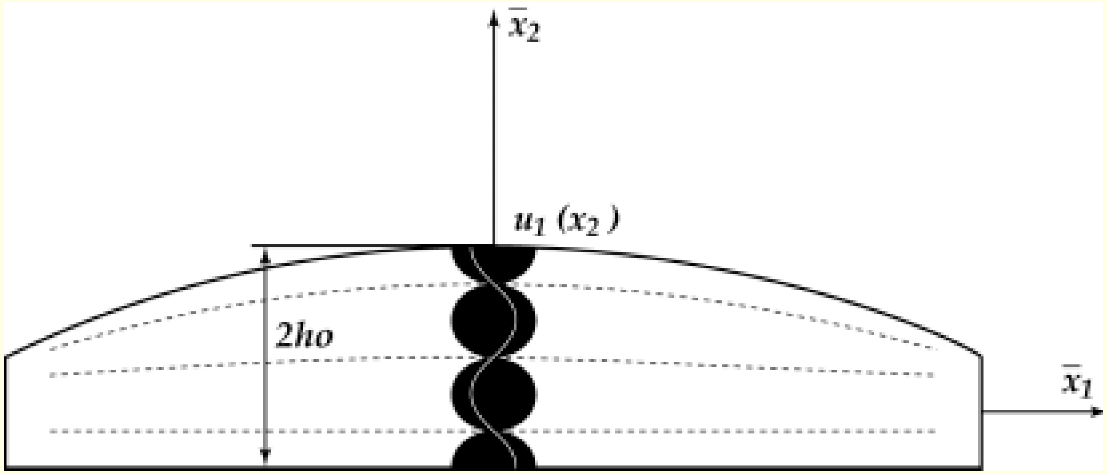
Stroboscopic topography





A. Zarka, B. Capelle, J. Détaint & J. Schwartzel
1987, Proc. 41th Freq. Contr. Symp. 236

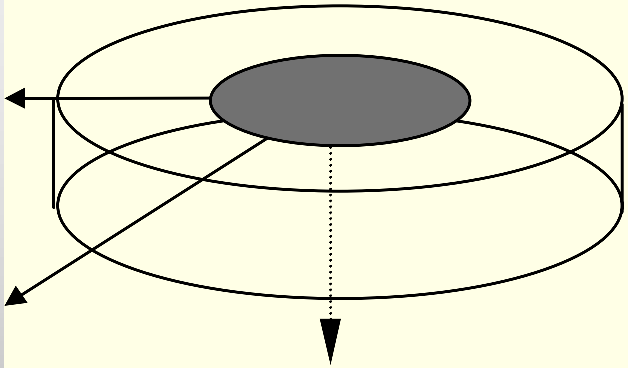
Ultra-waves Characteristics



Study of shear modes:

- Vibration parallel to the X_1 axis.
- N overtone: number of half acoustic wavelengths in the thickness.
- More than one resonant vibration for a given overtone.

Piezoelectric devices

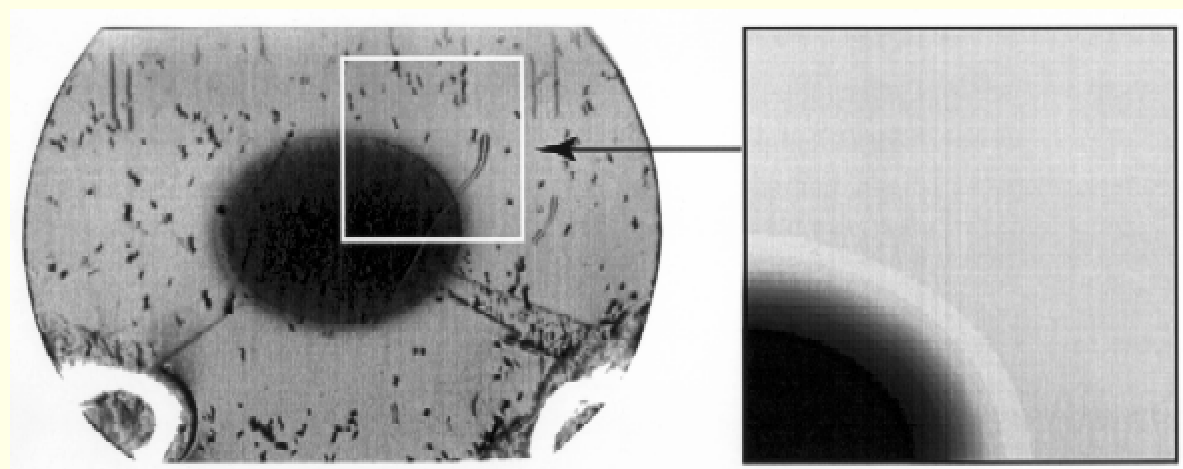


Circular plano-convex devices with elliptical electrodes

λ = 0.076 nm,

$\bar{2}1.0$ reflection ,

mode 1

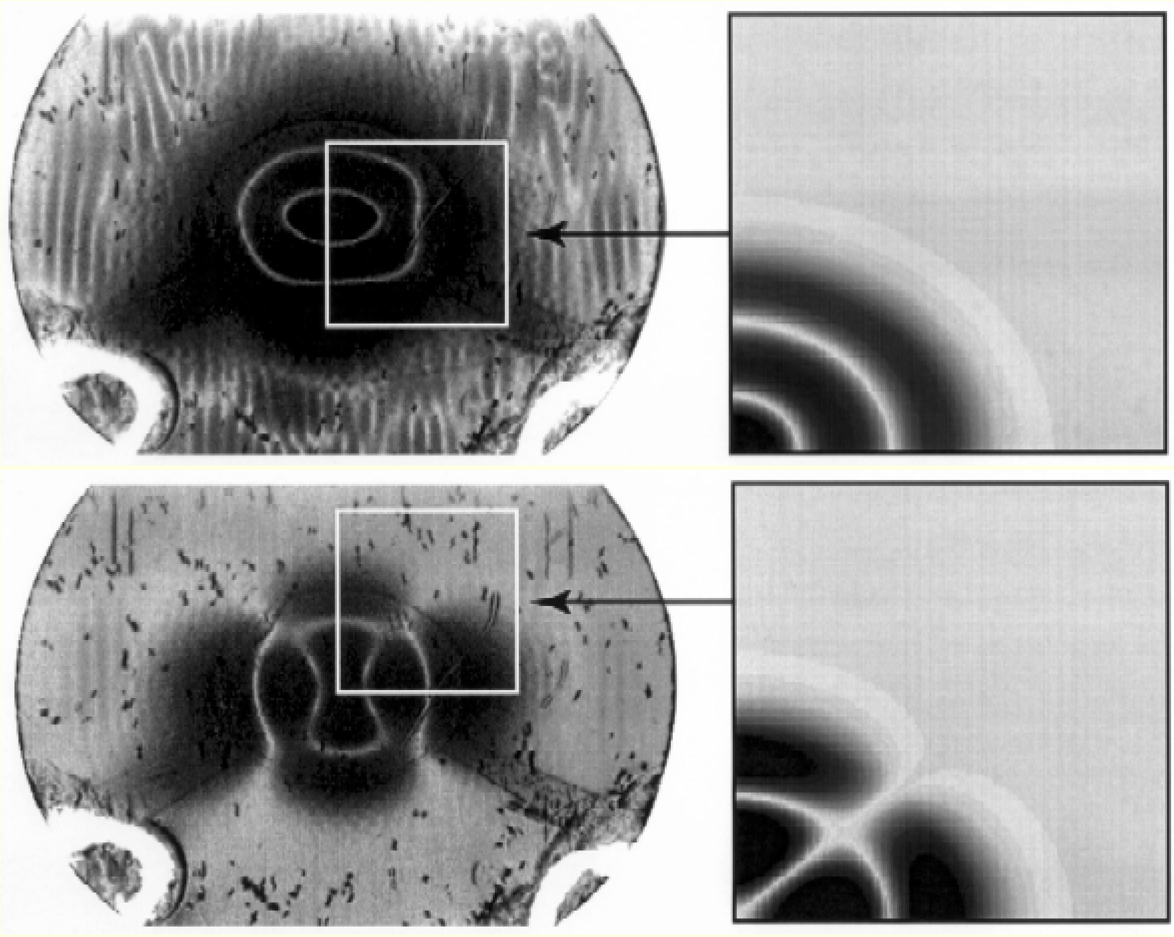


Piezoelectric devices ...

