LAUE MICRODIFFRACTION

French CRG beamline BM32 “InterFaces” @ ESRF

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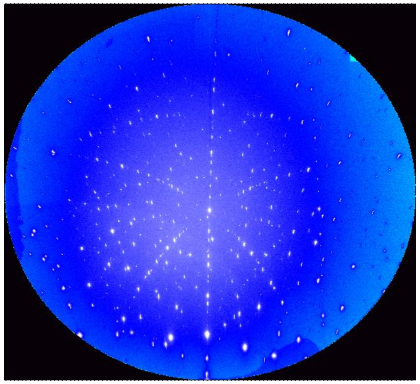
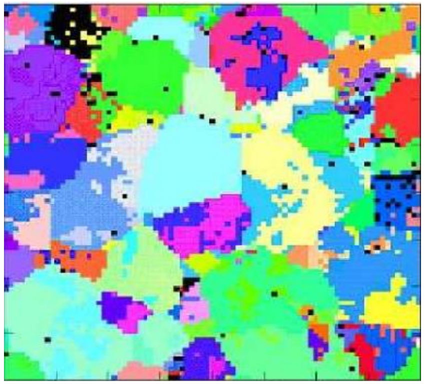
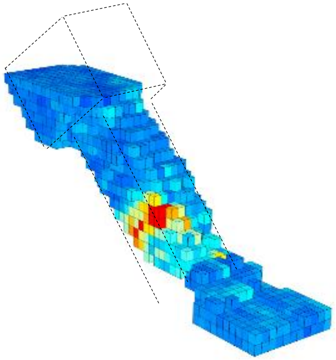


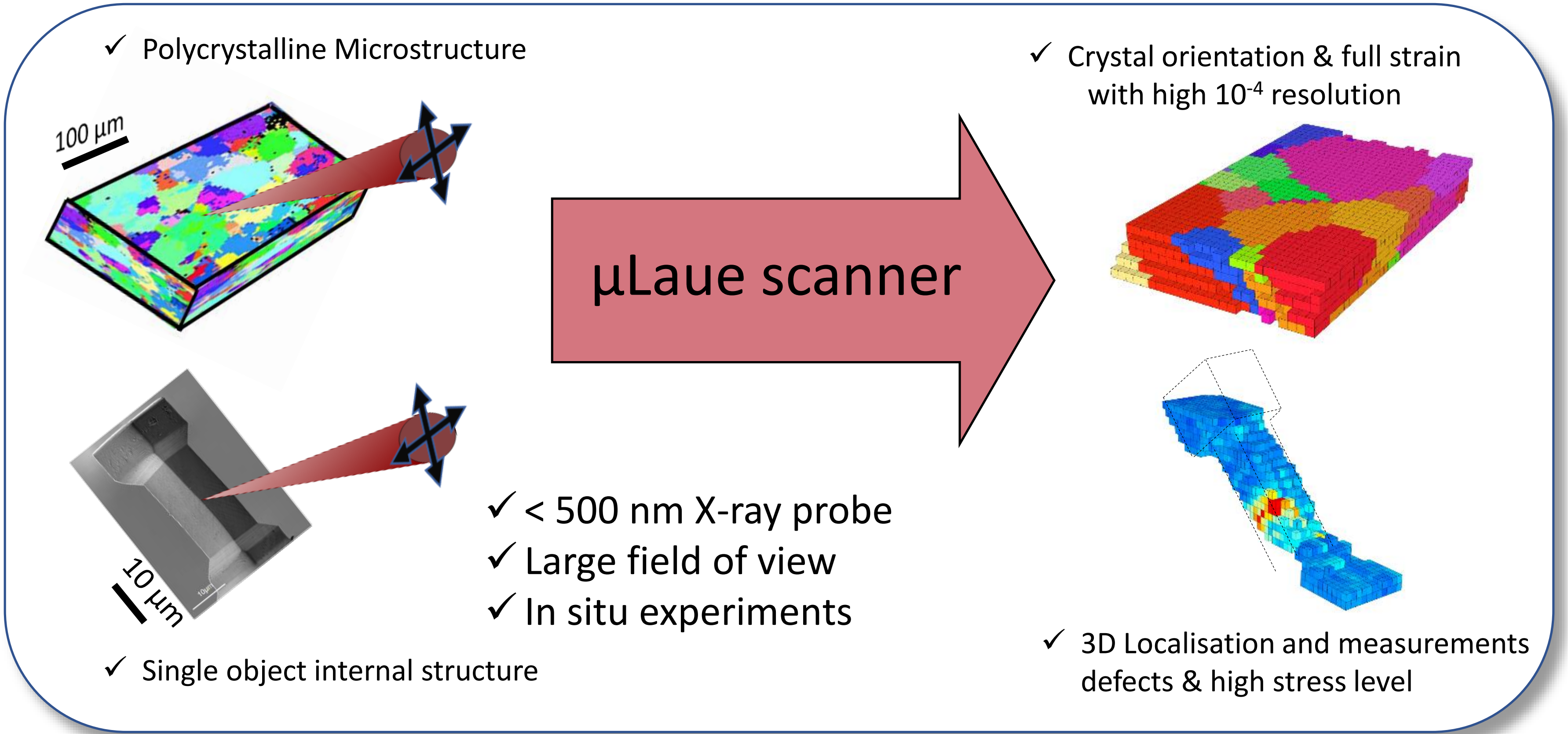




HERCULES Tutorials March 2020

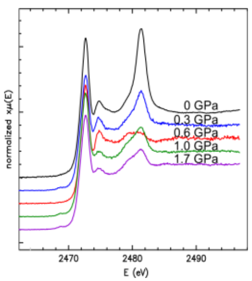
X-ray Laue diffraction microscope for structural imaging

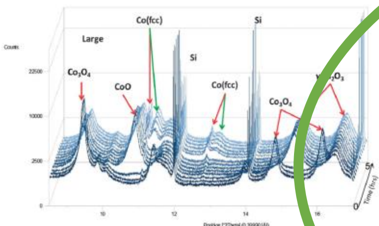
on CRG-IF beamline at ESRF, Grenoble, France

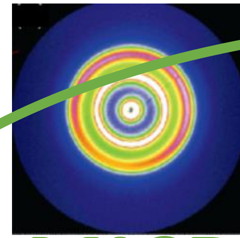


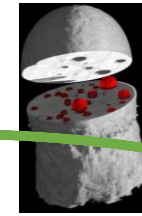
Materials x-ray based characterization techniques

- Multi-scale approach
- Multi-modal analysis









X-ray Spectroscopies

Atom

- *Fine chemical analysis*
- *Composition*
- *Physico-chemical properties*

X-ray Diffraction

Crystal

- *Microstructure*
- *Residual strain*
- *Phase recognition*

X-ray Scattering

Material

- *Morphology*
- *Structure*
- *Defects*

2D/3D X-ray Imaging

Final object

- *Failure analysis*
- *Compliance*
- *Reverse engineering*

LAUE MICRODIFFRACTION

- Coherence → resolution and contrast
- Hard X-rays → penetration (in-situ, operando)

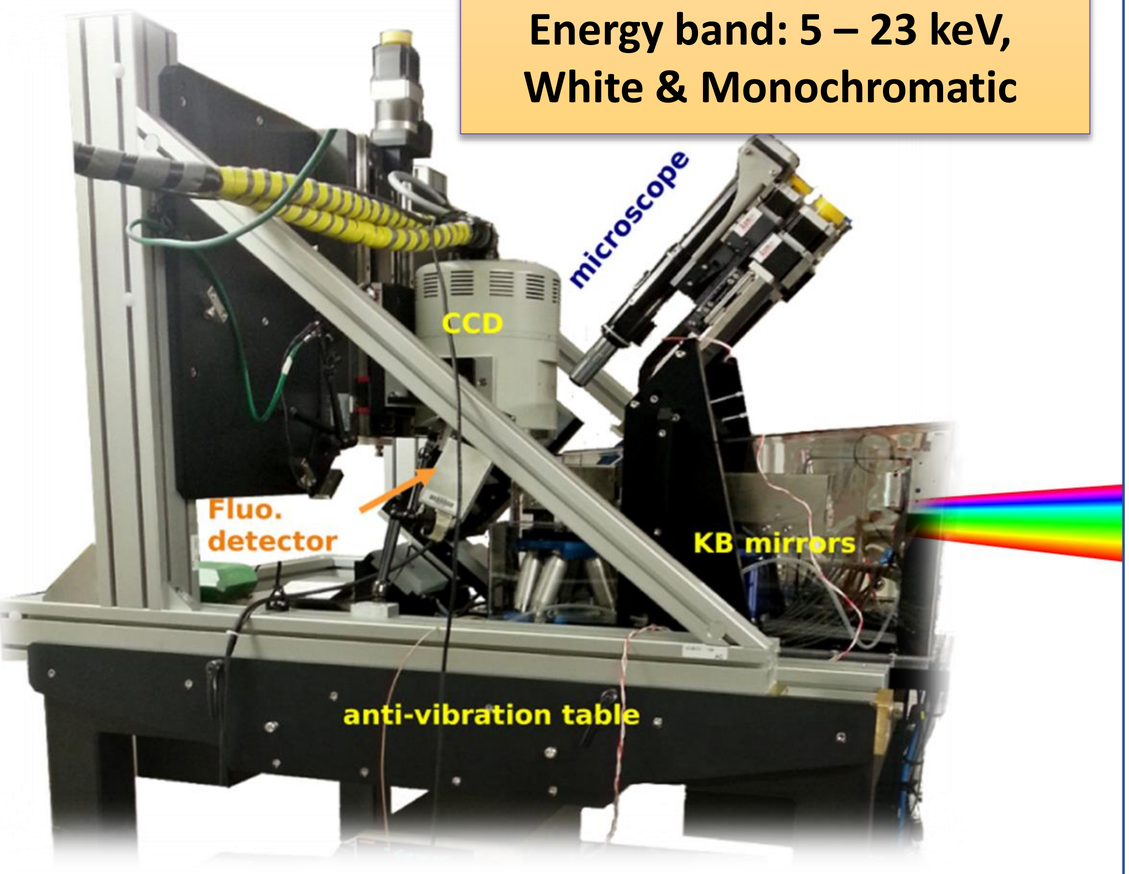
Courtesy J. Susini

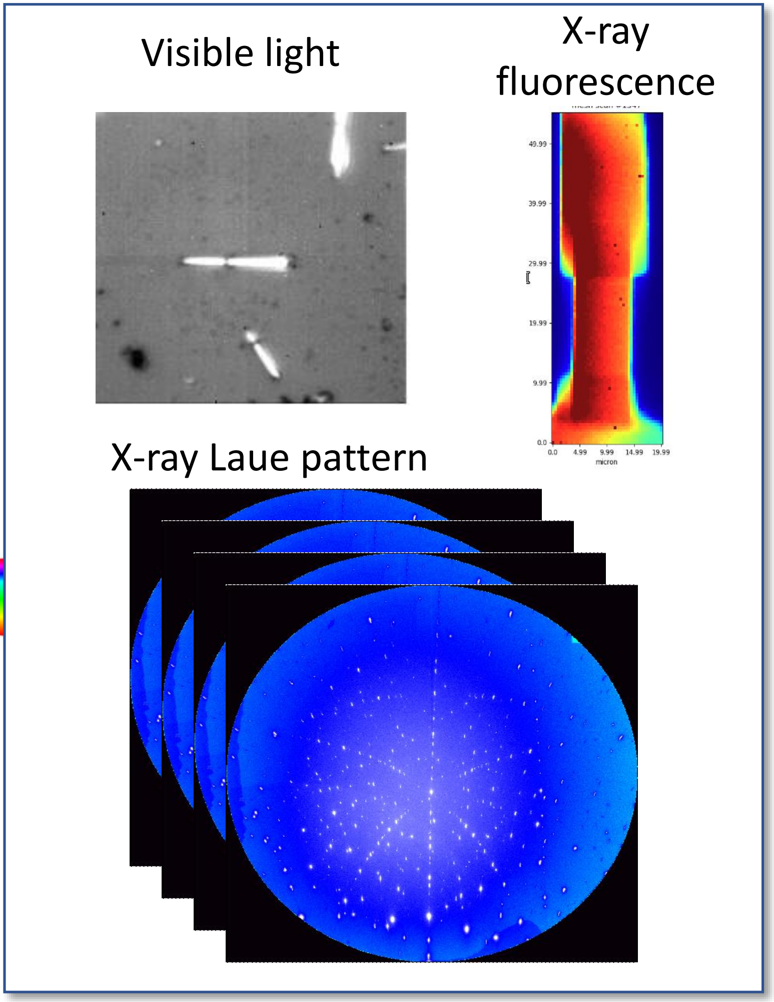
Microdiffraction Laue (µLaue)

Scattering @ InterFaces

Laue Diffraction Microscopy

Beamsize: 0.3 (H) x 0.3 (V) µm²
Energy band: 5 – 23 keV,
White & Monochromatic



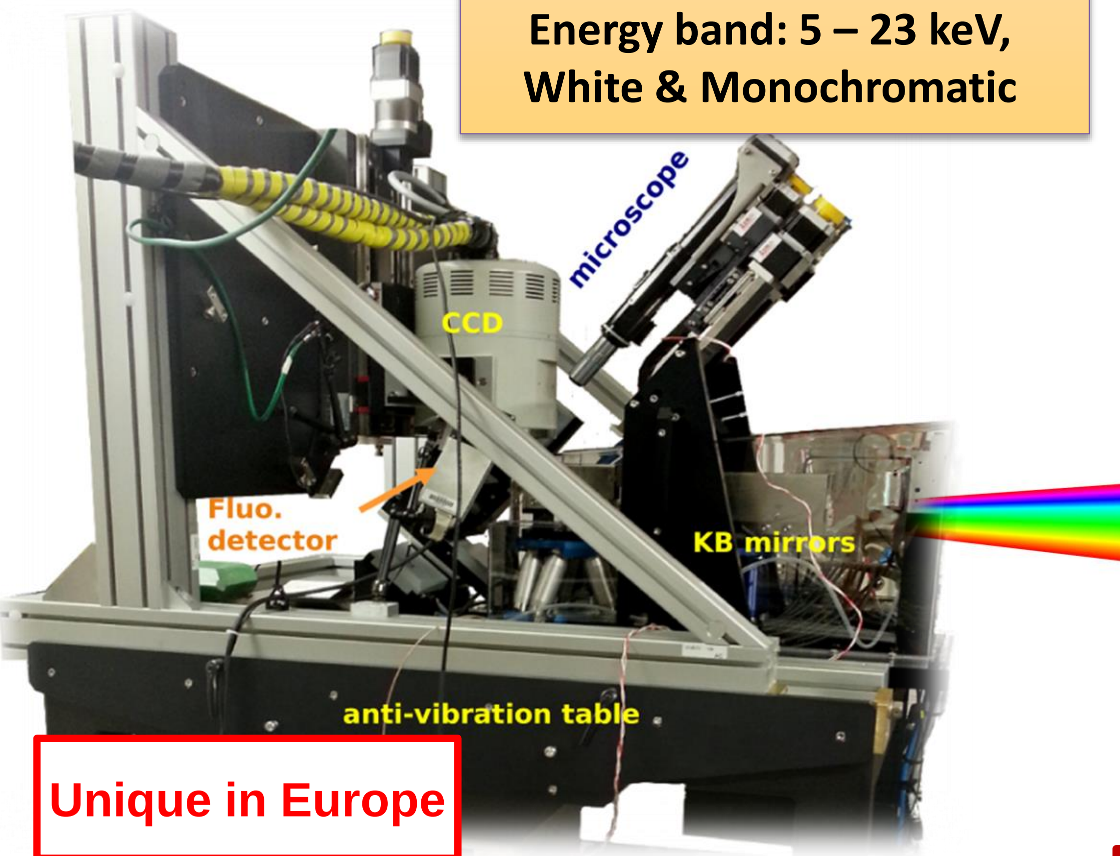


μLaue diffraction (μLaue)

Scattering @ InterFaces

Laue Diffraction Microscopy

Beamsize: 0.3 (H) x 0.3 (V) μm^2
Energy band: 5 – 23 keV,
White & Monochromatic



Unique in Europe

Routine

2D mapping
Orientation/Strain/phase
Crystalline defects

Mono- & poly-crystals
Single micro-object

In situ
furnace, stress, light …

Advanced
- Full stress
(energy resolution)
- 3D mapping
(depth resolution)

Structure: phase, orientation, strain
Morphology: Grain position & shape
Crystal defects: location & nature