λ = 0.076 nm,
$\bar{2}1.0$ reflection ,

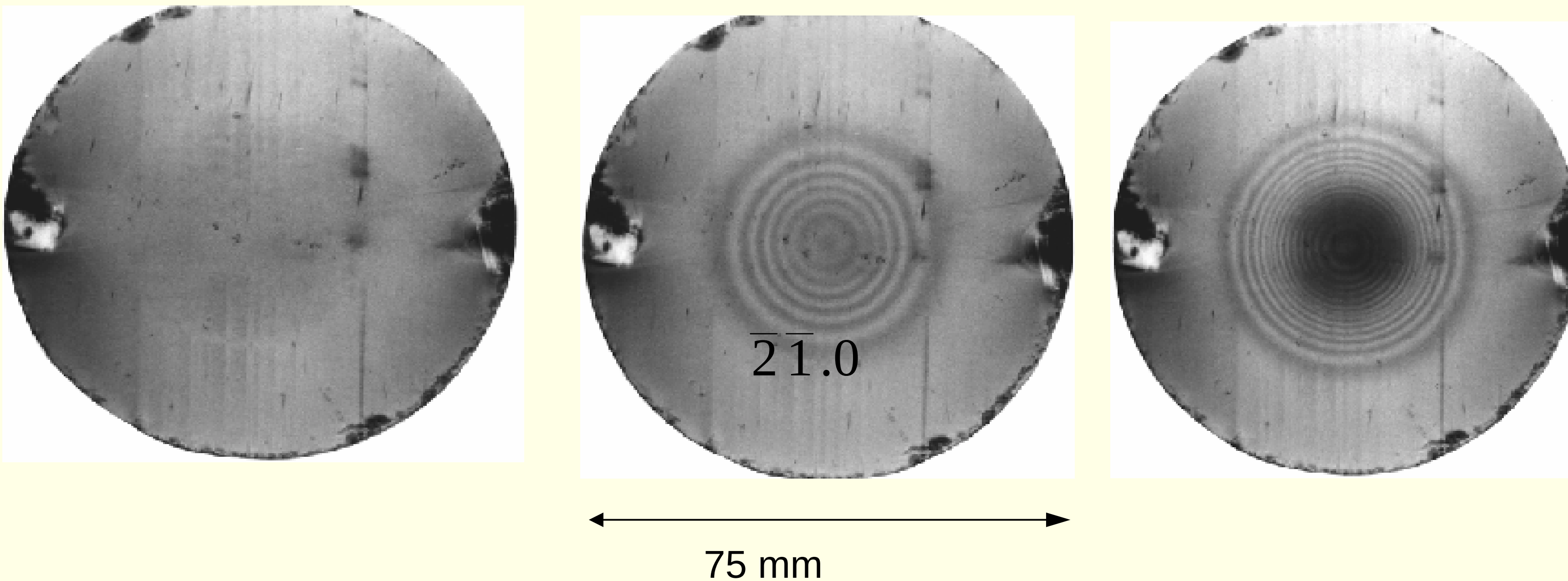
mode 3

mode 5



Y. Epelboin, J. Détaint & B. Capelle, J. Appl. Cryst. 1998, 31, 574

Influence of Voltage

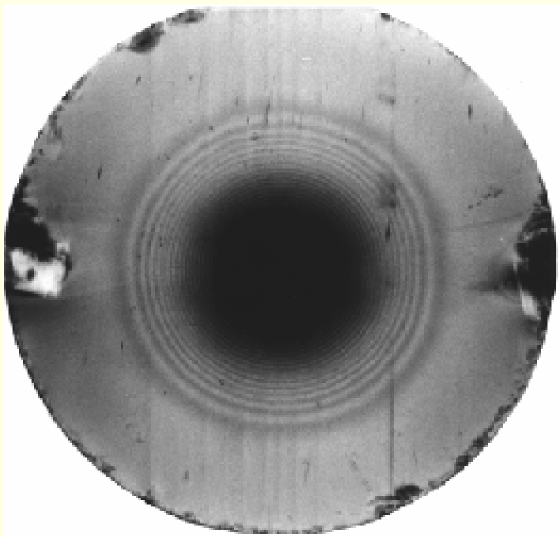


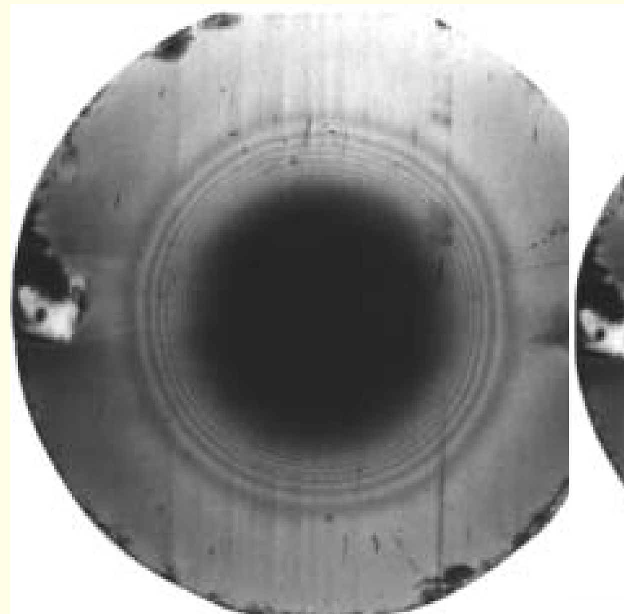
No vibration

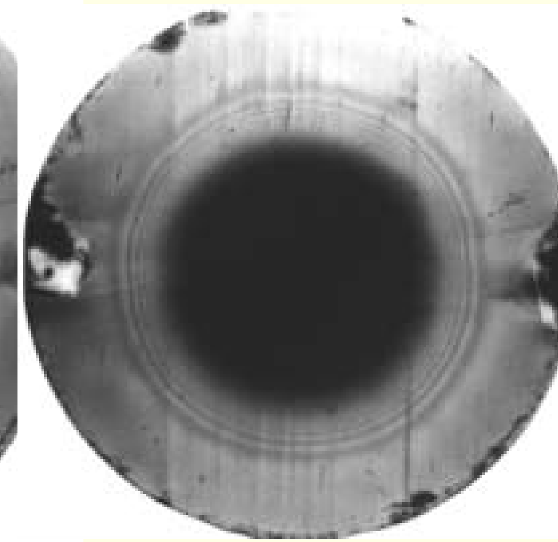
Increasing u_1 vibration

$\bar{2}\bar{1}.0$ reflection $\lambda \approx 0.07\,nm$

Influence of Voltage





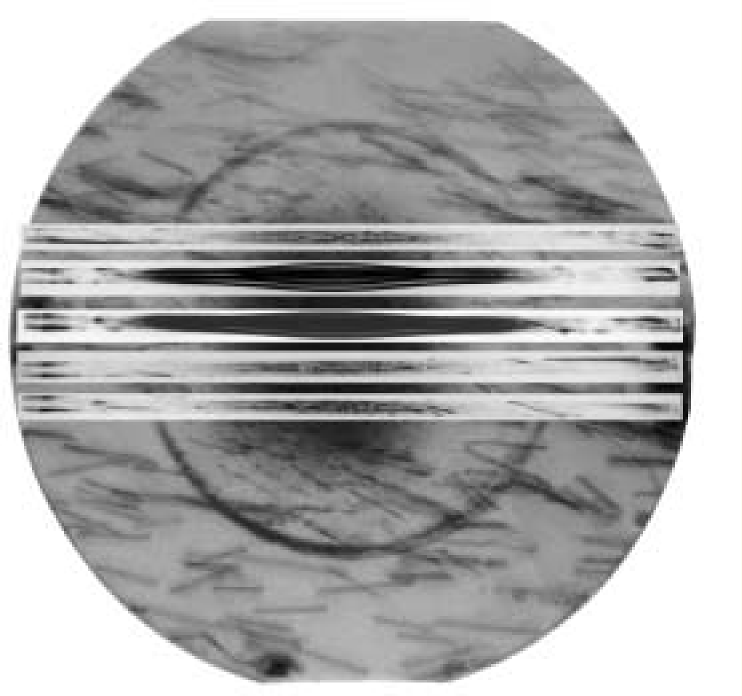


175 mm

The amplitude is maximum in the center and decreases toward the edges

Section Topographs

Section Topographs



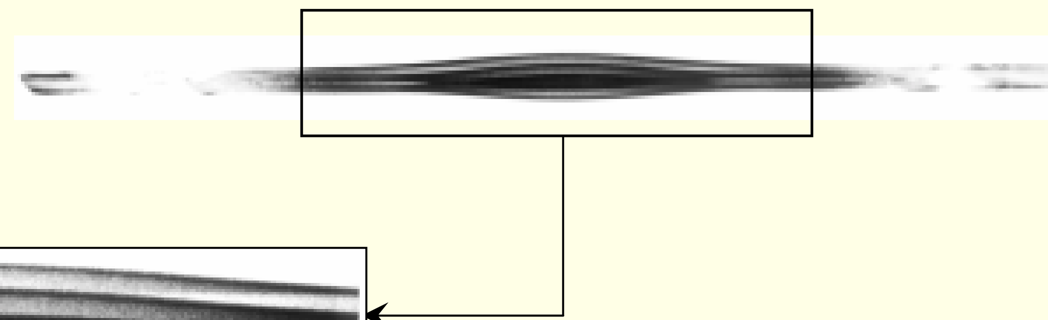
Section topographs present special features