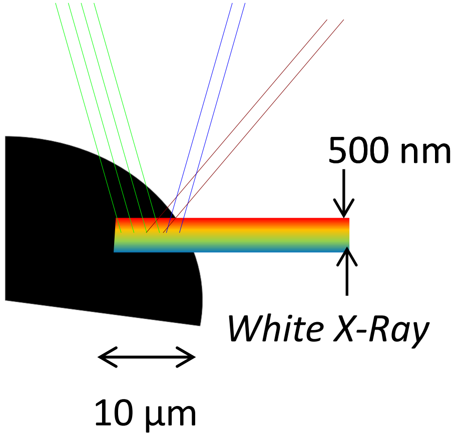
X-ray (scattering signal) microscopy for materials studies

•Local (residual) strains measurements

•Surface technique with useful probing depth : ~ several 10 µm

•Small gauge volume : < 500x500nm^2 x ~10 µm or grainsize limited

•X-ray equivalent to EBSD with:
- •Higher penetration, less sample preparation
- •Conducting or isolating materials
- •Finer resolution in orientation : x 1/100
- •High strain resolution: 10^{-4}



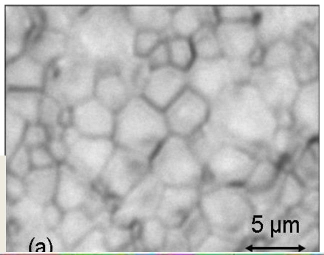
Still needs:
- •Accelerated acquisition - automated analysis
- •Synchrotron beamline

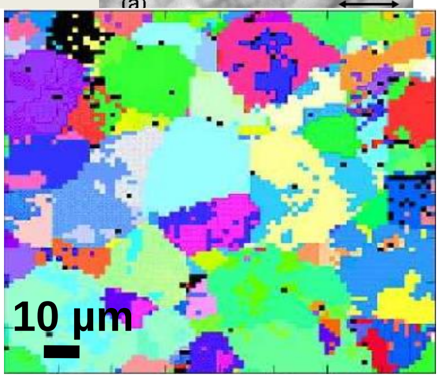
μLaue diffraction (μLaue)

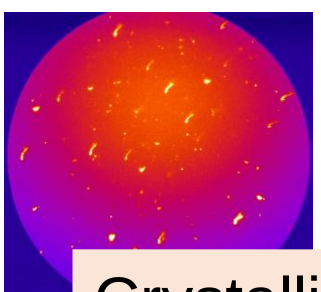
Scattering @ InterFaces

Strain/orientation resolution: 10^{-4}

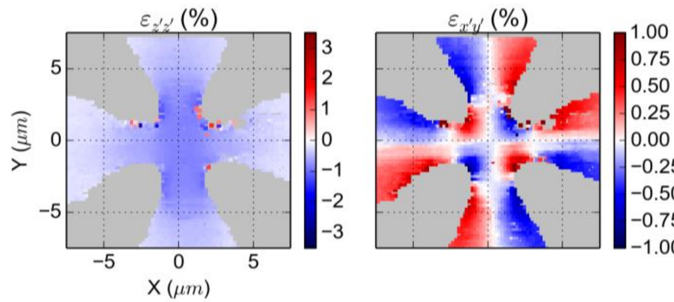
2D







Crystalline defects

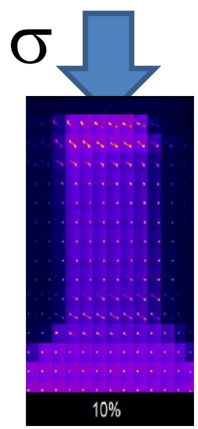


HR Strain map
In Ge strained membrane

stress

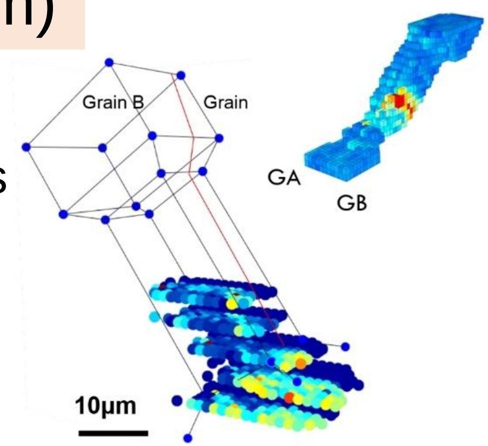
Microstructure & defects in Hard X-ray detector CdTe

High spatial resolution (< 1µm)



investigation of plasticity damages and stress level

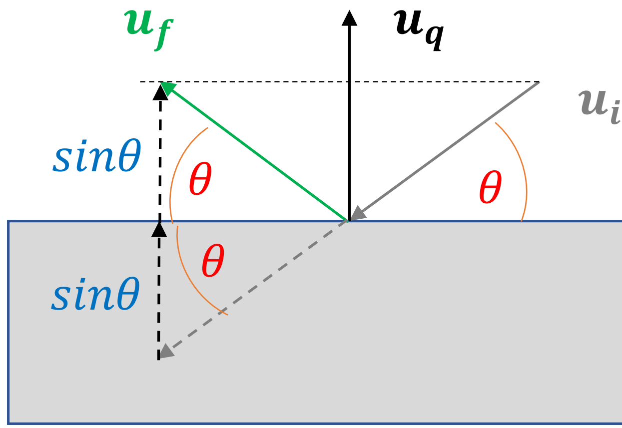
In situ



3D

Maths for Laue in a nutshell (1/2) : mirror reflection

Reflection of incident beam $\boldsymbol{u_i}$ by a mirror with normal vector $\boldsymbol{u_q}$ (elastic collision)

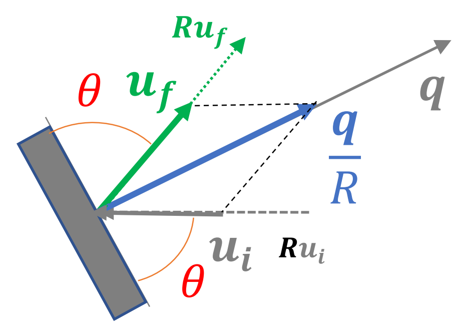


$$\boldsymbol{u_f} = u_i + 2sin\theta\boldsymbol{u_q} = u_i + \mathbf{2}\left(u_i.(-\boldsymbol{u_q})\right) = u_i - 2\frac{\boldsymbol{q}.u_i}{||\boldsymbol{q}||^2}\boldsymbol{q} = u_i + \frac{\boldsymbol{q}}{R}$$

$$||\boldsymbol{u_f}|| = ||u_i||$$

$$R\boldsymbol{u_f} = R\boldsymbol{u_i} + \boldsymbol{q}$$

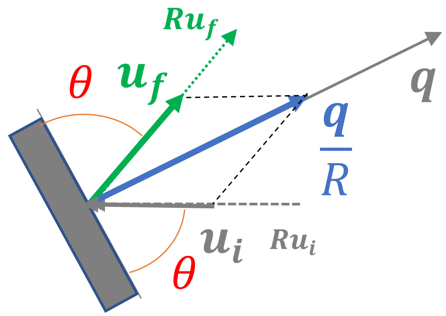
$$R(\boldsymbol{q}) = -\frac{||\boldsymbol{q}||^2}{2\boldsymbol{q}.\boldsymbol{u_i}}$$

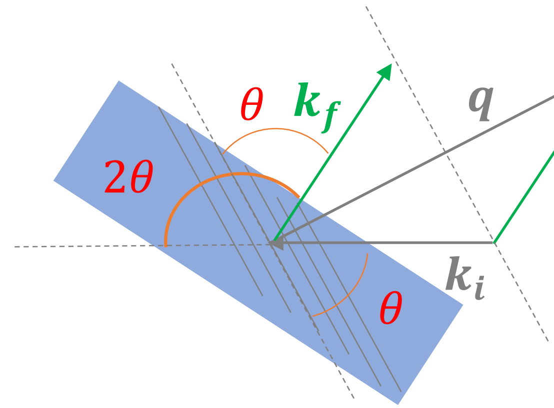


➢ Given $\boldsymbol{q}$ and u_i, it is easy to construct the reflected (or scattered) beam $\boldsymbol{u_f}$

Maths for Laue in a nutshell (2/2) : Bragg reflection

Atomic planes act as a mirror with normal $\boldsymbol{q}$ (elastic scattering).
Direction of the incoming wave (wave vector k_i) is changed into a new direction (wave vector $\boldsymbol{k_f}$) due to $\boldsymbol{q}$.





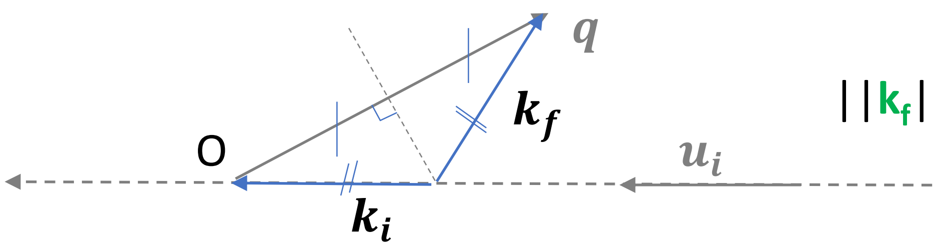
$$\frac{2\pi}{\lambda}\boldsymbol{u_f} = \frac{2\pi}{\lambda}u_i + \frac{4\pi}{\lambda}sin\theta\boldsymbol{u_q}$$

$$\boldsymbol{k_f} = k_i + ||\boldsymbol{q}||\boldsymbol{u_q} = k_i + \boldsymbol{q}$$

avec
$||\mathbf{k_f}||=||\mathbf{k_i}||=2\pi/\lambda$
$||\mathbf{q}|| = (2\pi/\lambda)\ \ 2\sin\theta$

$R(q)= 2\pi/\lambda =||q||/(2\sin\theta)$
$= E(keV)\ 2\pi/12{,}398\ = -\mathbf{q}^2/(2\mathbf{q}.\mathbf{u_i})$

Given $\boldsymbol{q}$ and incoming beam vectors u_i, the scattered vector $\boldsymbol{k_f}$ (direction and norm) is easily determined geometrically by means of the bisection of $\boldsymbol{q}$

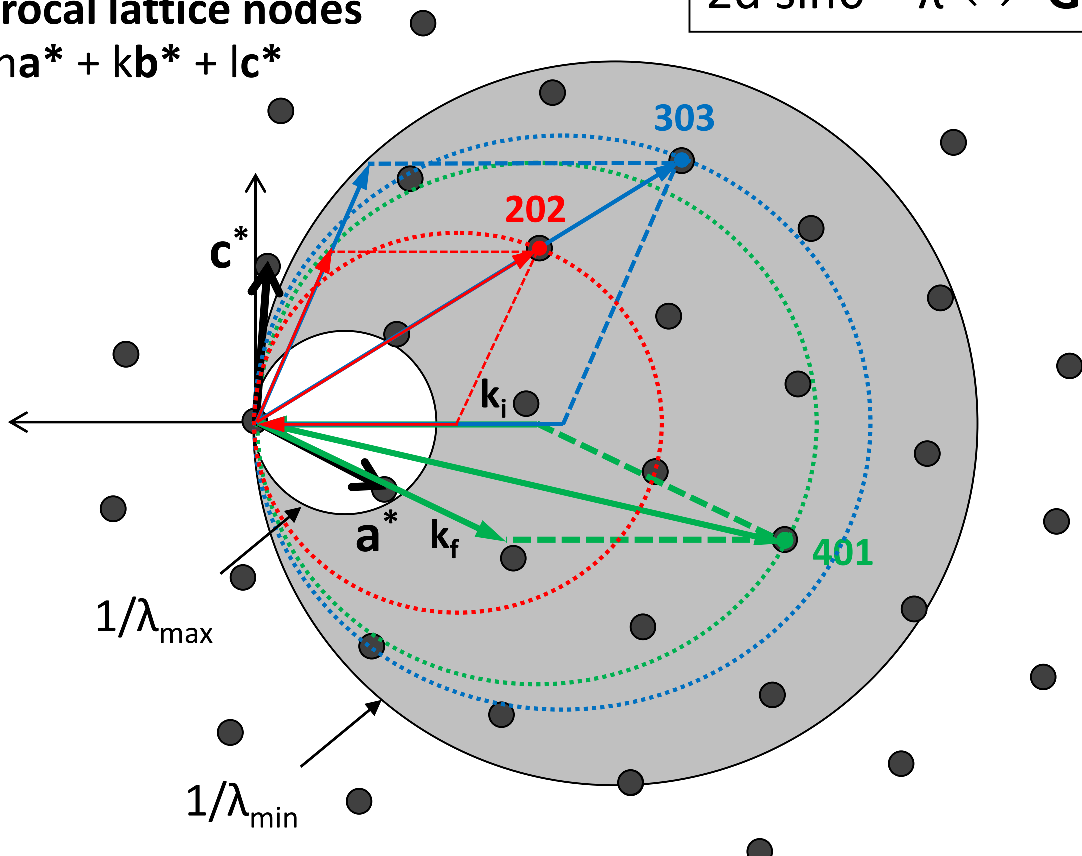


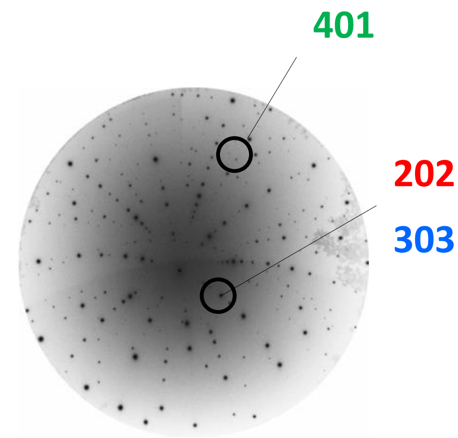
$||\mathbf{k_f}||=||\mathbf{k_i}||=2\pi/\lambda=-\mathbf{q}^2/(2\mathbf{q}.\mathbf{u_i})$

Laue Principles | measuring reciprocal directions & orientations

Reciprocal lattice nodes
$G^* = ha^* + kb^* + lc^*$

$2d \sin\theta = \lambda \Leftrightarrow G^* = k_f - k_i$



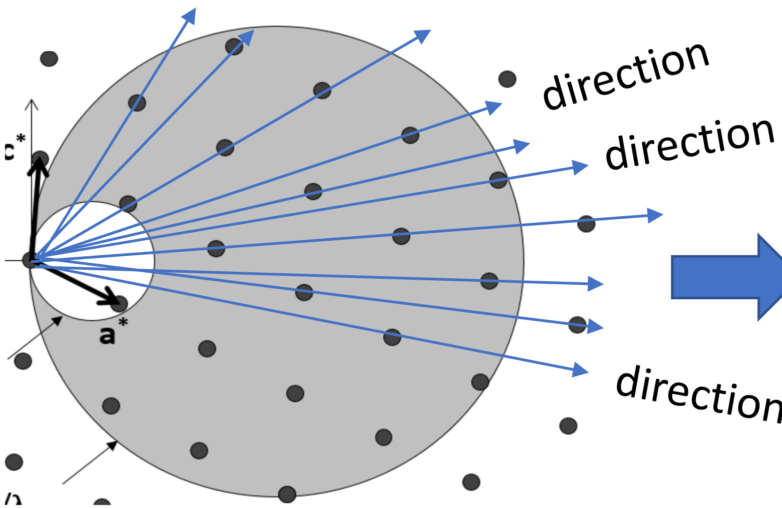


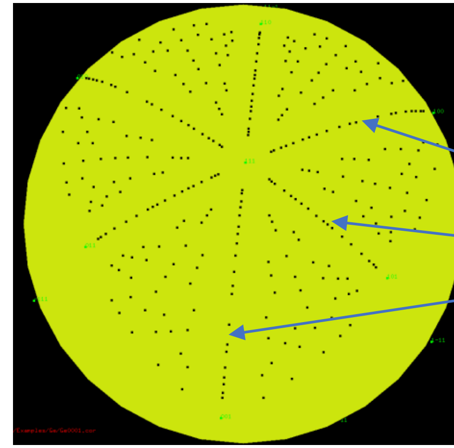
Mapping:
1 Reciprocal direction ⇔ 1 Laue spot
⇔ fund. and harmonics reciprocal nodes
$G^* / \|G^*\| \Leftrightarrow (X, Y)_{detector}$