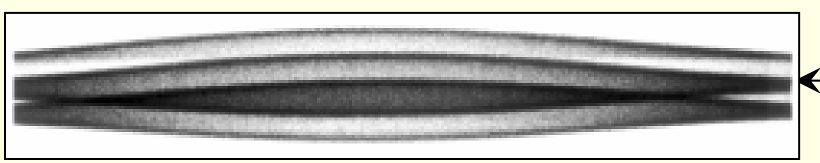
Experiment at ESRF

The image changes when the crystal to film distance changes

~ 5 cm

70 cm

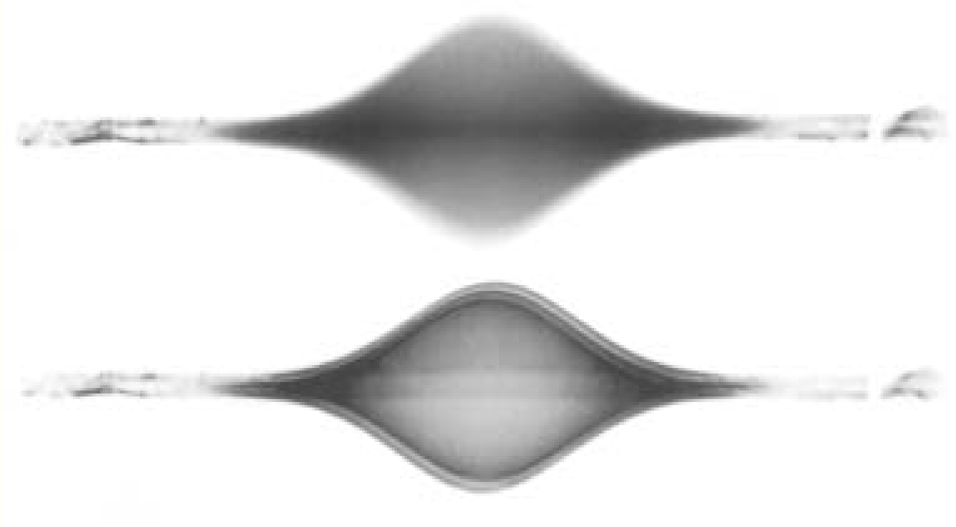




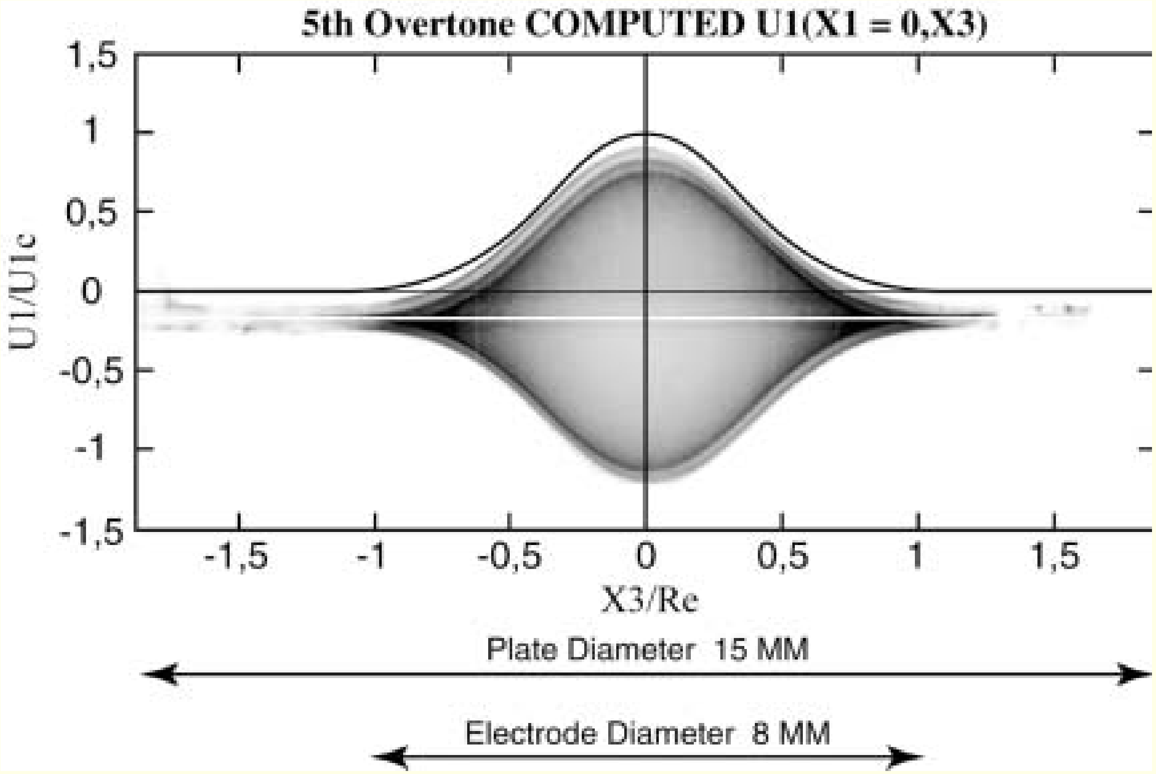
The fringes are shaped as a « muscle »!

Experiment at ESRF (…ctd)