Advantages

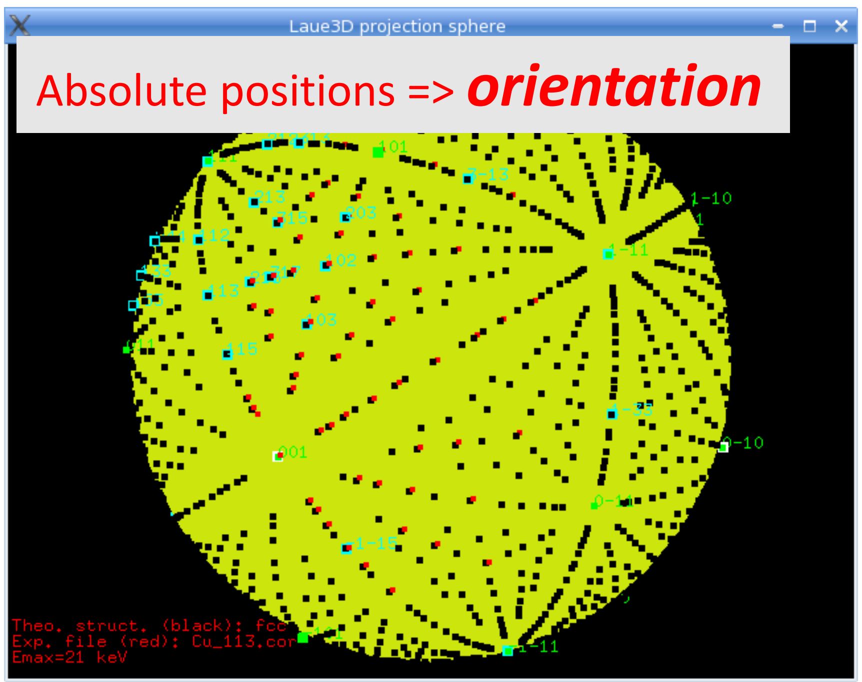
- Always diffraction signal for any crystal orientation
- Large number of Bragg reflections
- No sample rotation
- Determine easily orientation

Laue Principles | measuring reciprocal directions & orientations

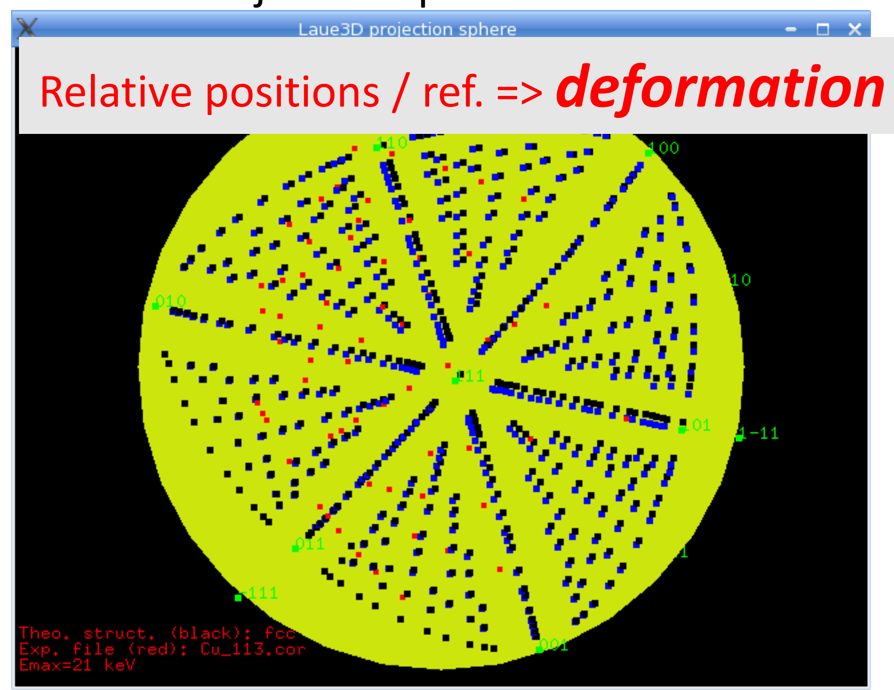




Projection sphere of directions

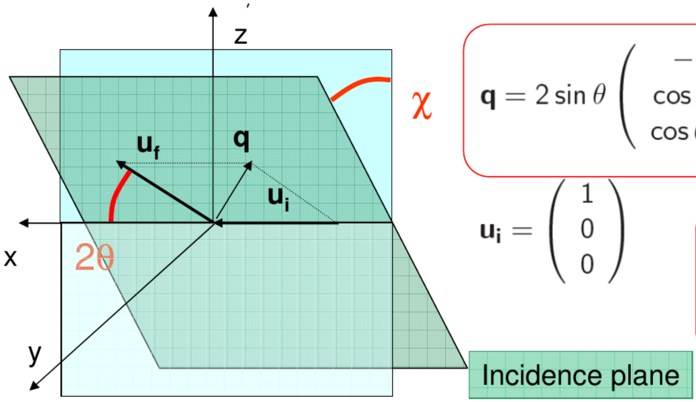


Matching exp. (red) Theo. (black) spots



Simulating strain (black->blue)

Laue Pattern visualisation. Frame



$$\mathbf{q} = 2\sin\theta \begin{pmatrix} -\sin\theta \\ \cos\theta\sin\chi \\ \cos\theta\cos\chi \end{pmatrix} = 2\sin\theta\tilde{\mathbf{q}}$$

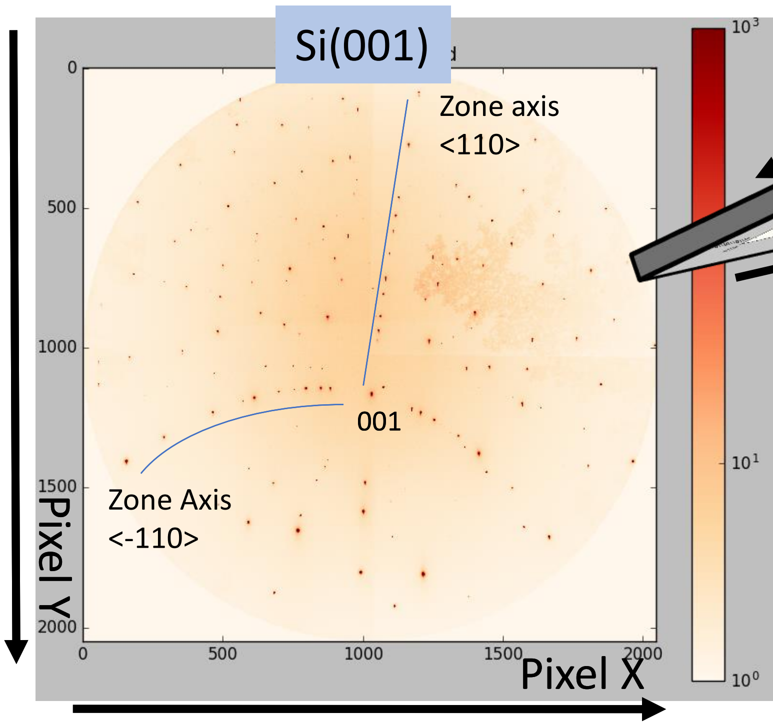
$$\mathbf{u_i} = \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}$$

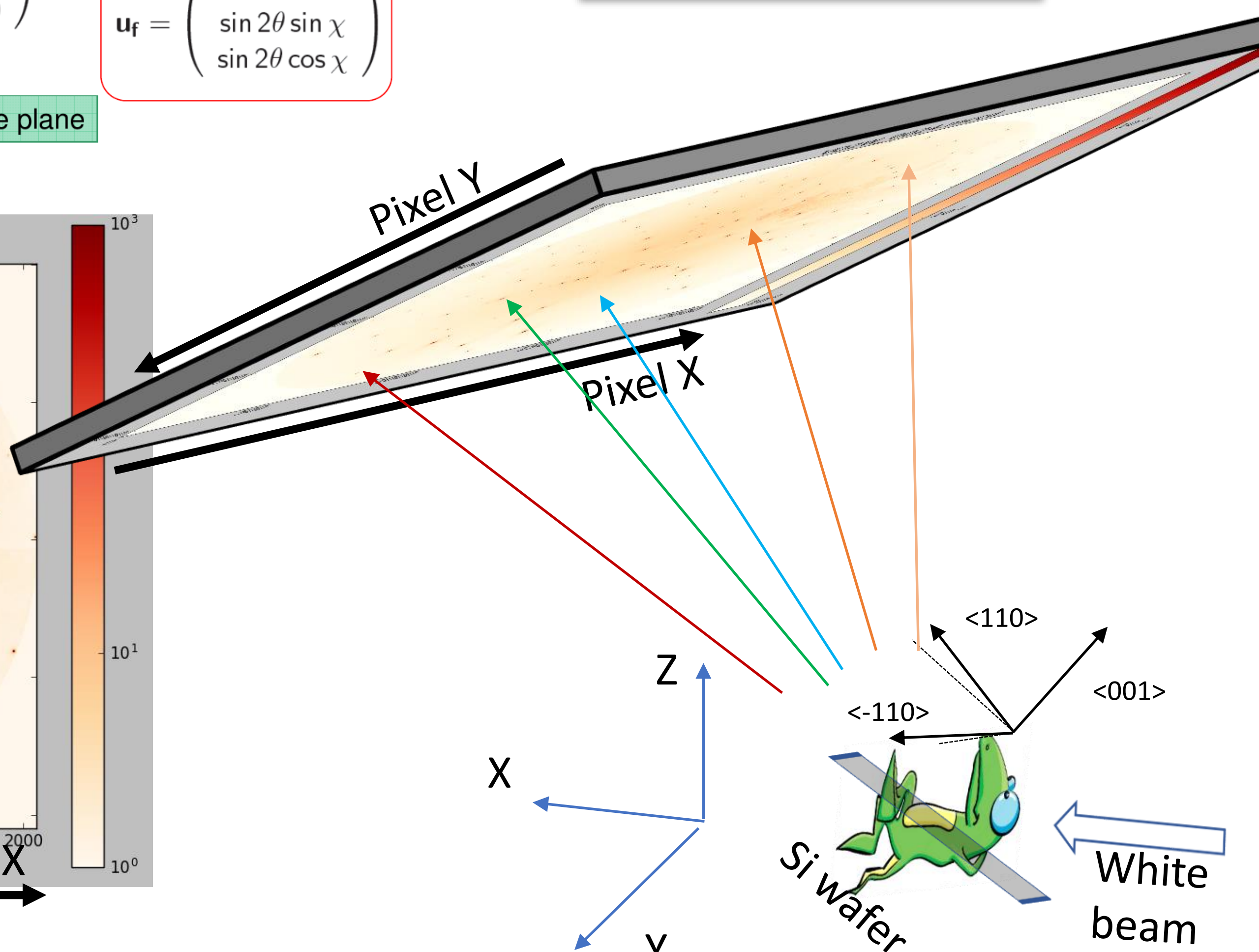
$$\mathbf{u_f} = \begin{pmatrix} \cos 2\theta \\ \sin 2\theta\sin\chi \\ \sin 2\theta\cos\chi \end{pmatrix}$$

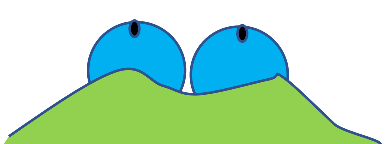
Scattering angles

$$(\boldsymbol{u_f}, \boldsymbol{u_i}) = 2\theta$$

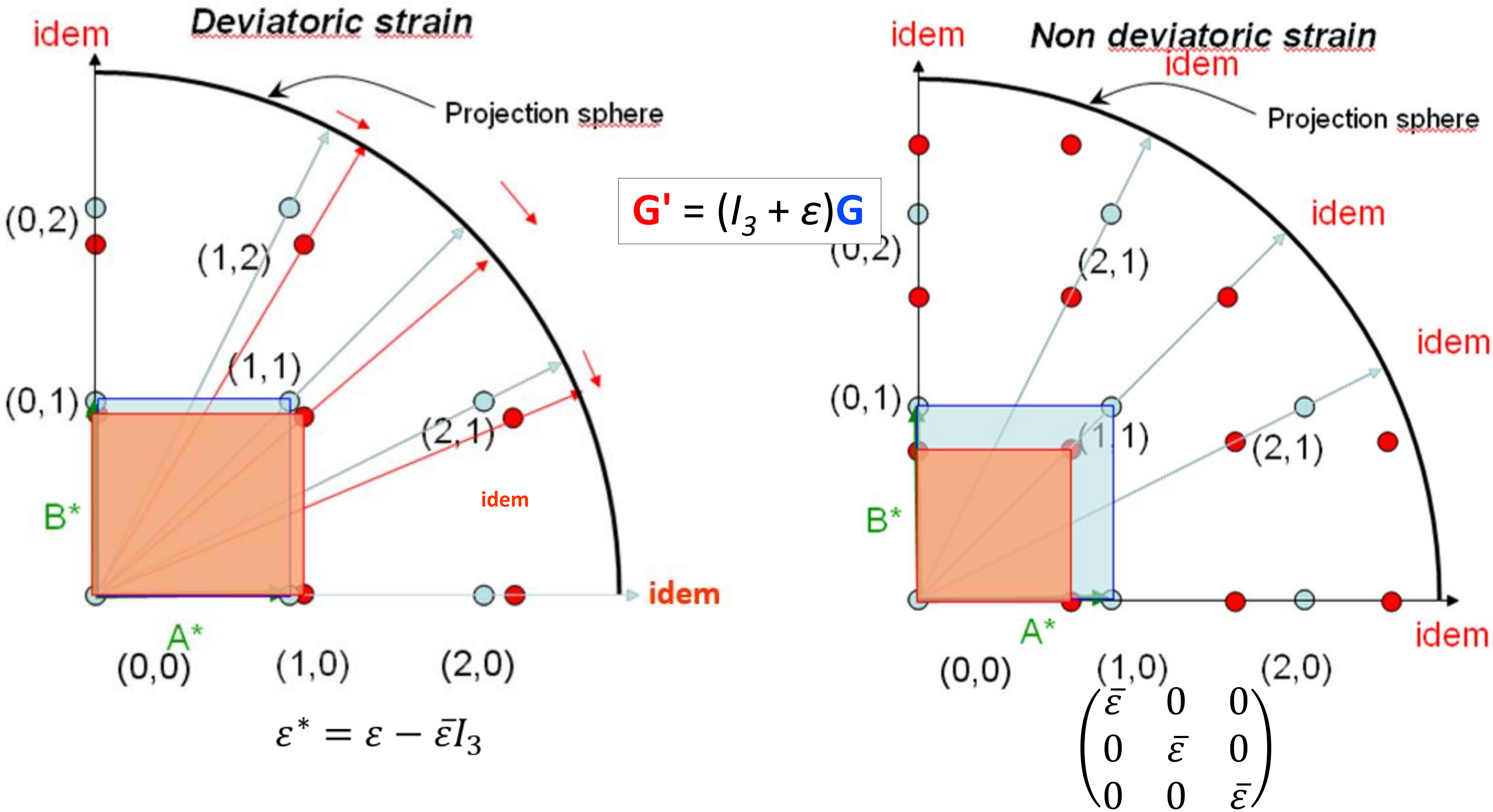
$$(\boldsymbol{u_f}, \boldsymbol{z}) = \chi$$







Laue Principles | Sensitivity to deviatoric strain

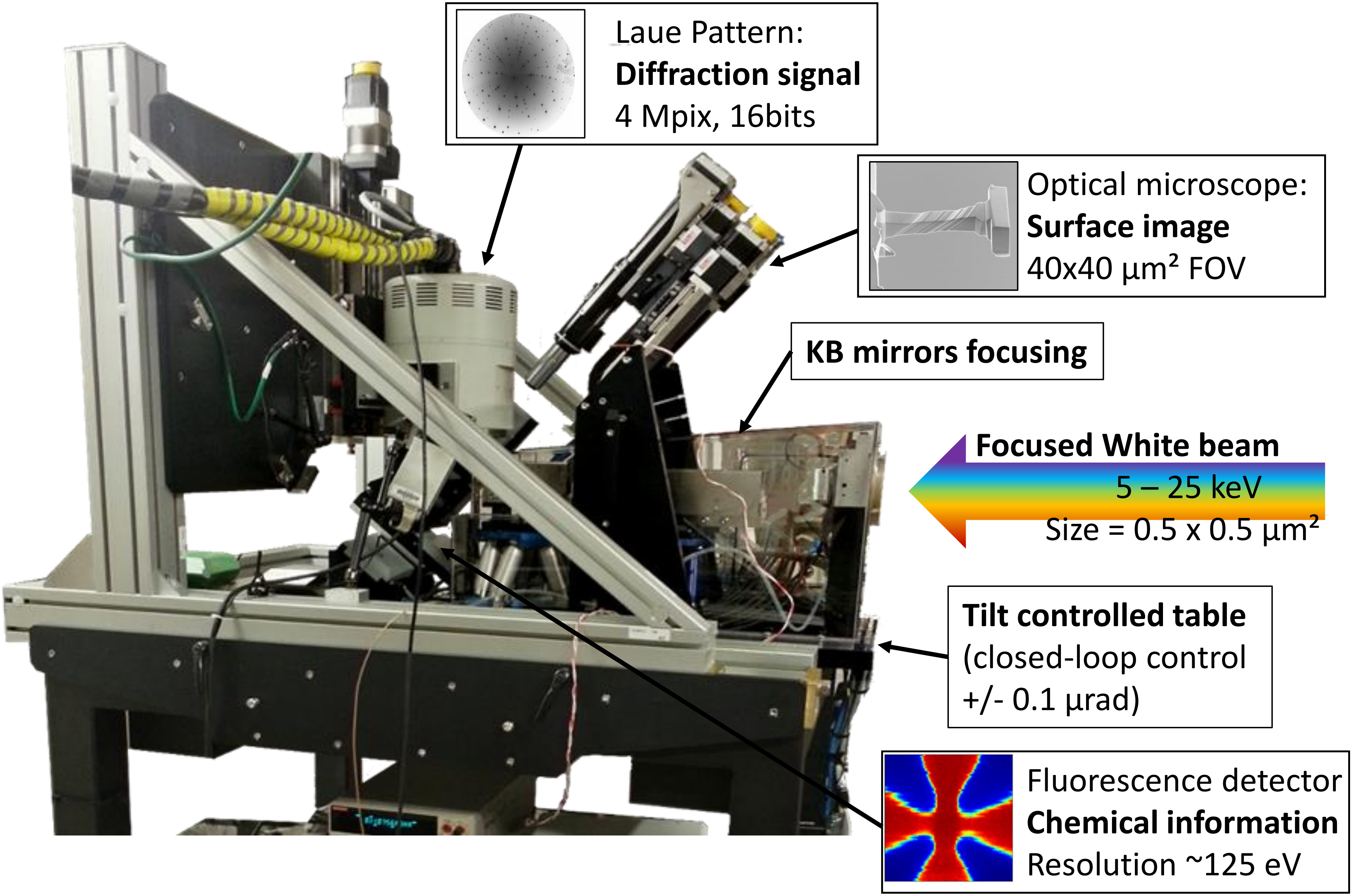


Laue => Deviatoric strain ε* : angular unit cell distortions i.e. b/a, c/a, α, β, γ

Laue + measuring 1 spot energy => 1 reciprocal vector length

=> **unit cell volume, a,b,c, α, β, γ (full strain)**

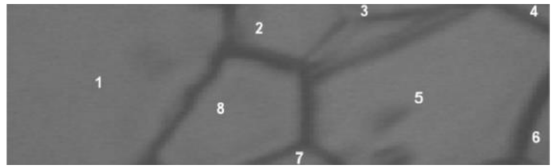
µLaue | Experimental setup



µLaue | 2D microscopy

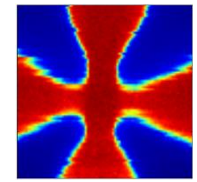
2D lateral resolution
< 0.5 x 0.5 µm²

surface optical microscope



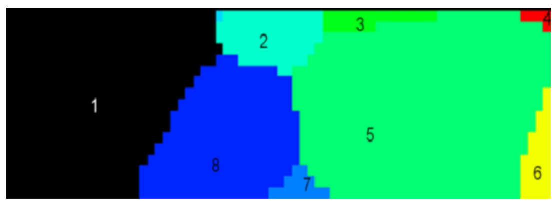
element/phase mapping

Fluo.
K_α Ge



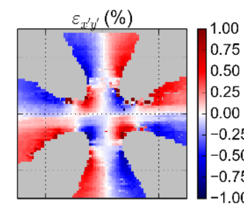
local crystal orientation

indexing

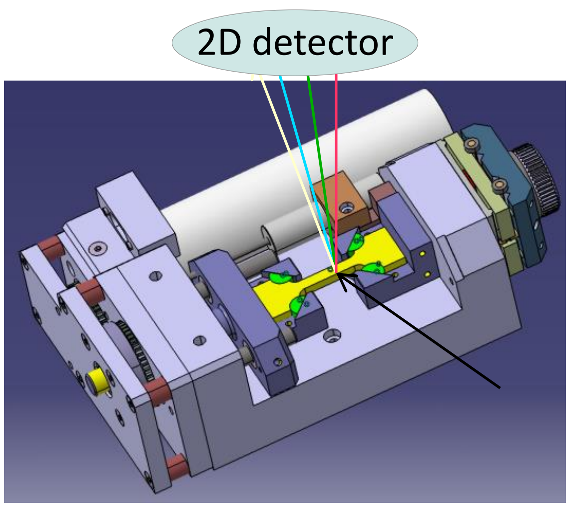


local unit cell strain

refinement



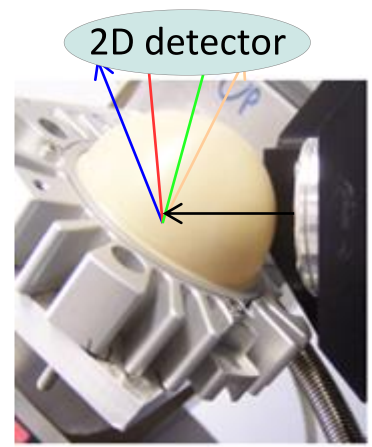
$\varepsilon_{xy,...}$



Tensile test

in situ & operando

σ, $k_B T$,
e-, light collection (XEOL),
…

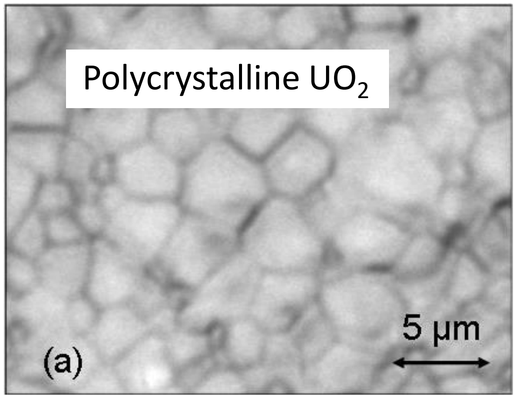


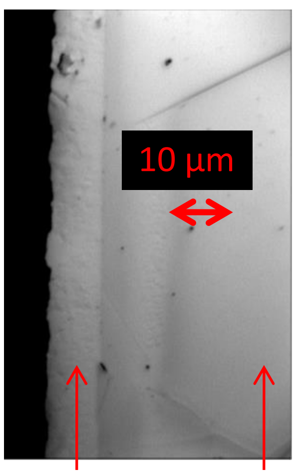
furnace

µLaue | First steps: (1) Locate & Measure

Visible microscope

Field of view: 200X200 to 40X40 μm^2

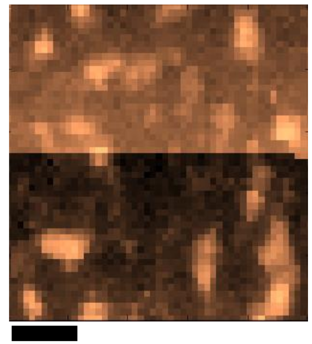




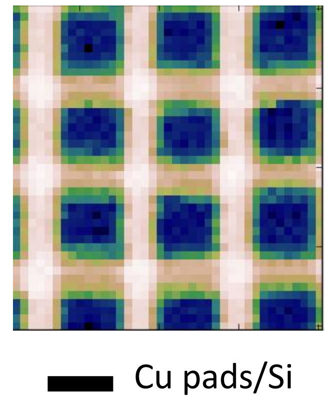
N-implanted Ni   Ni

Fluorescence map

Duration time: 2-10 min

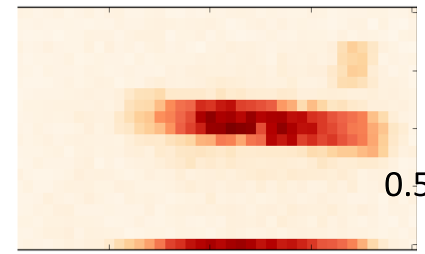


10µm   NdFeB



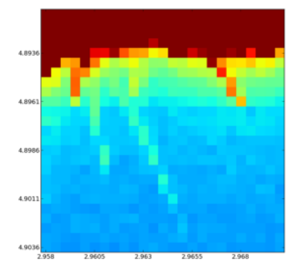
Cu pads/Si

2µm



0.5µm

Au nanowires



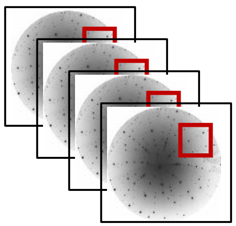
- Find micro region in macro sample (can be long!)
- Select single point/ROI for Laue diffraction

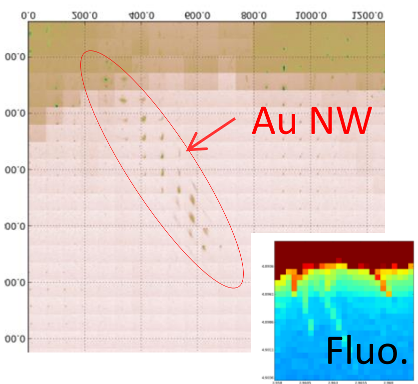
µLaue | First steps: (2) Imaging & Tracking

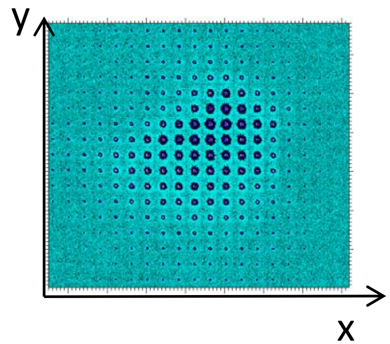
Single crystal & polycrystal mapping

from
Images
dataset

Images ROI mosaic





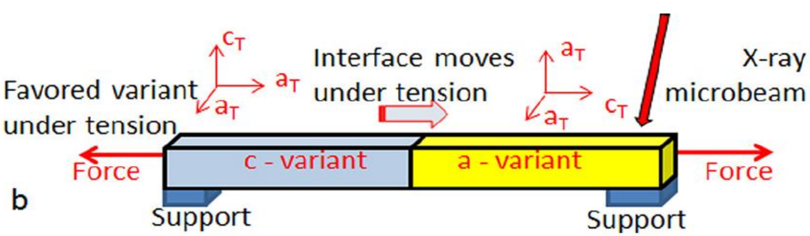


Single peak monitoring

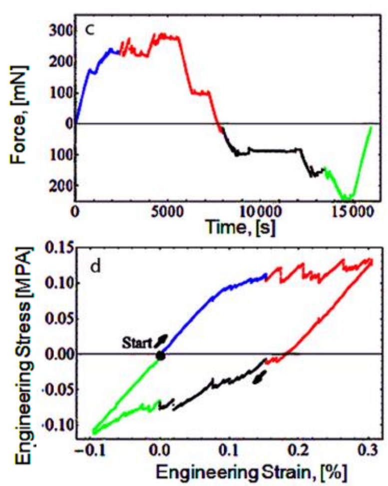
from
peaks lists
dataset

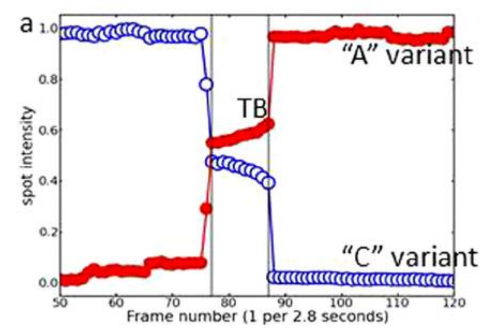
UO_2 polycrystals

GB & structural defects mapping



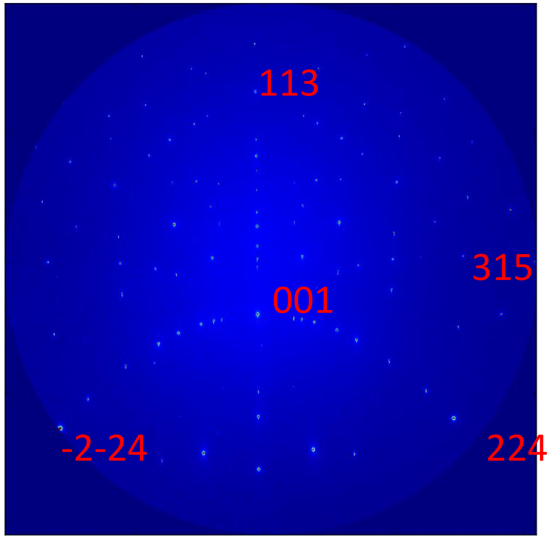
NiMnGa





R.I. Barabash *et al.*,
Appl. Phys. Lett. 103 (2), 021909 (2013)

µLaue | Second steps: (1) Indexation & (raw) Orientation



Indexing a ***single*** Laue pattern

$\mathbf{G^*} = h.\mathbf{a^*} + k.\mathbf{b^*} + l.\mathbf{c^*}$

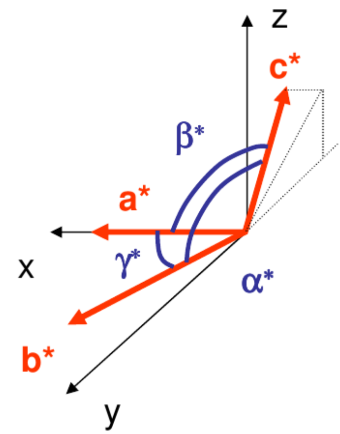
$\mathbf{q} = \mathbf{U_{raw}}.B_0.\mathbf{G^*} = \mathbf{k_f} - \mathbf{k_i}$

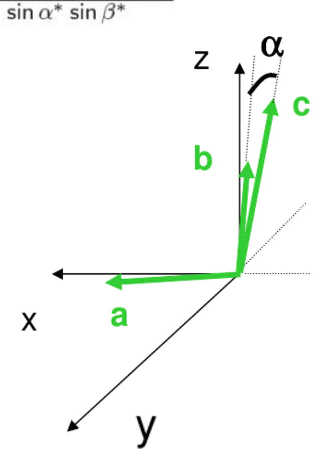
B_0 initial matrix a*,b*,c* in Lab. frame

$U_{raw} = U.(I_3 + \varepsilon)$ ε Strain

$$B0 = \begin{pmatrix} a^* & b^* \cos\gamma^* & c^* \cos\beta^* \\ 0 & b^* \sin\gamma^* & -c^* \sin\beta^* \cos\alpha \\ 0 & 0 & c^* \sin\beta^* \sin\alpha \end{pmatrix}$$

$$\cos\alpha = \frac{\cos\beta^* \cos\gamma^* - \cos\alpha^*}{\sin\alpha^* \sin\beta^*}$$





Indexing results

- Miller indices (hkl)
- crystal orientation ($\mathbf{U_{raw}}$)

Indexing Basic method principles:
Recognition of angle between 2 atomic planes normals

Reference Table (*AnglesLUT*) des angles α_{ij} entre plans $(hkl)_i$ et $(hkl)_j$