This effect can be observed in stroboscopic images only



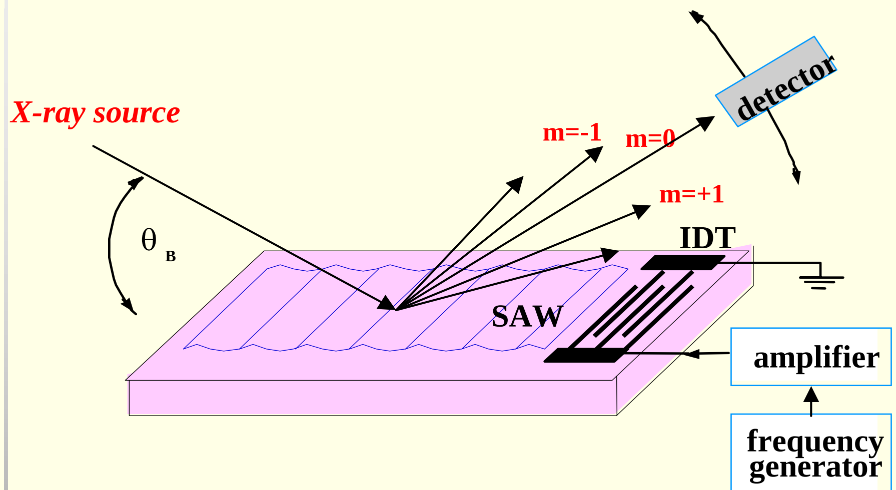
Quantitative analysis

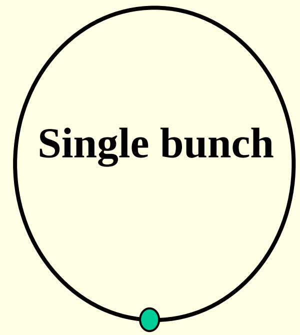


The comparison with a theoretical curve qualitatively fits with the theory

B. Capelle, J. Détaint & Y. Epelboin JAC 2001, 34, 625

Study of surface waves

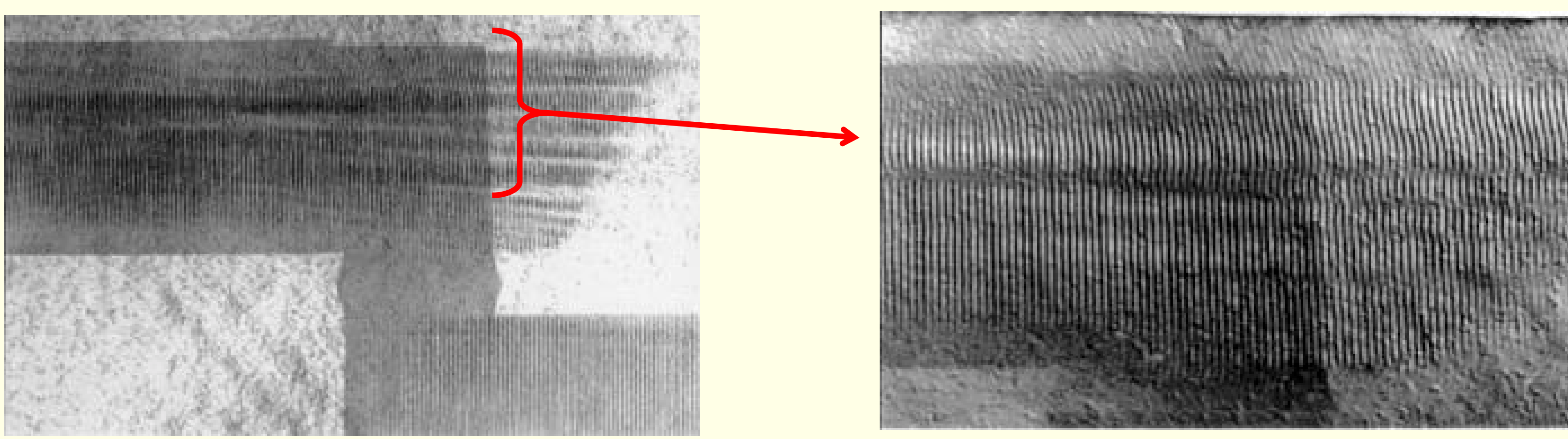




ν = 9.507 Mhz, $\lambda_R/2$ = 166 μm

LURE: B. Capelle, J. Détaint 1996

Good agreement with simulations V. Mocella & Y. Epelboin, JAC 1999, 154



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- Heating of monochromators

Characterization of Biological Materials

Work performed at ESRF:

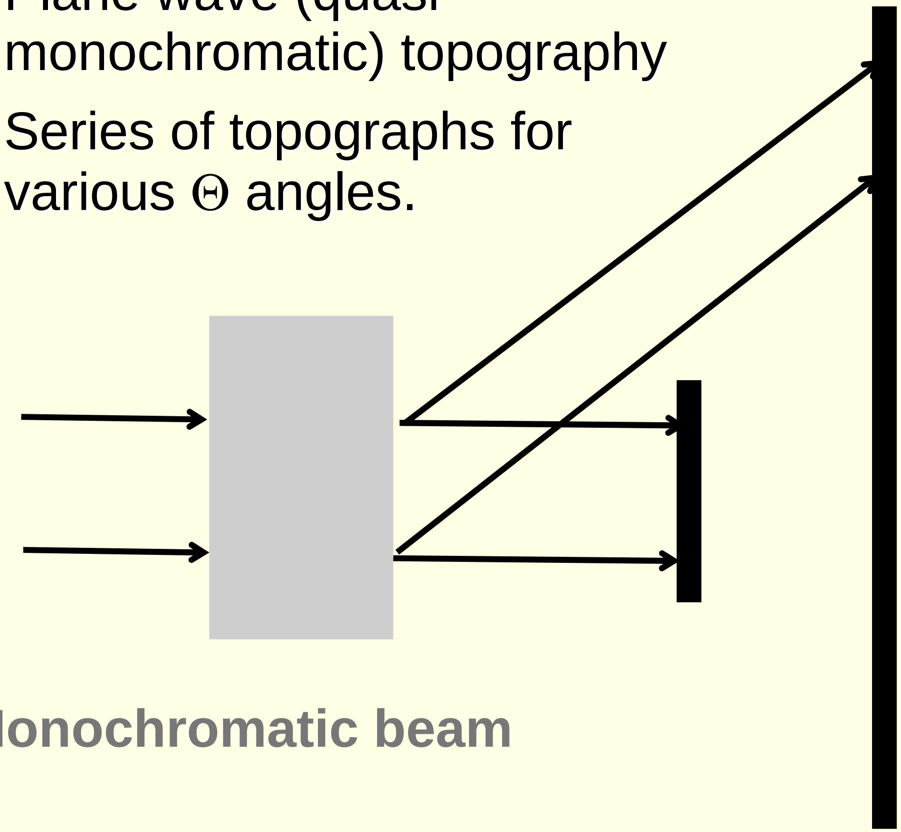
- B. Capelle (LMCP, Paris)
- Y. Epelboin (LMCP, Paris)
- J. Härtwig (ESRF)
- F. Otalora (Granada, Spain)
- V. Stojanoff (NSLS, USA)

Question:

- How is the determination of the molecular structure of a protein affected by the defect structures in the biomolecular crystal?
- Do dislocations exist in the crystals?

Biological materials

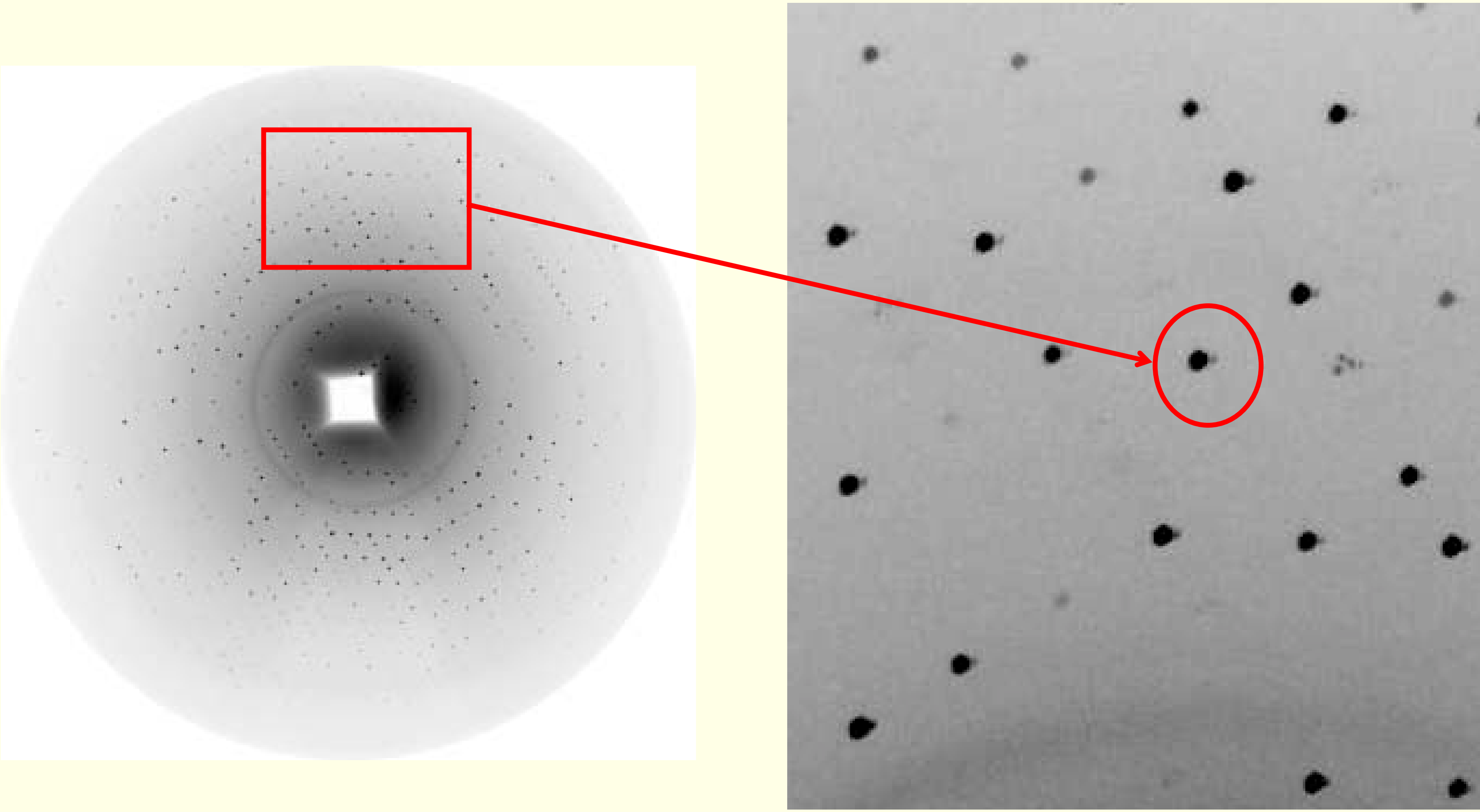
- Plane wave (quasi monochromatic) topography
- Series of topographs for various Θ angles.