Example: Cubic structure with highest Miller indices (*nLUT*) =3

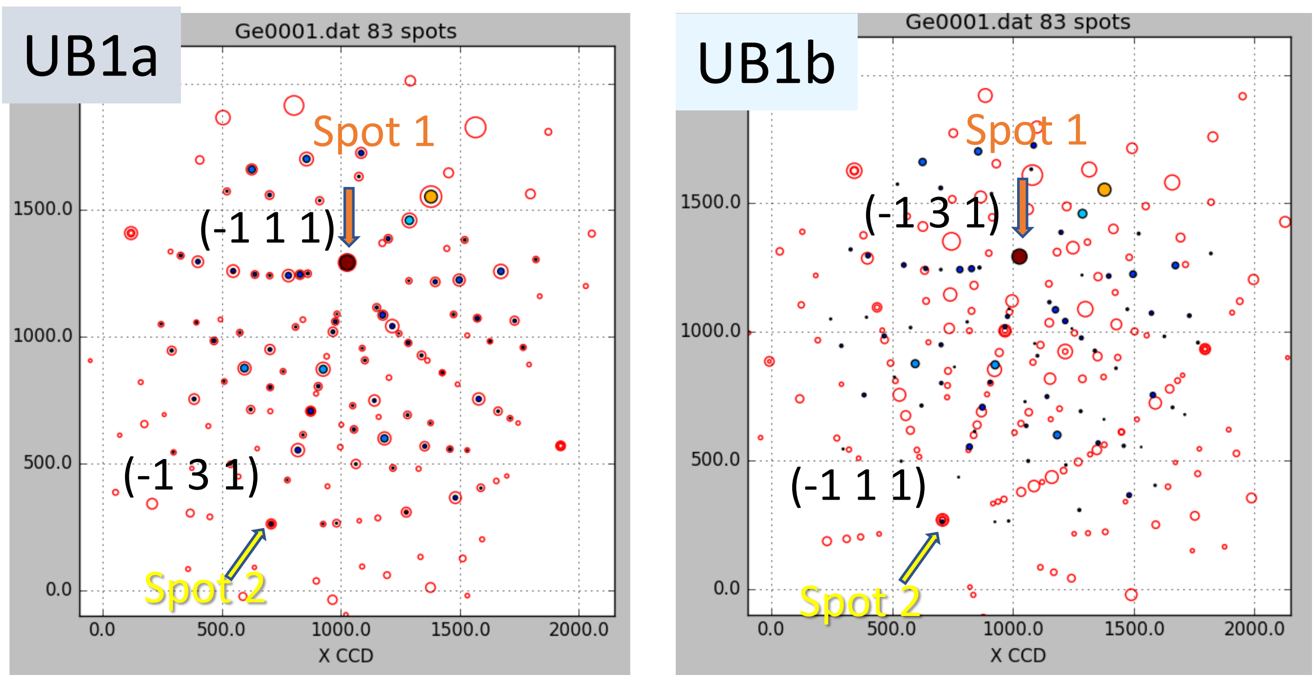
$\{h_2k_2l_2\}$	$\{h_1k_1l_1\}$						
	100	110	111	210	211	221	310
100	0 90						
110	45 90	0 60 90					
111	54.7	35.3 90	0 70.5 109.5				
210	26.6 63.4 90	18.4 50.8 71.6	39.2 75.0	0 36.9 53.1			
211	35.3 65.9	30 54.7 73.2 90	19.5 61.9 90	24.1 43.1 56.8	0 33.6 48.2		
221	48.2 70.5	19.5 45 76.4 90	15.8 54.7 78.9	26.6 41.8 53.4	17.7 35.3 47.1	0 27.3 39.0	
310	18.4 71.6 90	26.6 47.9 63.4 77.1	43.1 68.6	8.1 58.1 45	25.4 49.8 58.9	32.5 42.5 58.2	0 25.9 36.9
311	25.2 72.5	31.5 64.8 90	29.5 58.5 80.0	19.3 47.6 66.1	10.0 42.4 60.5	25.2 45.3 59.8	17.6 40.3 55.1
320	33.7 56.3 90	11.3 54.0 66.9	61.3 71.3	7.1 29.8 41.9	25.2 37.6 55.6	22.4 42.3 49.7	15.3 37.9 52.1
321	36.7 57.7 74.5	19.1 40.9 55.5	22.2 51.9 72.0 90	17.0 33.2 53.3	10.9 29.2 40.2	11.5 27.0 36.7	21.6 32.3 40.5
331	46.5	13.1	22.0				

Indexing: recognition of angle between 2 atomic planes normals

if: $|\alpha_{k,l} - \alpha_{i,j\ ref}| < \alpha_{max}$, angle $\alpha_{i,j}$ in LUT is *recognised* and corresponds to a pair of planes {$h_1k_1l_1$, $h_2k_2l_2$} and *2 potential* solutions:

	Spot 1	Spot 2	Orientation Matrix
Solution 1a	$h_1k_1l_1$	$h_2k_2l_2$	UB1a
Solution 1b	$h_2k_2l_2$	$h_1k_1l_1$	UB1b

Then, 2 Laue patterns are simulated:



experimental

simulated

$\alpha_{1,2}$= 29,501°

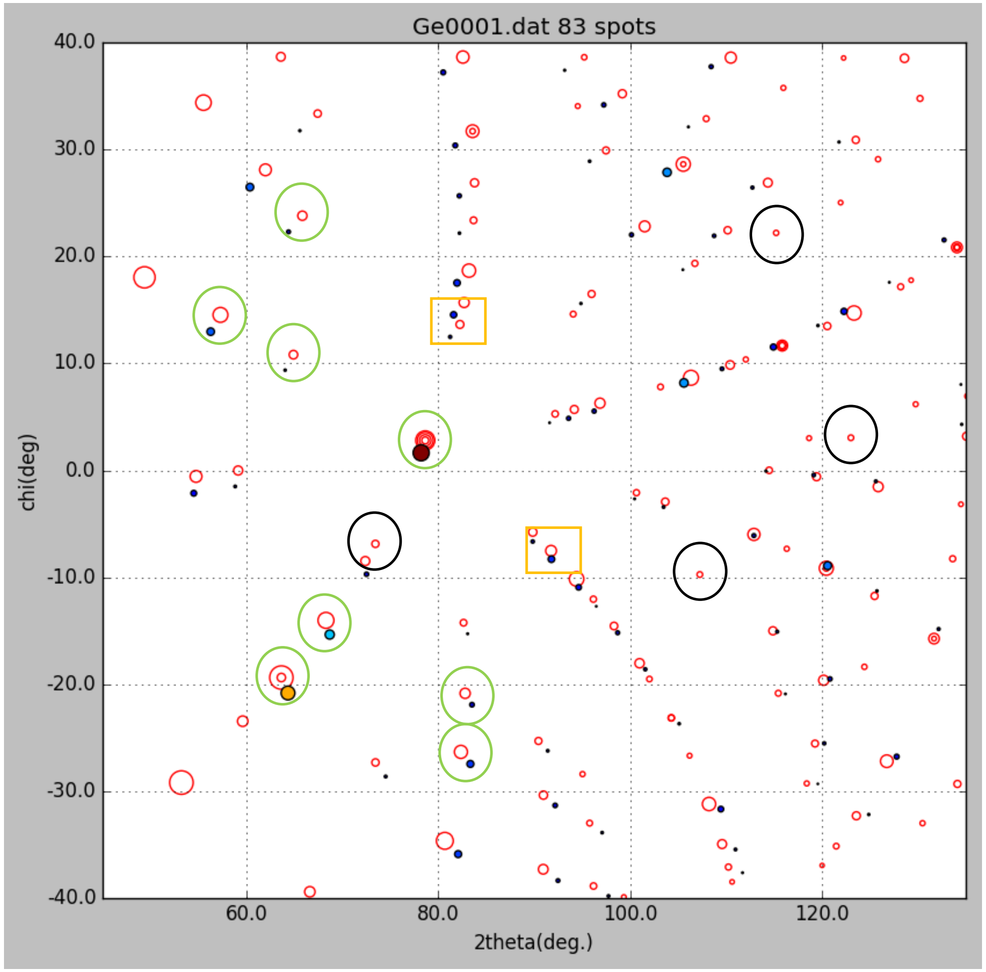
$h_1k_1l_1$ =(-1 1 1)

$h_2k_2l_2$ =(-1 3 1)

Evaluate matching level between two Laue Patterns

Automatic pairing between neighbouring theoretical (hollow red circles) and experimental (filled circles) is performed by (*Auto Links)* to:

- ① assess the Laue patterns 'matching rate'
- ② identify unambiguous pairs of (exp., theo.) for structure refinement



① Matching Rate

$(N_b(\text{green}+)+N_b(\text{orange}))\ /Nspots_{theo}$

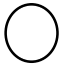
② Nb of unambiguous links

$N_{pairs}(\text{green})$

Within Matching tolerance angle

1 theo. spot linked close to 1 exp. spot

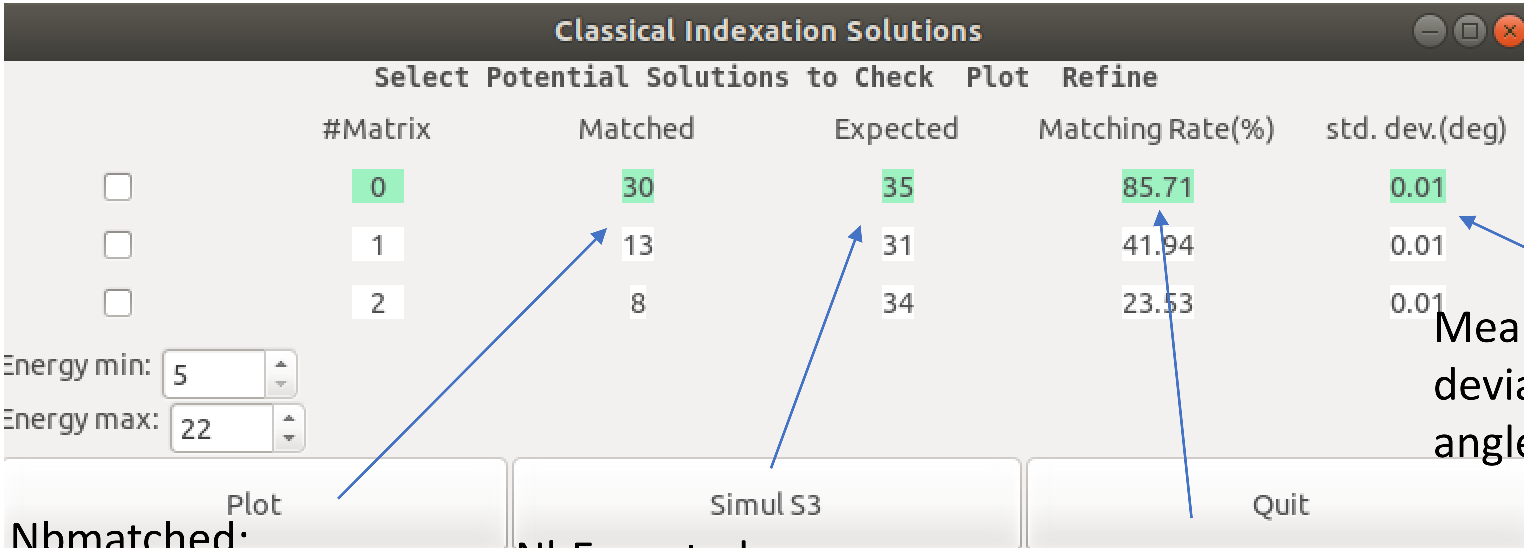
1 theo. spot linked close to several exp. spots



1 theo. Spot without neighb. Exp. Spots (*Missing Reflection* in exp. Spots list)

Matching rates report of potential indexing results

Given two spots angle recognition, several potential indexing solutions (i.e. UB orientation matrix) are listed:



Mean Standard deviation of pairs angles

Nbmatched:
Nb of exp. Spots (including harmonics) linked to a theo. spot

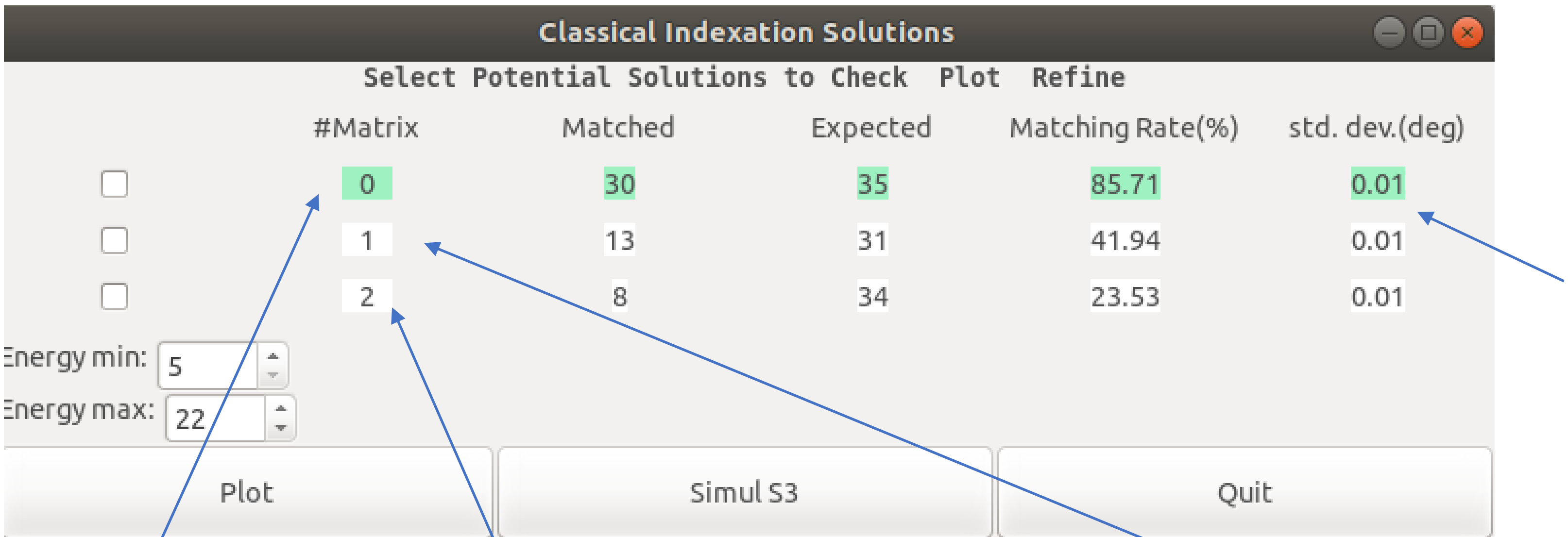
NbExpected:
Number of theo. Spots (including harmonics)

Matching Rate:
Nbmatched / NbExpected

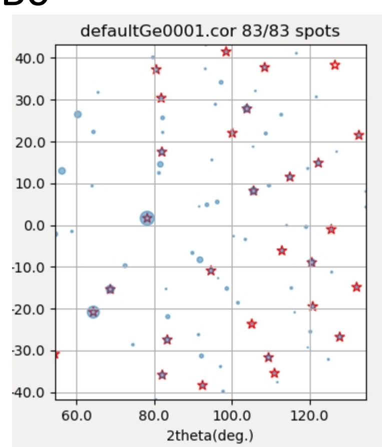
A poor de Matching Rate can be due to:
- A small number of exp. Spots (see Peak Search procedure)
- The reference structure for indexing is not well chosen (strong lattice deformation ?)
- Matching tolerance angle is too small

Matching rates report of potential indexing results

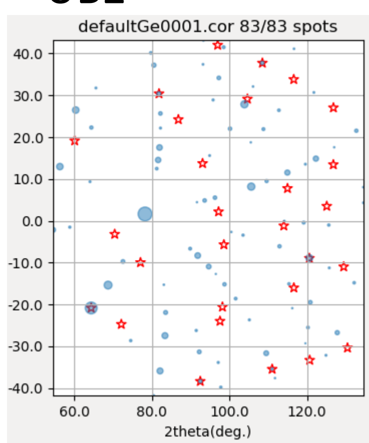
Given two spots angle recognition, several potential indexing solutions (i.e. UB orientation matrix) are listed:



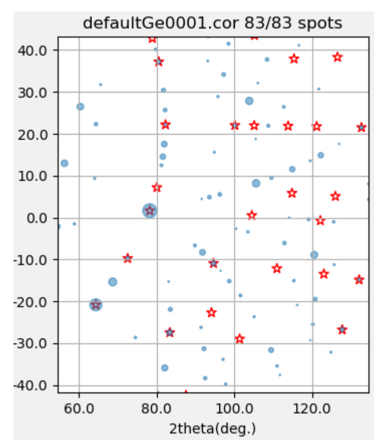
UB0



UB2

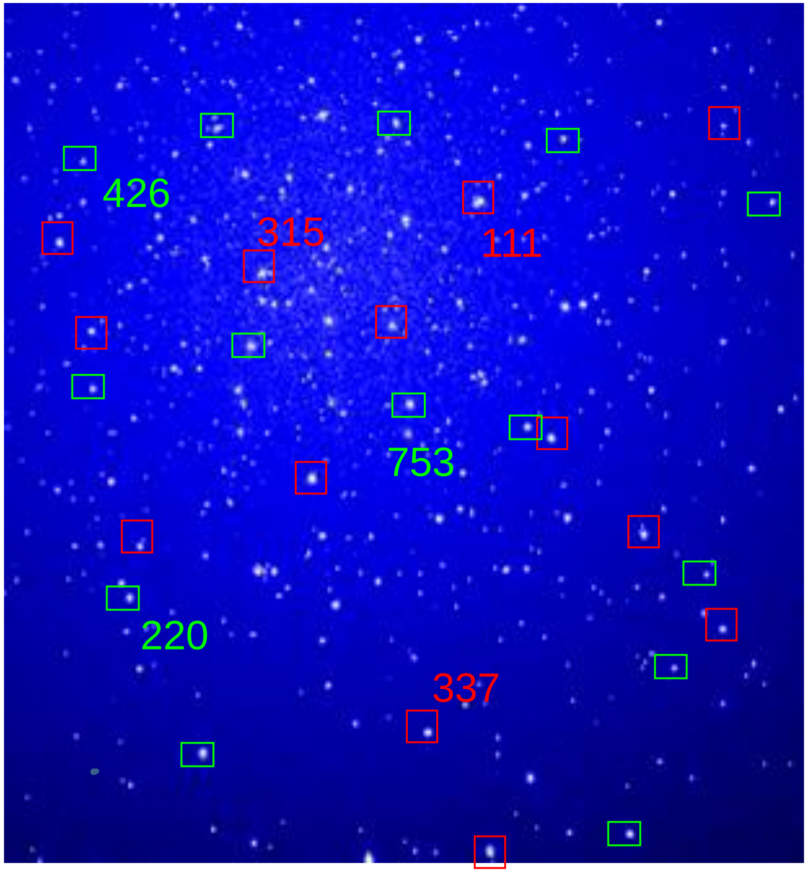


UB1



µLaue | Second steps: (1) Indexation & (raw) Orientation

Multiple indexation
of superimposed Laue patterns



Grain 1

Grain 2

...