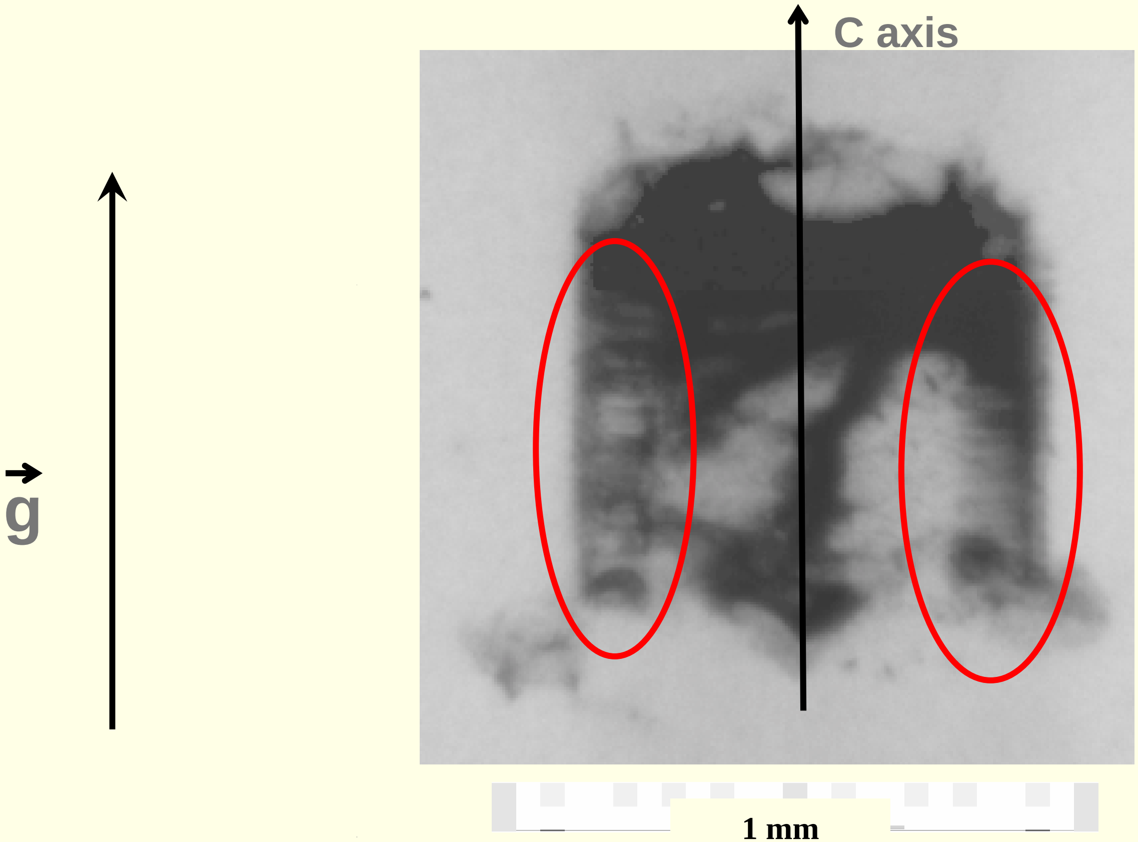
Film or image plate

Monochromatic beam

Biological materials



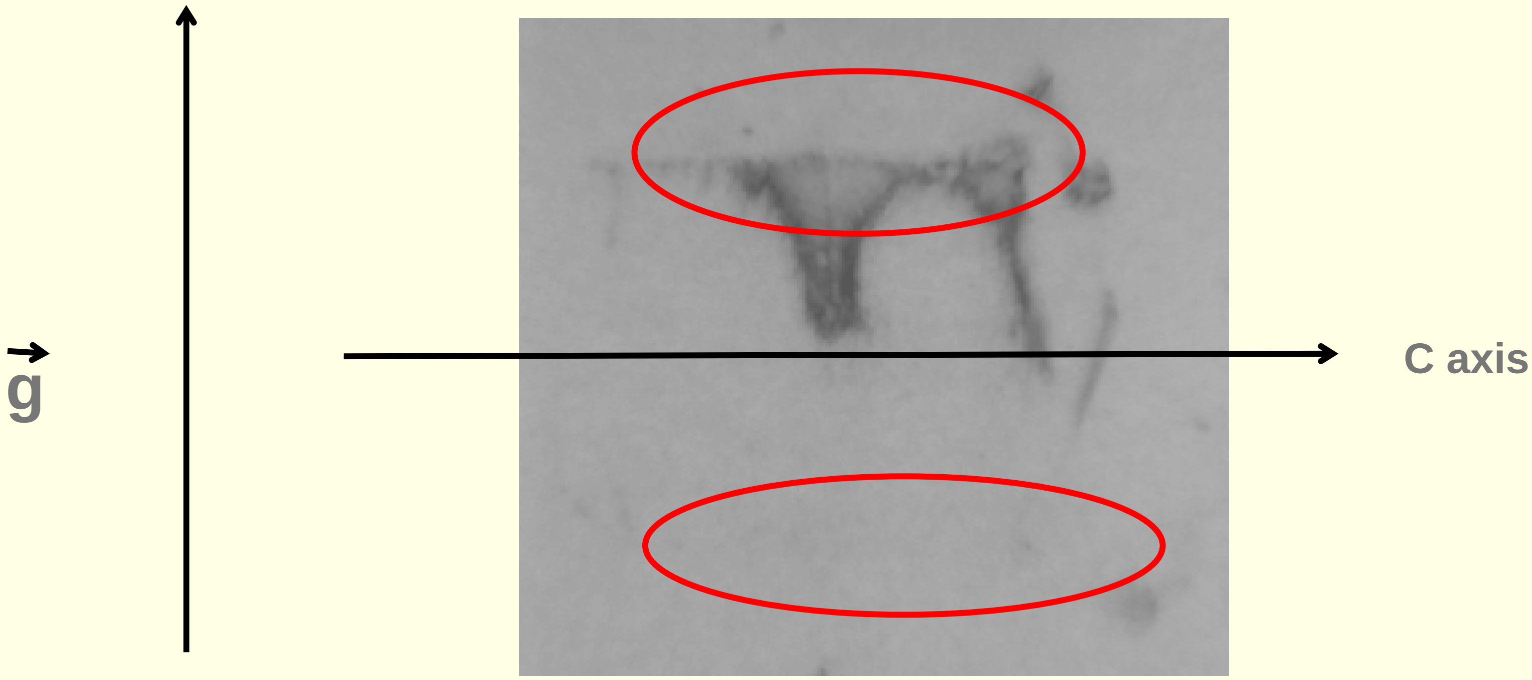
Visibility of dislocations



Dislocations?

HEWL batch grown in gel media; λ=0.81Å

Visibility of dislocations



Poor visibility of lines!