Grain n

Indexation

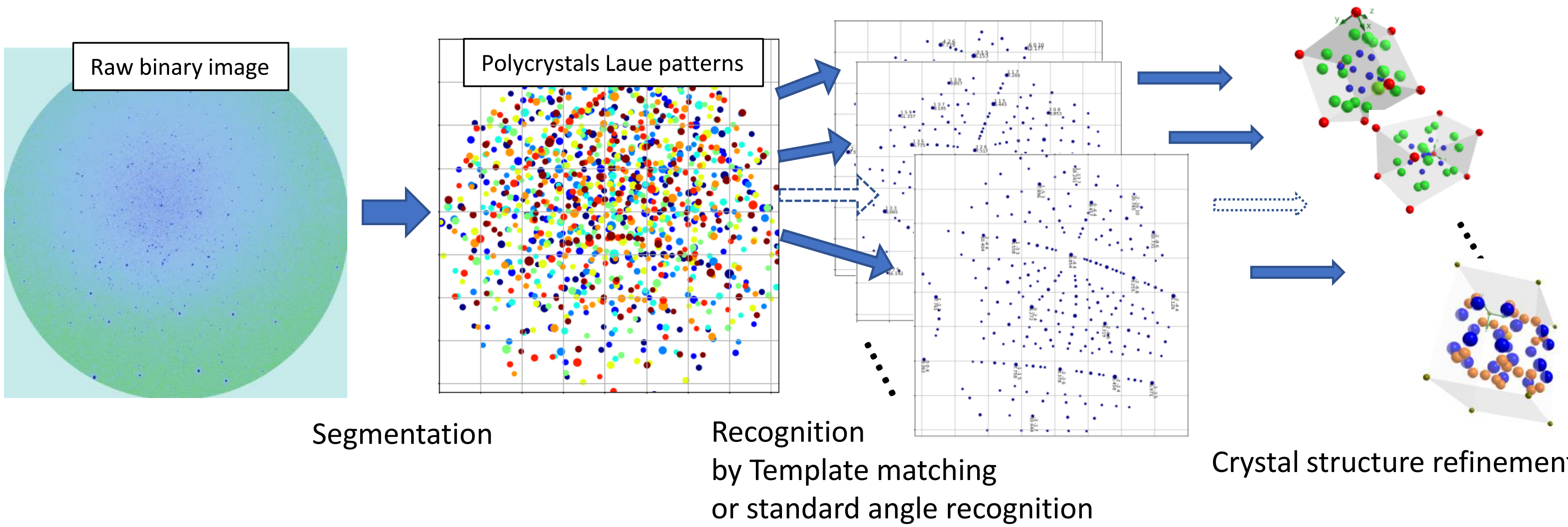
- Miller indices (hkl)
- crystal orientation (U_{raw})
- **Separate grains contributions**

✓ **Sequential indexation**

in case of no orientation relationships between grains

µLaue microscopy challenges

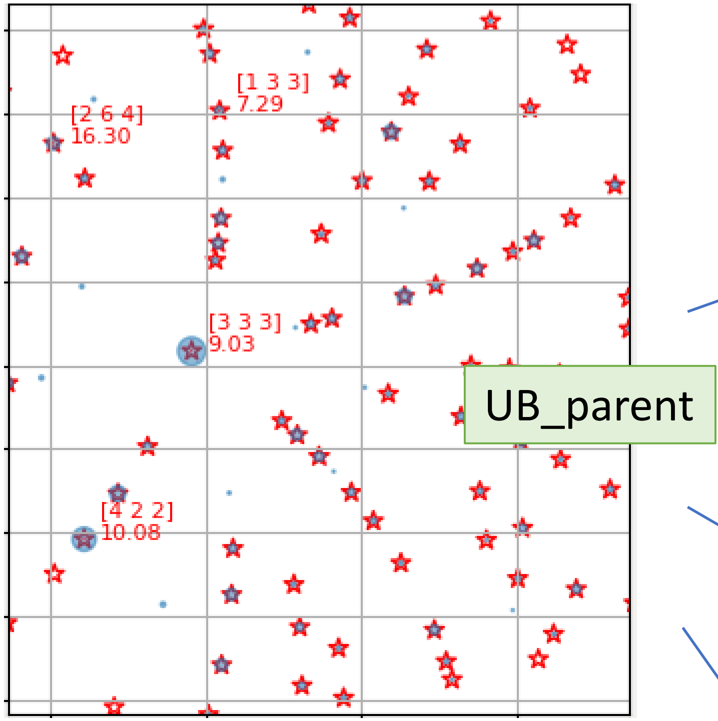
Single image analysis of complex Laue patterns



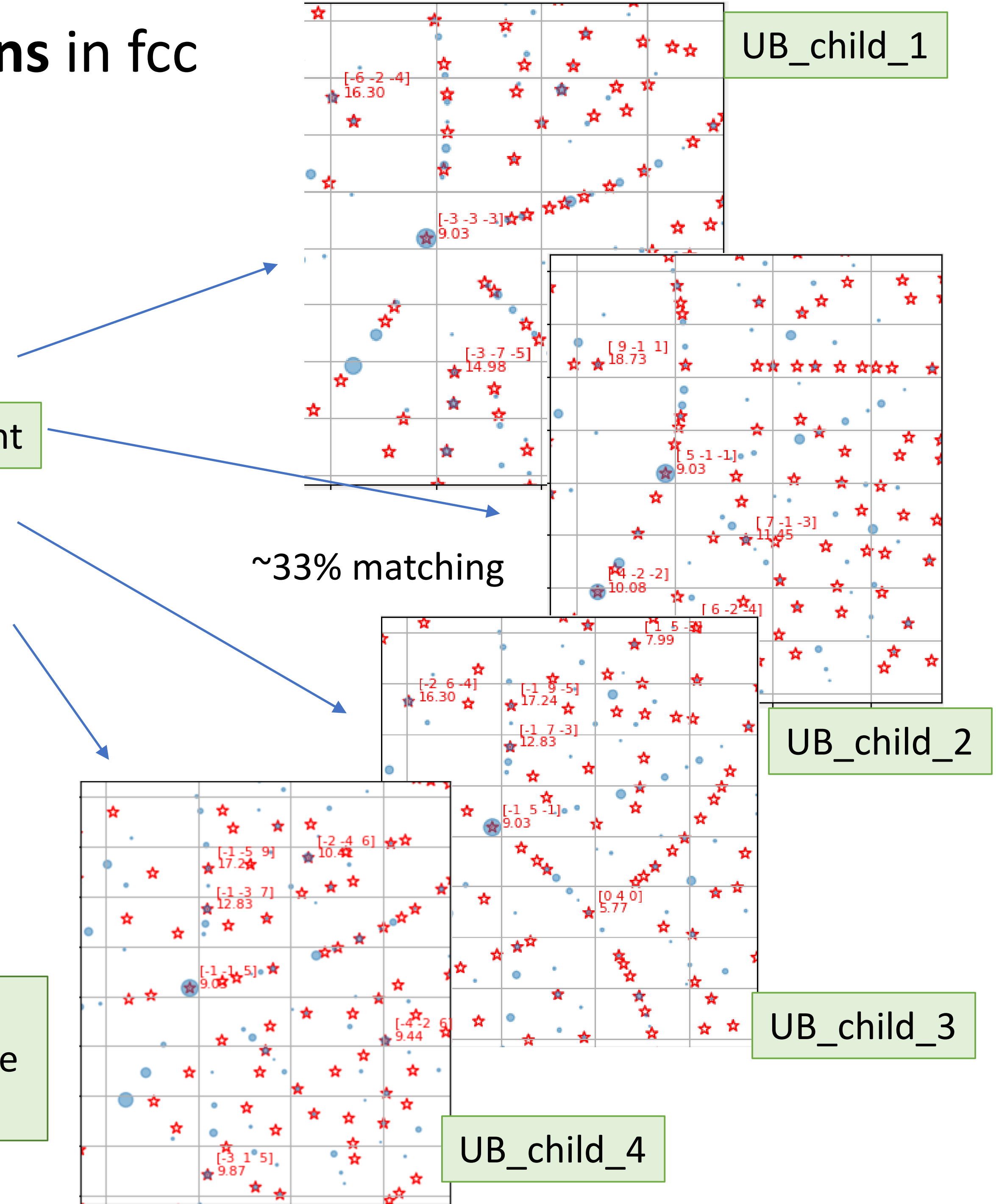
✓ **parallel indexation**

to handle orientation relationships between grains

Dealing with Σ3 **twins** in fcc



100% matching

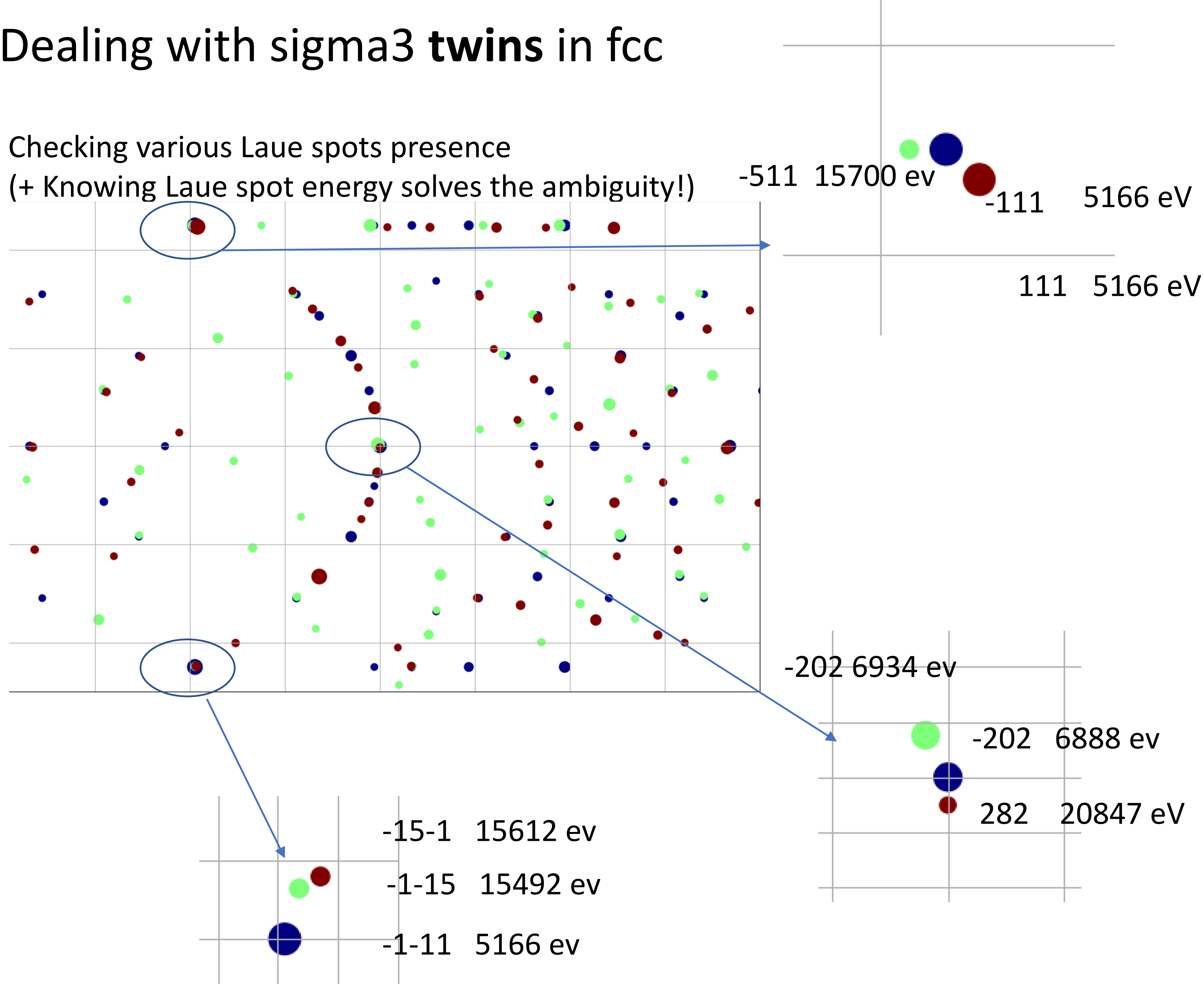


~33% matching

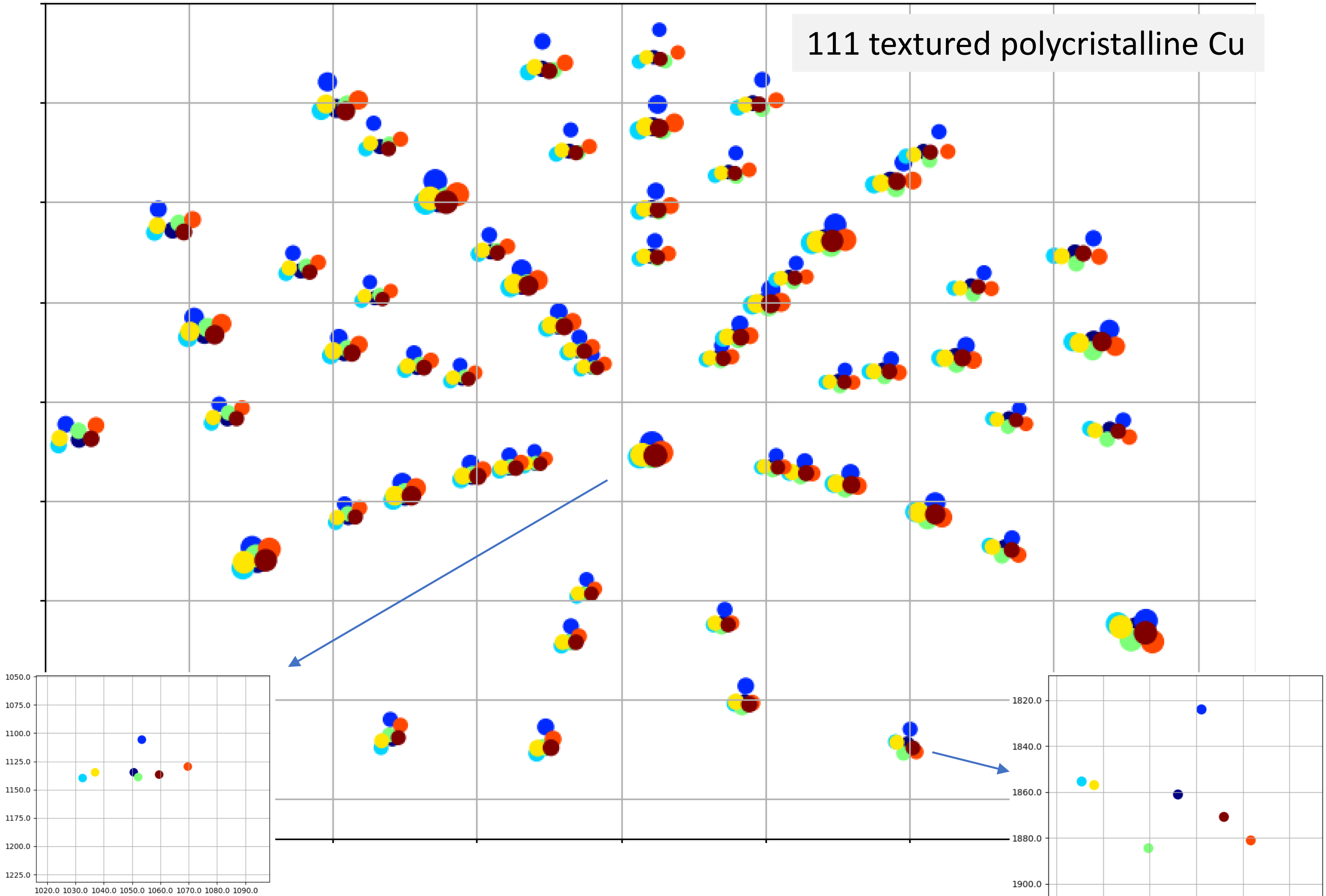
Twins
Ambiguous superimposition of Laue spots (of different grains and hkl)

Dealing with sigma3 **twins** in fcc

Checking various Laue spots presence
(+ Knowing Laue spot energy solves the ambiguity!)