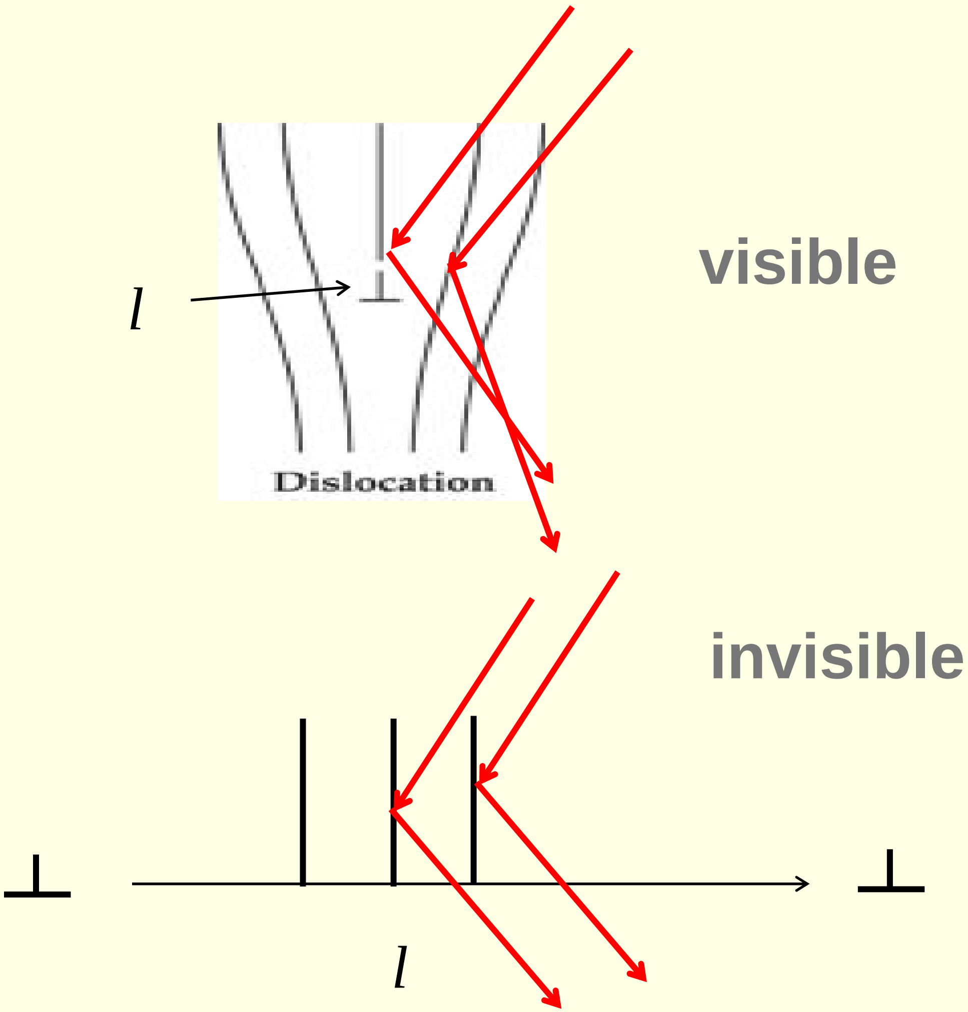
Visibility of dislocations

Rule:

$$\left(g, b, l\right) = 0$$

or (simplification)

$$g.b = 0$$



Visibility of dislocations

Thus if these linear contrasts are dislocations:

- Bürgers vector parallel to c axis (A4)
- Lines perpendicular to c axis

They are edge dislocations

Is this acceptable from the point of view of

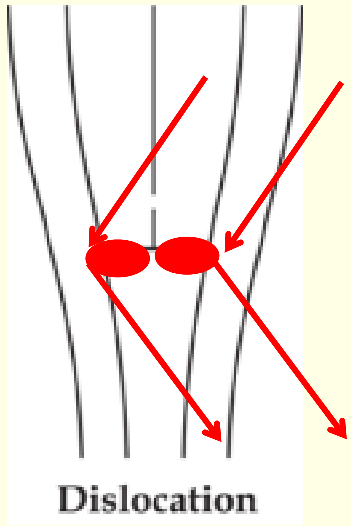
•crystal growth?

•Dislocation visibility?

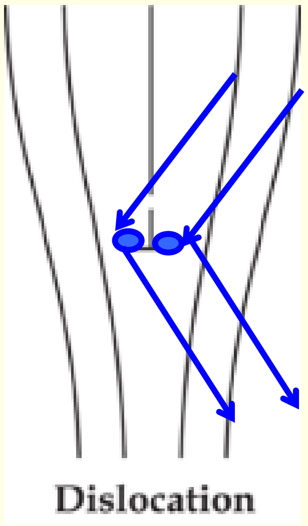
Dislocations

Width of dislocation images proportional to:

- Length of Bürgers vector
- 1/ΔΘ, ΔΘ being the departure from exact Bragg angle



Near Bragg peak:
large image

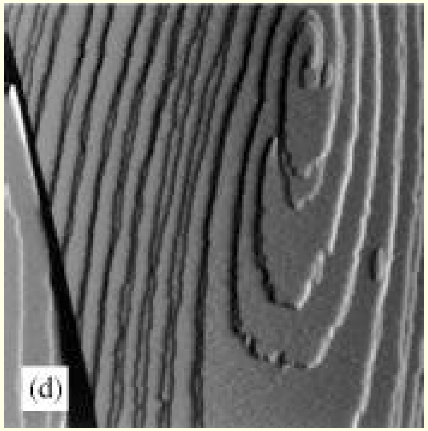


Weak beam condition:
small image

Dislocations

- Dislocations can be distinguished only far from Bragg condition
- Near the peak all contrasts mix together to show only a black area

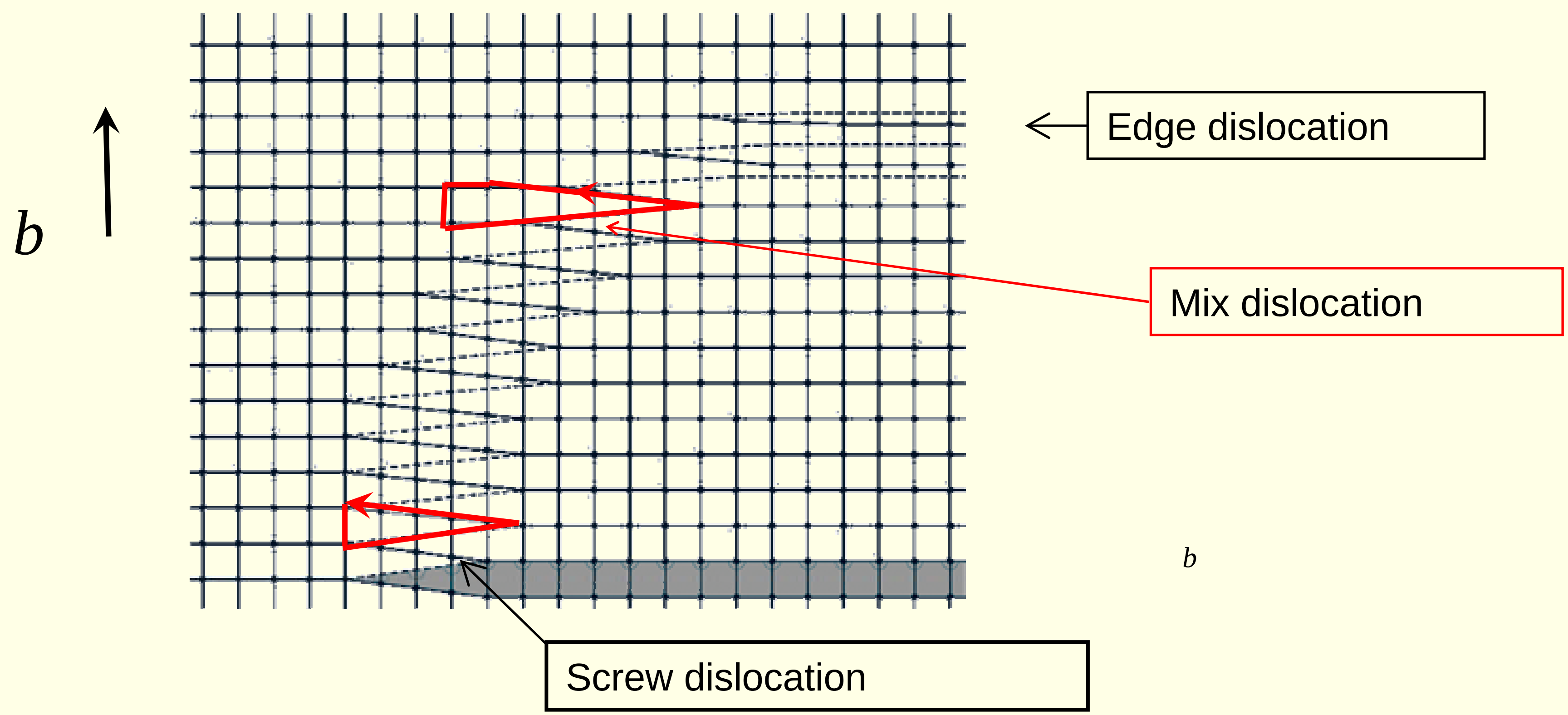
- Why do people see spiral loops in surface studies (AFM…)?