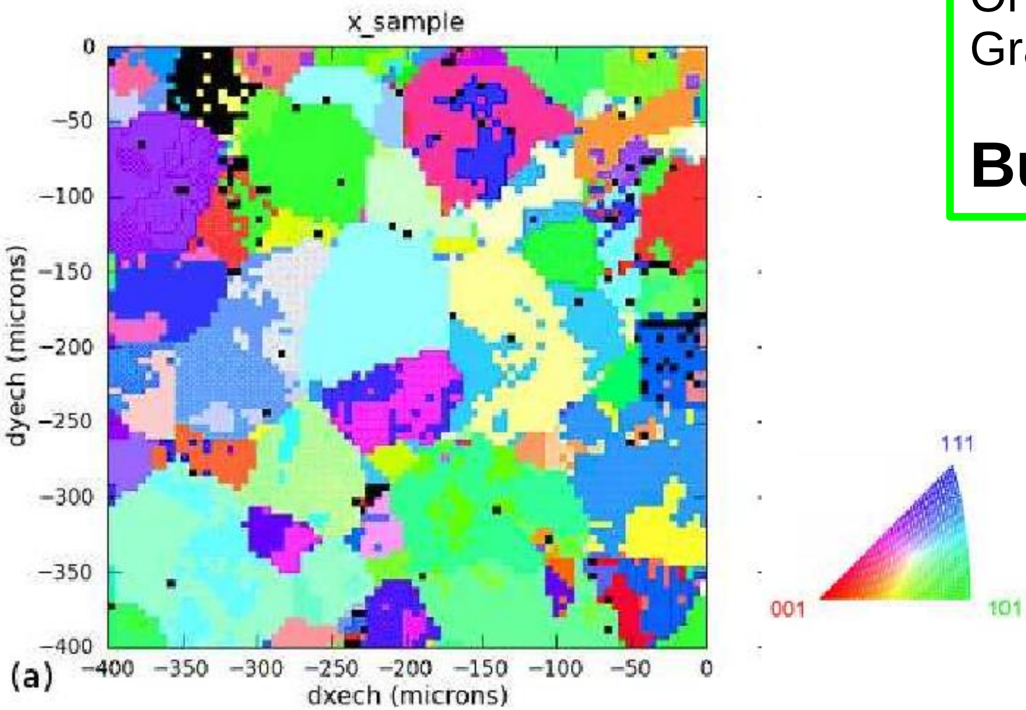


Fine Texture can be resolved (at least manually)



µLaue | Second steps: (1) Indexation & (raw) Orientation

Orientation map



V. Consonni *et al*, Phys. Rev. B 89 (2014) 035310

Useful to Correlate

scattering – **spatial** informations

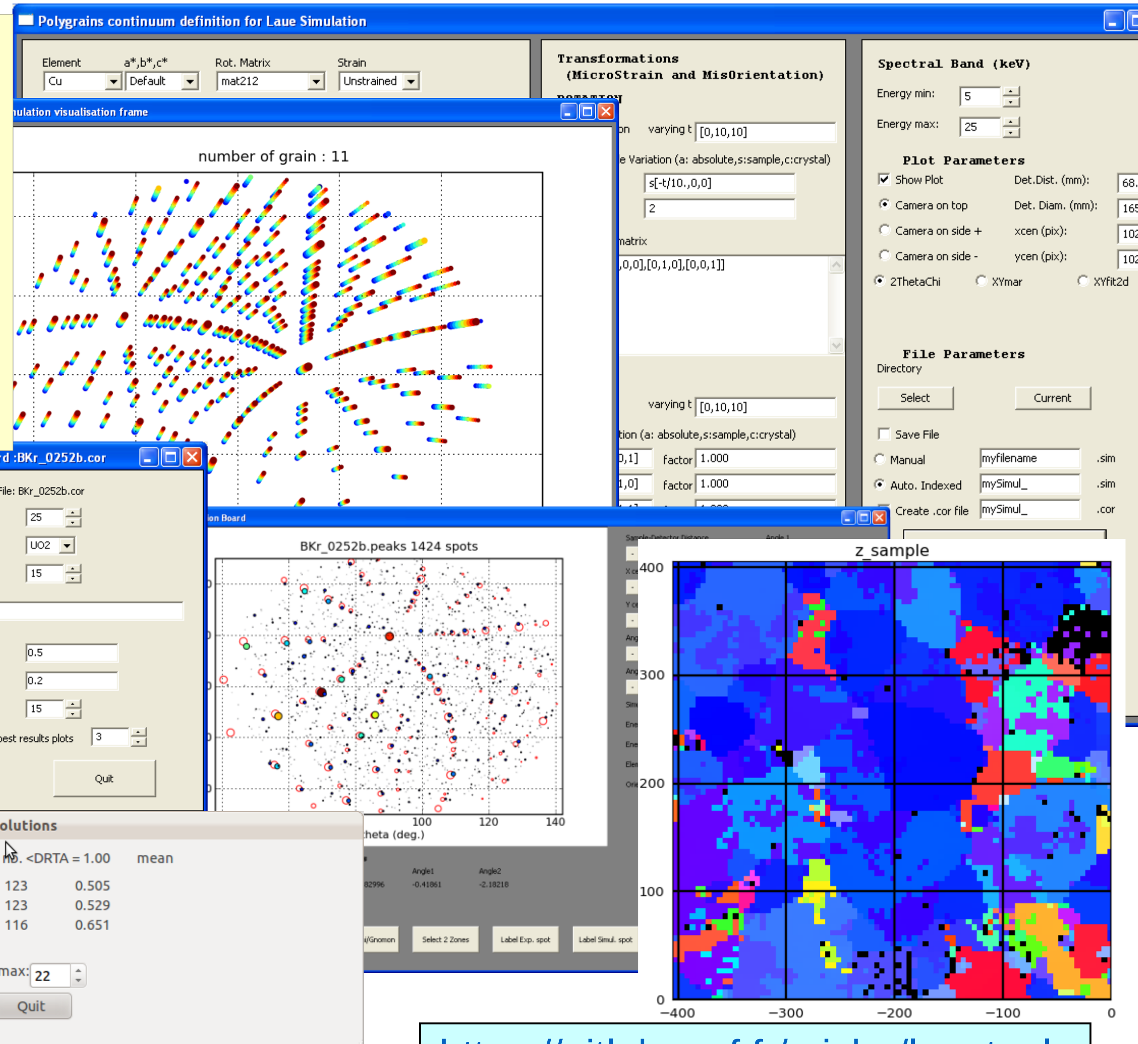
Orientation, texture,
Grains size, shape, positions, phase

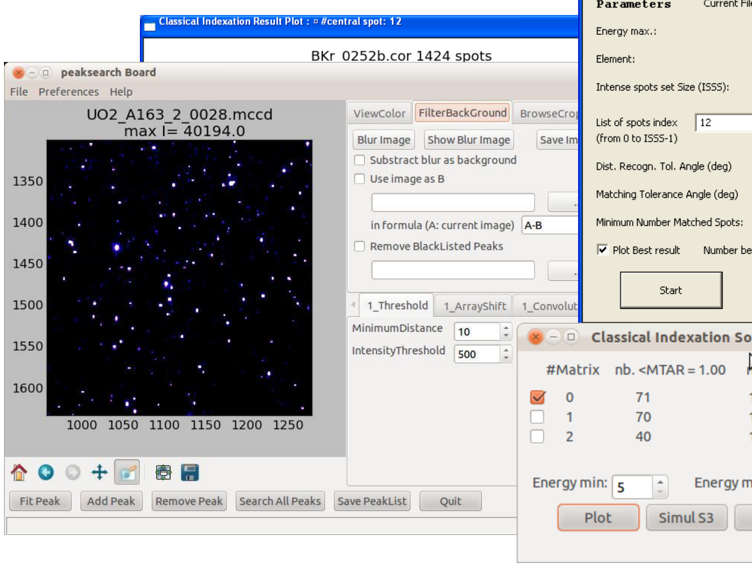
But Also: strain, defects,...

Laue Diffraction | Data analysis

LaueTools Suite Software for Laue microdiffraction data analysis

1- Peak Search
2- Indexing
3- Unit Cell Refinement
4- Map results plot
5- Automatic Analysis



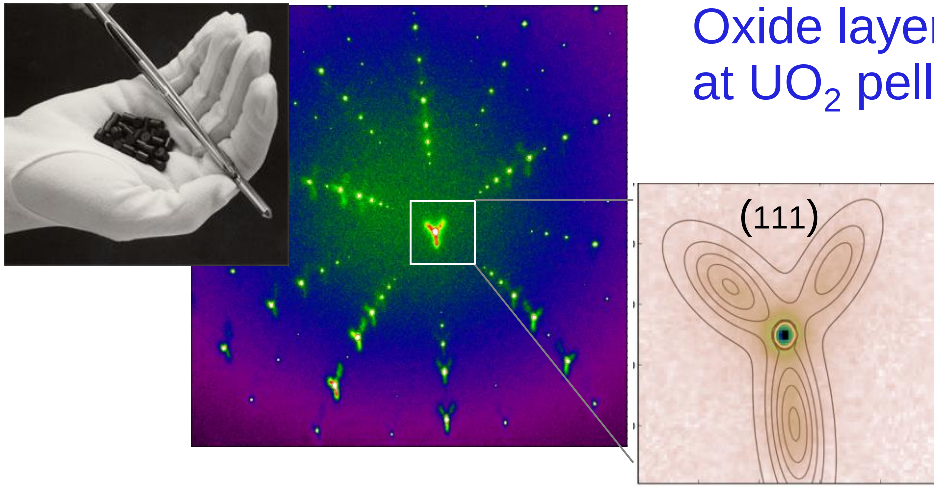


J.S. Micha et al, to be submitted to J. Appl. Cryst.

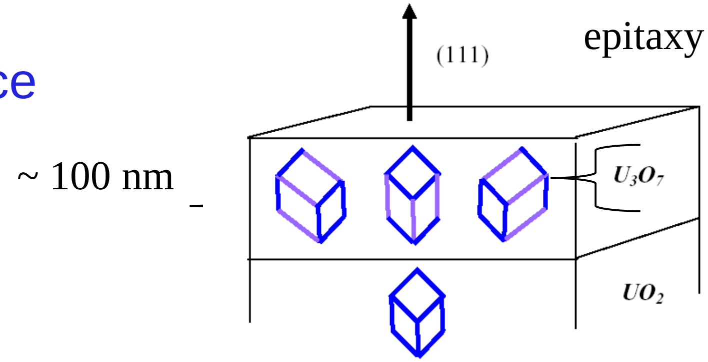
https://gitlab.esrf.fr/micha/lauetools

http://sourceforge.net/projects/lauetools/

µLaue Diffraction | Phase Identification



Oxide layer growth
at UO_2 pellet surface

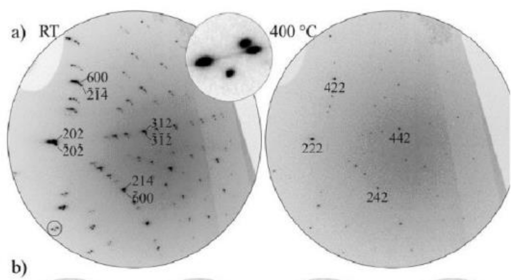


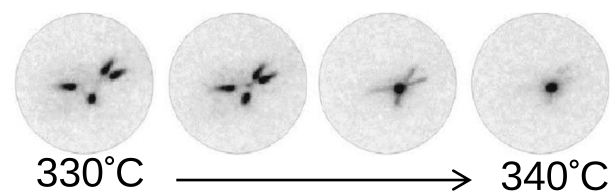
L. Desgranges *et al.*,J. Nucl. Mat. 402 p 167 (2010)

Single shot measurement :
3 variants with quadratic strain c/a ~ 1.03

In situ Phase transitions of
$(GeTe)_nSb_2Te_3$ (6 <= n <= 15)

DVD





Spatially resolved study of
defects and distorted structures distribution

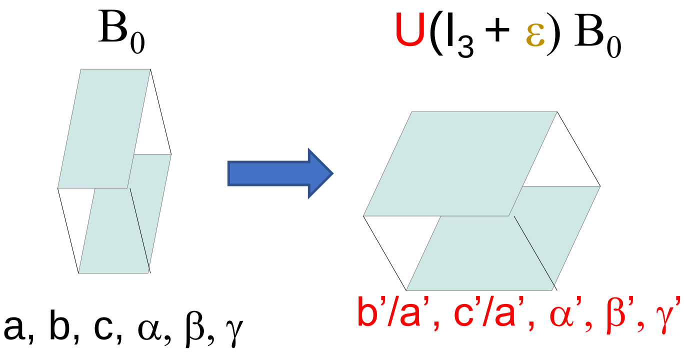
M. Schneider *et al*, Chem. Comm. 48 (16), 2192 (2012)

µLaue | Second steps: (2) Strain & Orientation refinement

Strain refinement by least squares minimization
- needs refined detector geometry (calibration)
- provides :

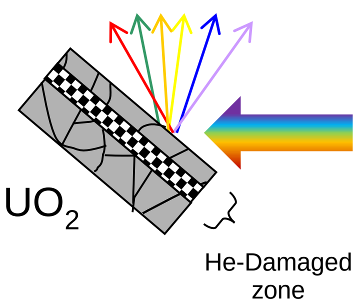
– unit cell deviatoric strain **ε***
– crystal orientation **U**

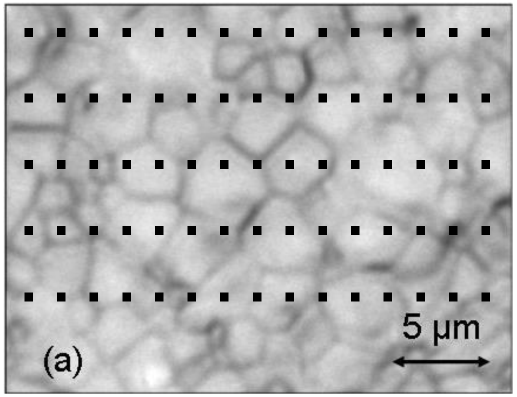
$$\mathbf{q} = \mathbf{U}(I_3 + \boldsymbol{\varepsilon})\, B_0\, \mathbf{G}^* = \mathbf{k_f} - \mathbf{k_i}$$

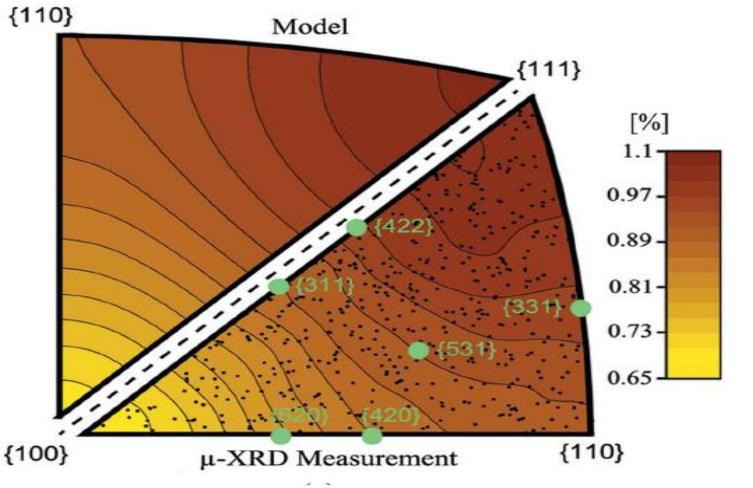


Rotation
& deformation

Normal-to-surface strain level
in polycrystalline He-implanted UO_2





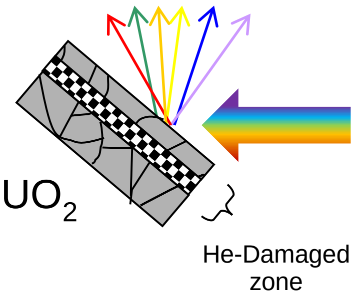


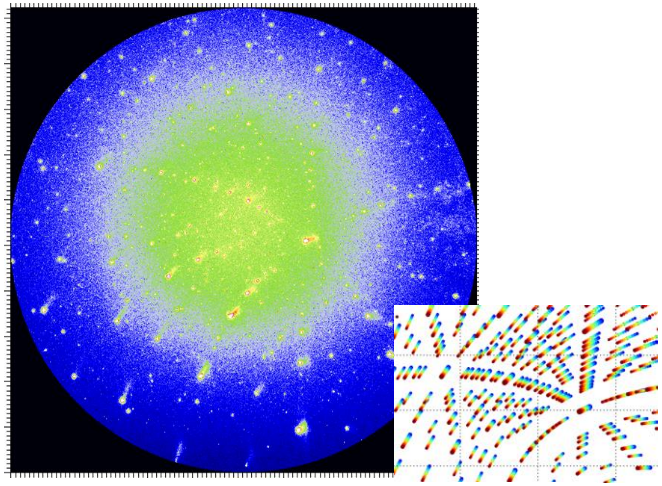
M. Ibrahim et al., accepted J. Appl. Cryst. (2015)
A. Richard et al., Nucl. Instr. Meth. B 326 (2013) 251
A. Richard et al., J. Appl. Cryst. 45 (4) (2012) 826