Thaumatin crystal 12x12 μm^2

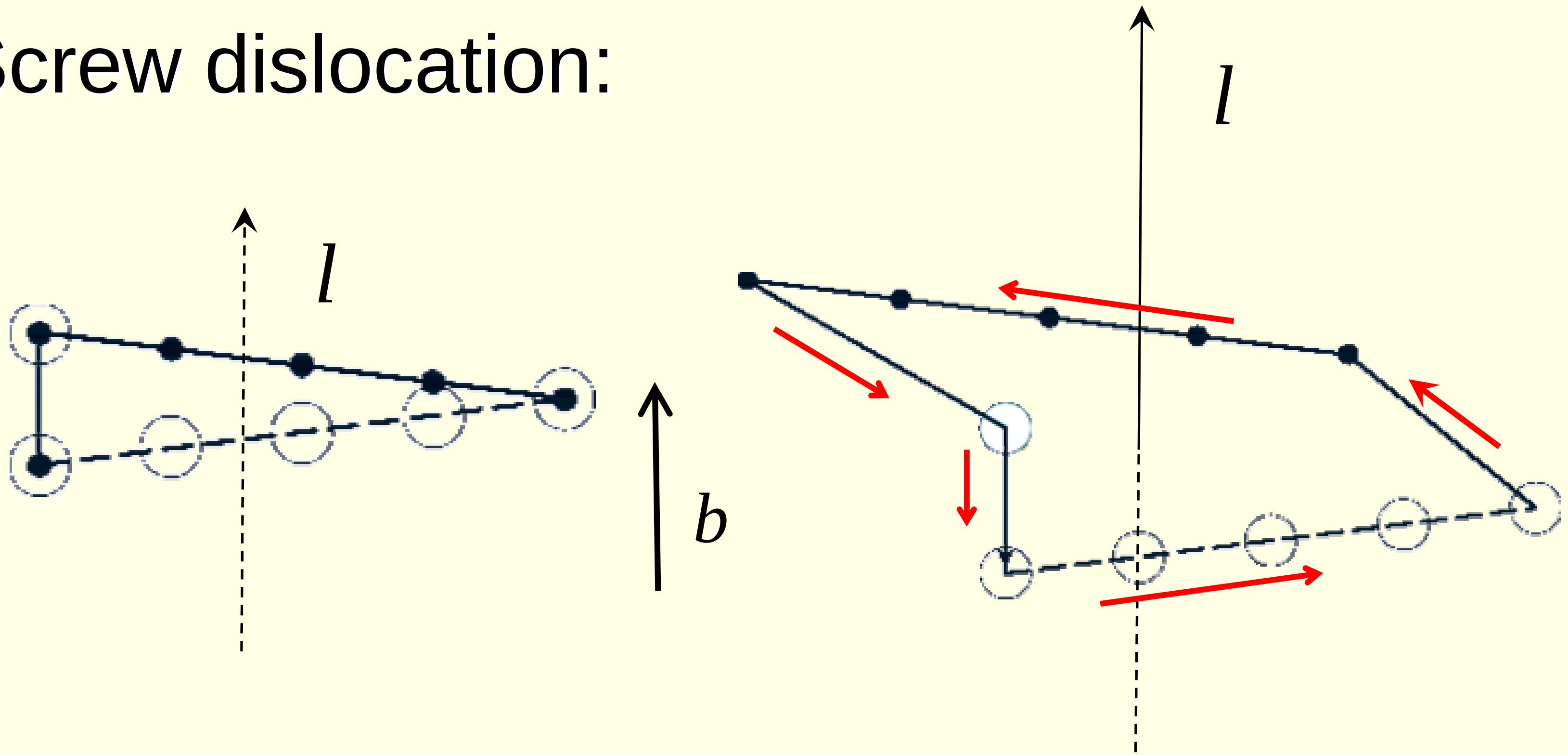
Malkin & al. J. Cryst. Growth 1999, 471

Nature of dislocations



Nature of dislocations

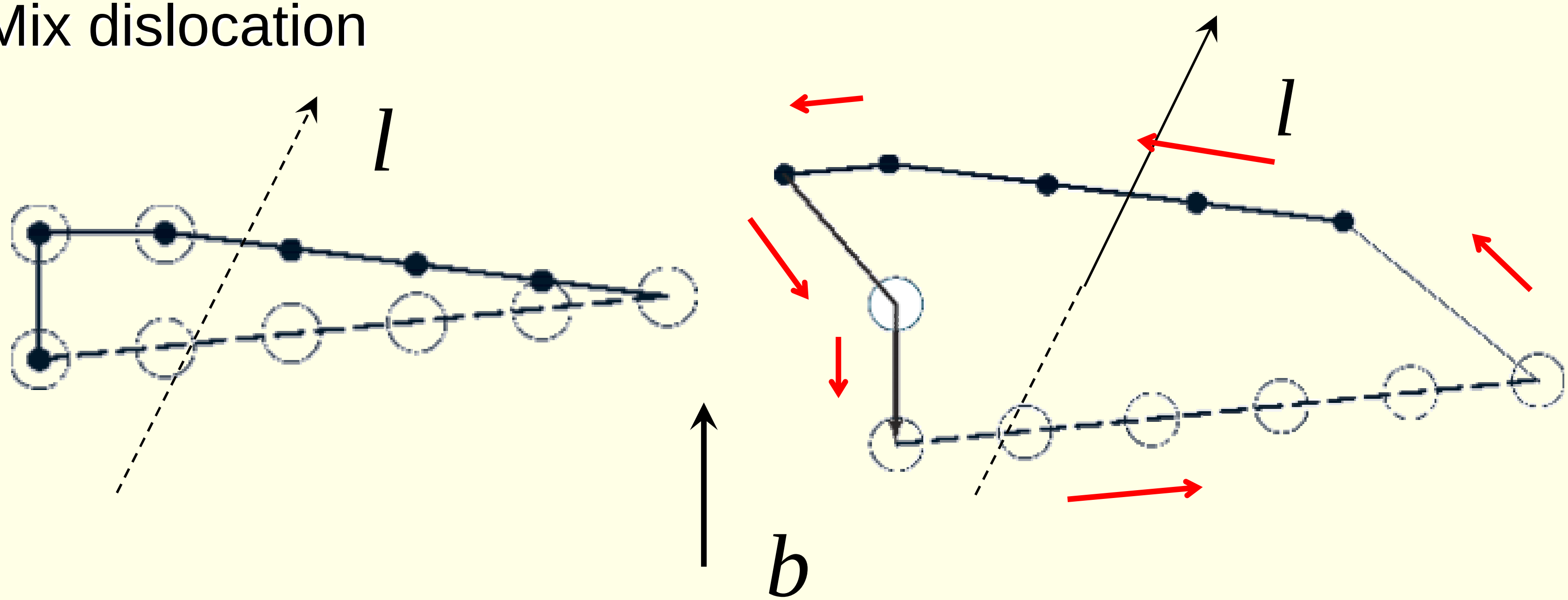
Screw dislocation:



A screw dislocation appears as a spiral on a surface perpendicular to the line.

Nature of dislocations

Mix dislocation



A mix dislocation also appears as a spiral, inclined on one side, on a cross surface

Nature of dislocations

Surface studies can seldom determine the nature of a volume defect.

They cannot determine the nature of a dislocation.

Dislocations

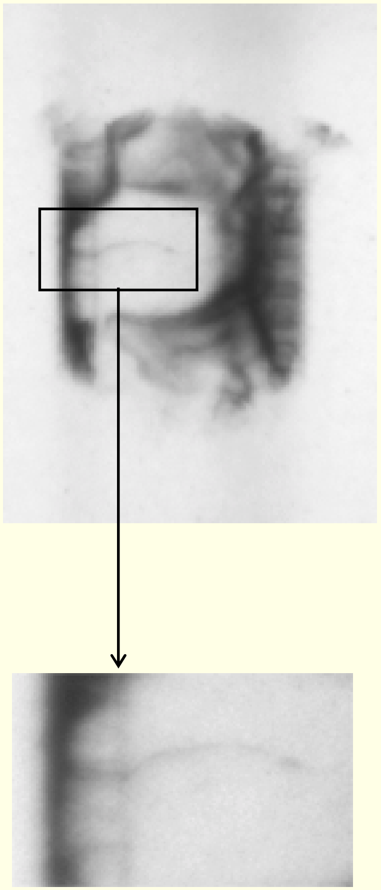
- Edge dislocations are common in hydrothermal growth:
  - Growth speed improvement: 1 – 2
  - More edge than screw dislocations

F. Lefaucheux, M-C. Robert & A. Authier J. Cryst. Growth (1973), 329 (in french)

S. Gits-Léon, F. Lefaucheux & M-C. Robert, J. Cryst. Growth (1978), 345

Dislocations

- Dislocations exist in HEWL hydro thermally grown crystals
- Features resemble to the known ones in inorganic hydro thermally grown crystals
- There are good reasons to think that biological crystals resemble to inorganic crystals from the point of view of crystal growth



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Heating of monochromators

- V. Mocella (ESRF & APS)
- W. K. Lee (APS)
- G. Tajiri (APS)
- D. Mills (APS)
- C. Ferrero (ESRF)
- Y. Epelboin (LMCP-UPMC, Paris)

Study of a cryogenically cooled double crystal monochromator

(submitted to J. Appl. Cryst.)

Methodology

Calorimetry and rocking curve measurements

↓

Finite elements modeling of thermal load and related strain

↓

Simulation with Takagi - Taupin equations

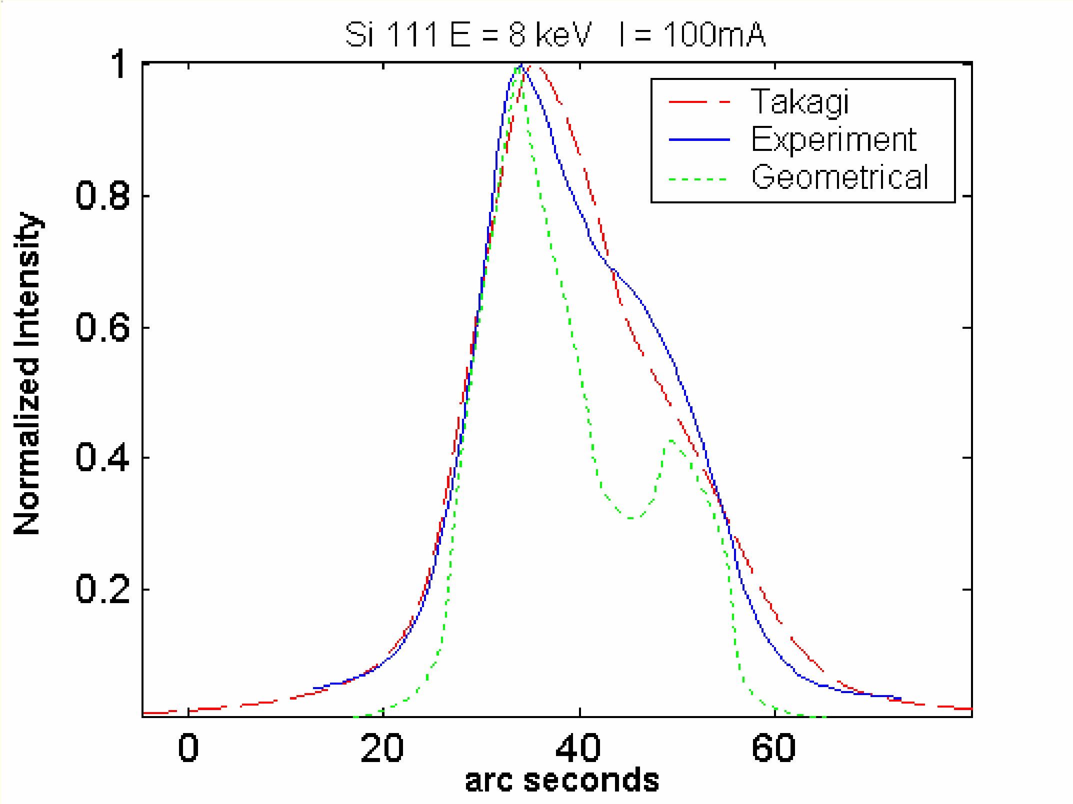
↓

Comparison between simulation and experiment

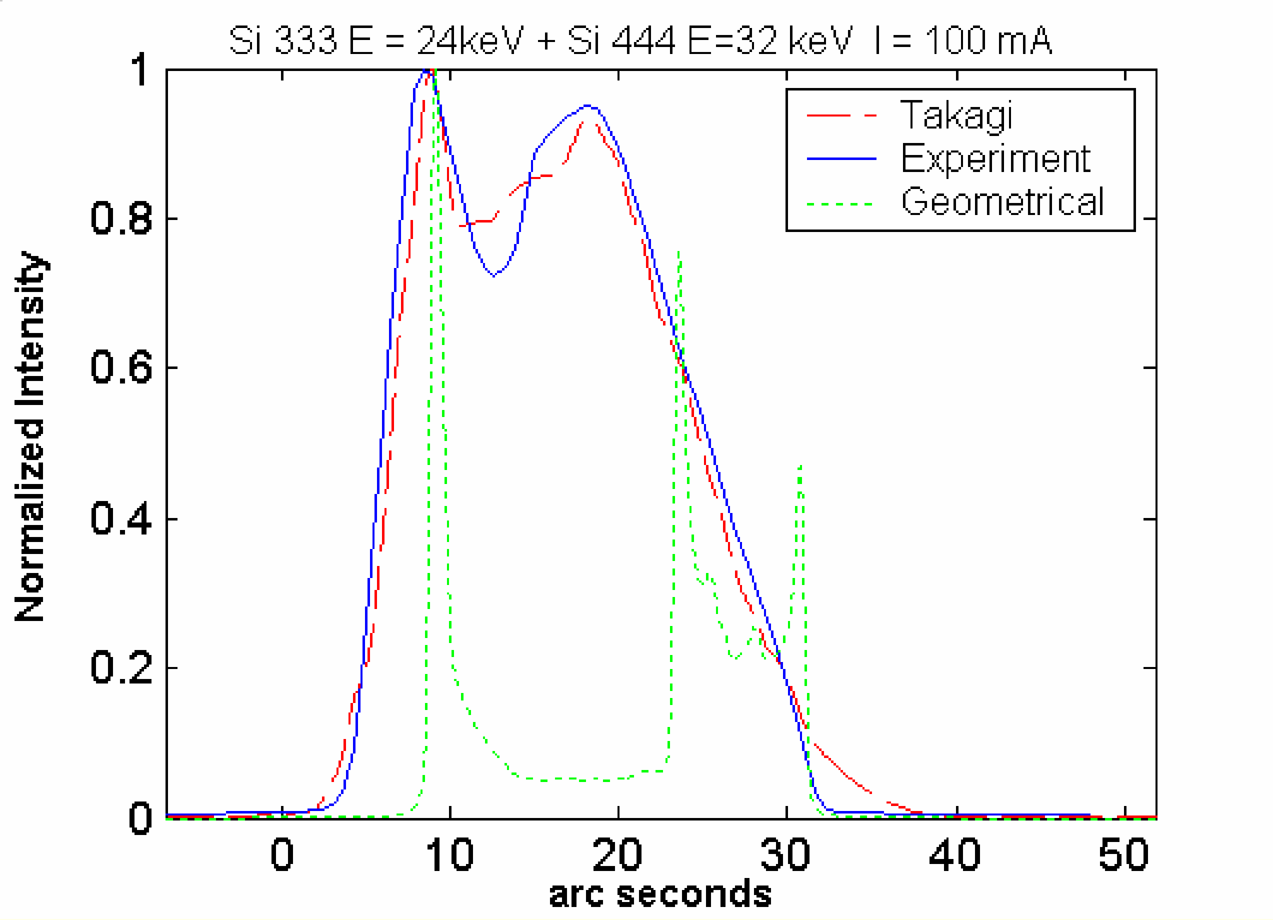
Si 111, E = 8 keV, I = 100 mA

Crystal dimensions 85 mm x 50 mm x 35 mm

(Lee, W. K., Fernandez, P. & Mills, D. M. (2000). *J. Synch. Rad* **7**, 12)



Si 333, E = 24 keV + Si 444 E = 32 keV, I = 100 mA



Results

- Simulations match very well the experiment
- Discrepancies may be explained by:
  - The approximation of the FEA heat load model
  - Inaccurate spatial beam distribution in the simulation
- A geometrical approach is satisfactory only when the X - Ray penetration is negligible:
  - Low intensity
  - Low energy

Thanks

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http://www.lmcp.jussieu.fr/~epelboin