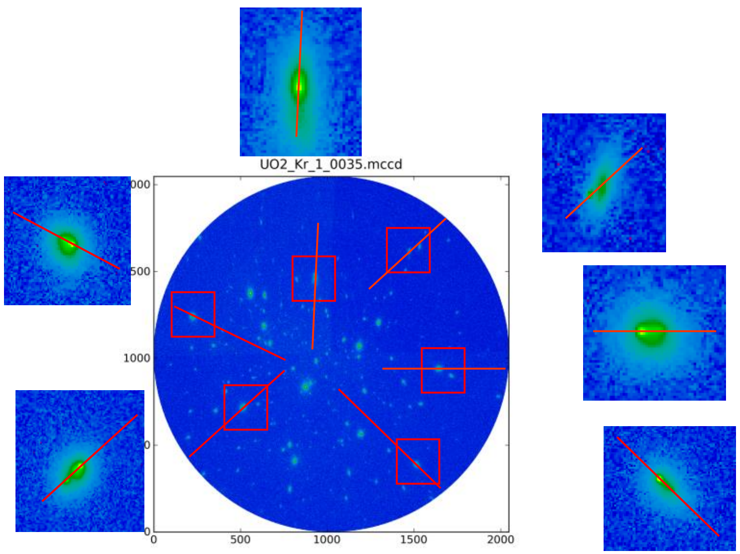
Mechanical properties under load
Data => input for FEA

μLaue Diffraction | Peak shape Analysis

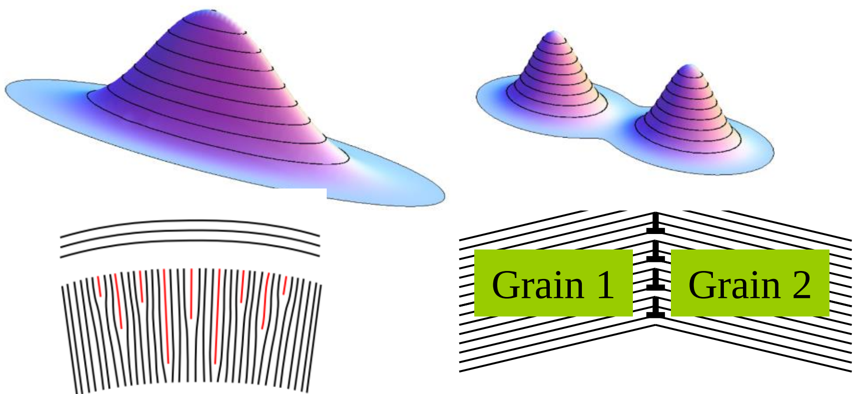
Defects induced by He implantation in UO_2 polycrystal







Radial streaking => Surface swelling



Geometrically **N**ecessary **D**islocations

⇒Intragrain fine misorientation
⇒Activated slip system

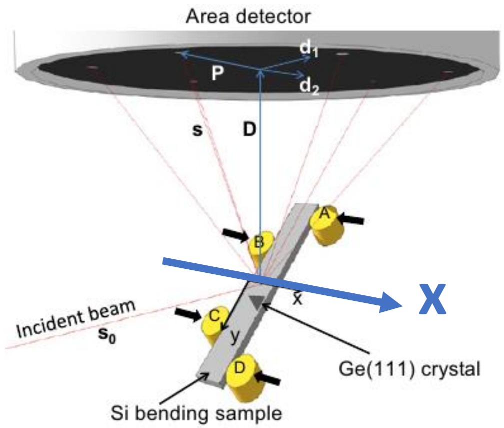
=> Stress release modes, Slip system recognition

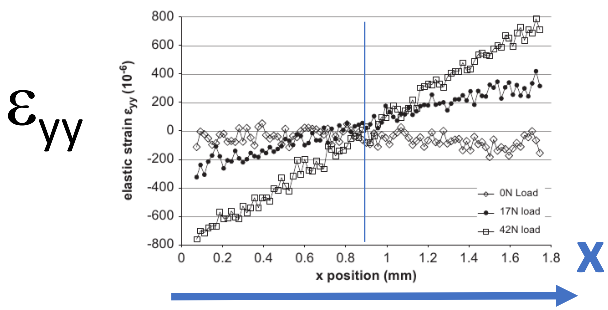
A. Richard et al., J. Appl. Cryst. 45 (4) (2012) 826

µLaue Diffraction | *in situ* mechanical test

Data measurements & analysis validation
Errors estimation

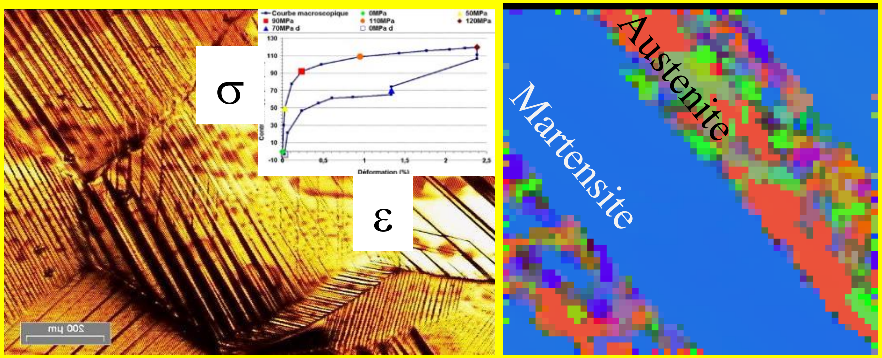
In situ **elastic** bending





F. Hofmann *et al*, Nucl. Instr. Meth. A 660 (2011) 130
E. Plancher Thesis 2015

Reversible phase transformation
(SMA)
during cycle in CuBeAl

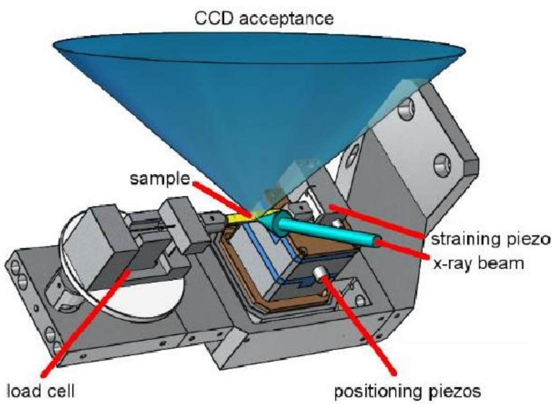


S. Berveiller *et al*, Acta Mat. 59 (2011) 3636
B. Malard, Compt. Rend. Physique 13 (2012) 280–292

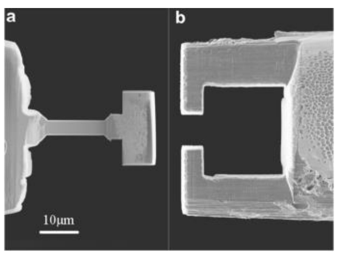
- Validation:
- uncertainties on strain
- reliability of data analysis chain

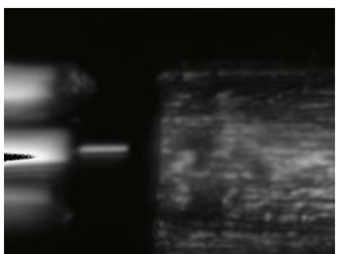
- In situ structural characterization
- Determine behaviour law

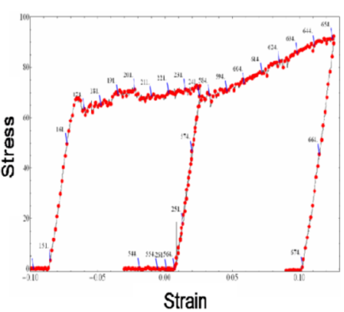
µLaue Diffraction | *in situ* mechanical test

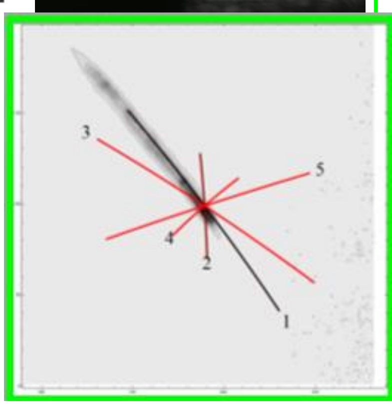


Compatible SEM & Synchrotron BL

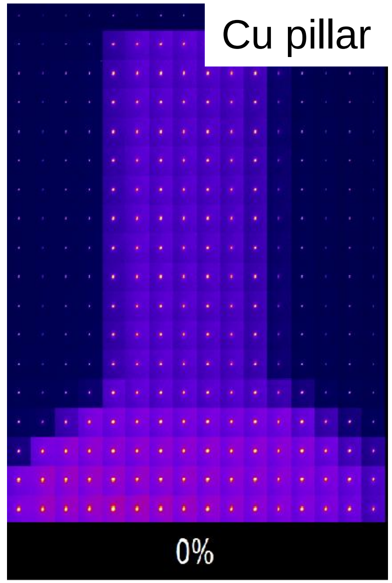


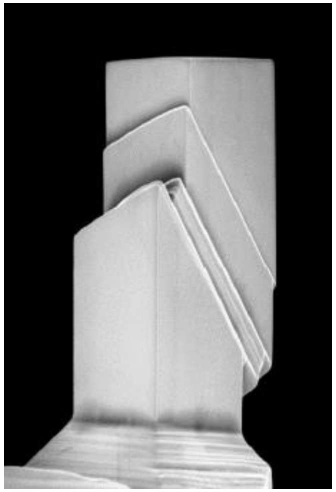


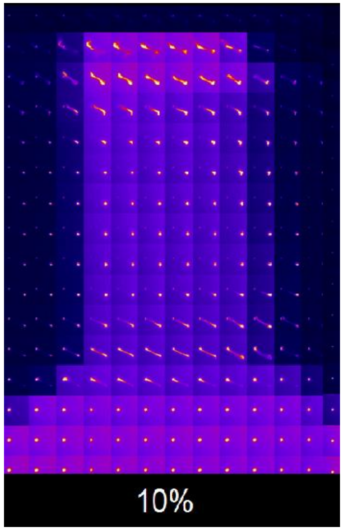




In situ plasticity







Well controlled micromechanical tests
- Crystal & GB orientation
- Fine Force resolution
- Tension, compression, bending, torsion,...

Measurements of **peak elongation direction and width**
→ Activated slip systems
→ Dislocations density

See C. Kirchlechner et al, Acta Mat (2011) to (2016)

µLaue Add-ons | Going beyond phase/orientation/strain determination

- Full stress/6 lattice parameters measurements

$$\varepsilon^* = \varepsilon - \bar{\varepsilon} I_3$$

Deviatoric strain
Standard Laue Pattern analysis

Full strain

hydrostatic strain

- Mechanical assumption e.g. $\sigma_{33} = 0$

  Solve $\sigma = C\varepsilon$ for $\bar{\epsilon}$ and 5 σ_{ij}

- Laue spot energy fine experimental measurement: $\epsilon_{hkl} = \bar{\epsilon} + (\epsilon^* \tilde{\mathbf{G}}).\tilde{\mathbf{G}}$

  Determine $\bar{\epsilon}$ then ε then $\sigma = C\varepsilon$

  How?

  insert monochromator (optics hutch)
  use of a 0D movable energy-resolved detector
  insert monochromator (diamond) in transmission (in exp. Hutch)

- Depth resolution: **3D** microscopy

From the deviatoric to the full strain tensor

Hypothesis : No stress perpendicular to the free surfaces (σ_{33} = 0)

Hooke's law **F** = k**x** … *with tensors and Voigt's notation:*

$$\begin{pmatrix} \sigma_{11} \\ \sigma_{22} \\ \sigma_{33}=0 \\ \sigma_{23} \\ \sigma_{13} \\ \sigma_{12} \end{pmatrix} = \begin{pmatrix} c_{11} & c_{12} & c_{12} & 0 & 0 & 0 \\ c_{12} & c_{11} & c_{12} & 0 & 0 & 0 \\ c_{12} & c_{12} & c_{11} & 0 & 0 & 0 \\ 0 & 0 & 0 & c_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & c_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & c_{44} \end{pmatrix} \begin{pmatrix} \varepsilon'_{11}+\varepsilon_h/3 \\ \varepsilon'_{22}+\varepsilon_h/3 \\ \varepsilon'_{33}+\varepsilon_h/3 \\ 2\varepsilon'_{23} \\ 2\varepsilon'_{13} \\ 2\varepsilon'_{12} \end{pmatrix}$$

$1,2,3 = x, y, z = [100], [010], [001]$

σ = stress (unknown)

c_{11}, c_{12}, c_{44} = 126, 44.0, 67.7 GPa = elastic coefficients of Ge

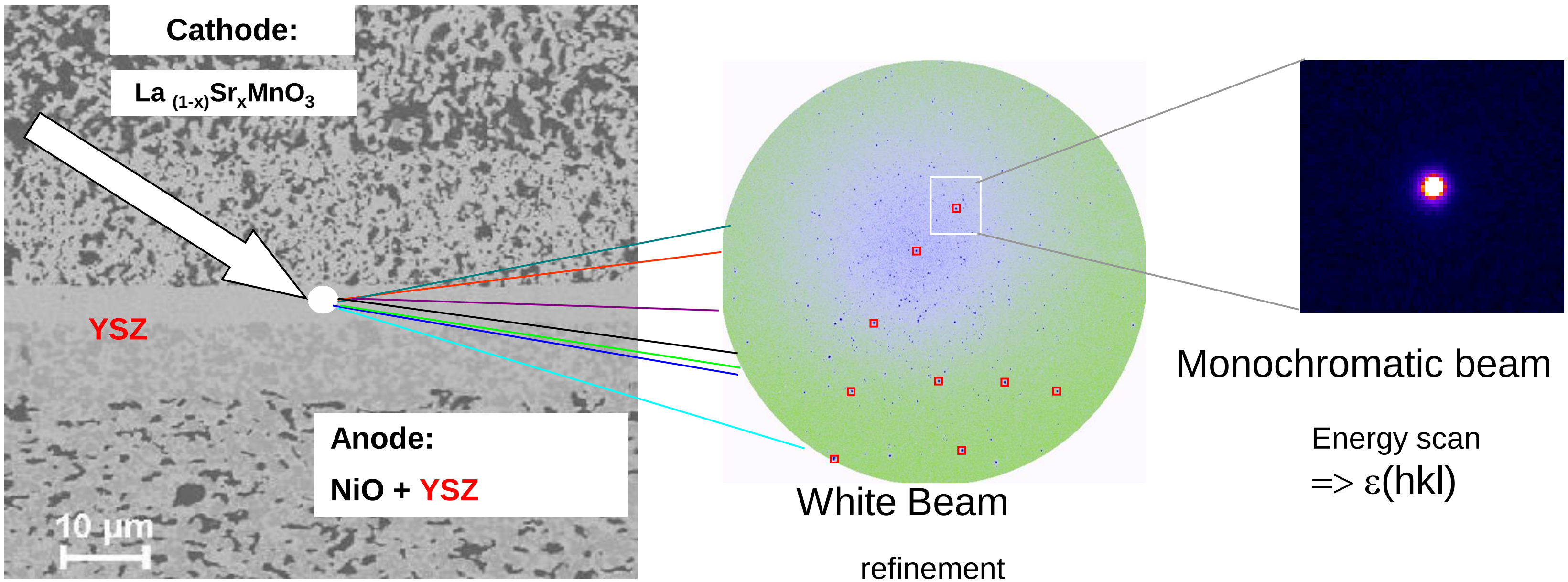
ε' = deviatoric strain (measured)

ε_h = hydrostatic strain (unknown)

Solve the 6 equations for the 6 unknowns to get the full strain tensor

μLaue Diffraction | Full Stress

Spatial distribution of full stress tensor in polycrystalline ZrO_2-based electrolyte for Solid Oxide Fuel cell



Electrolyte YSZ : $(ZrO_2)_{1-x}(Y_2O_3)_x$

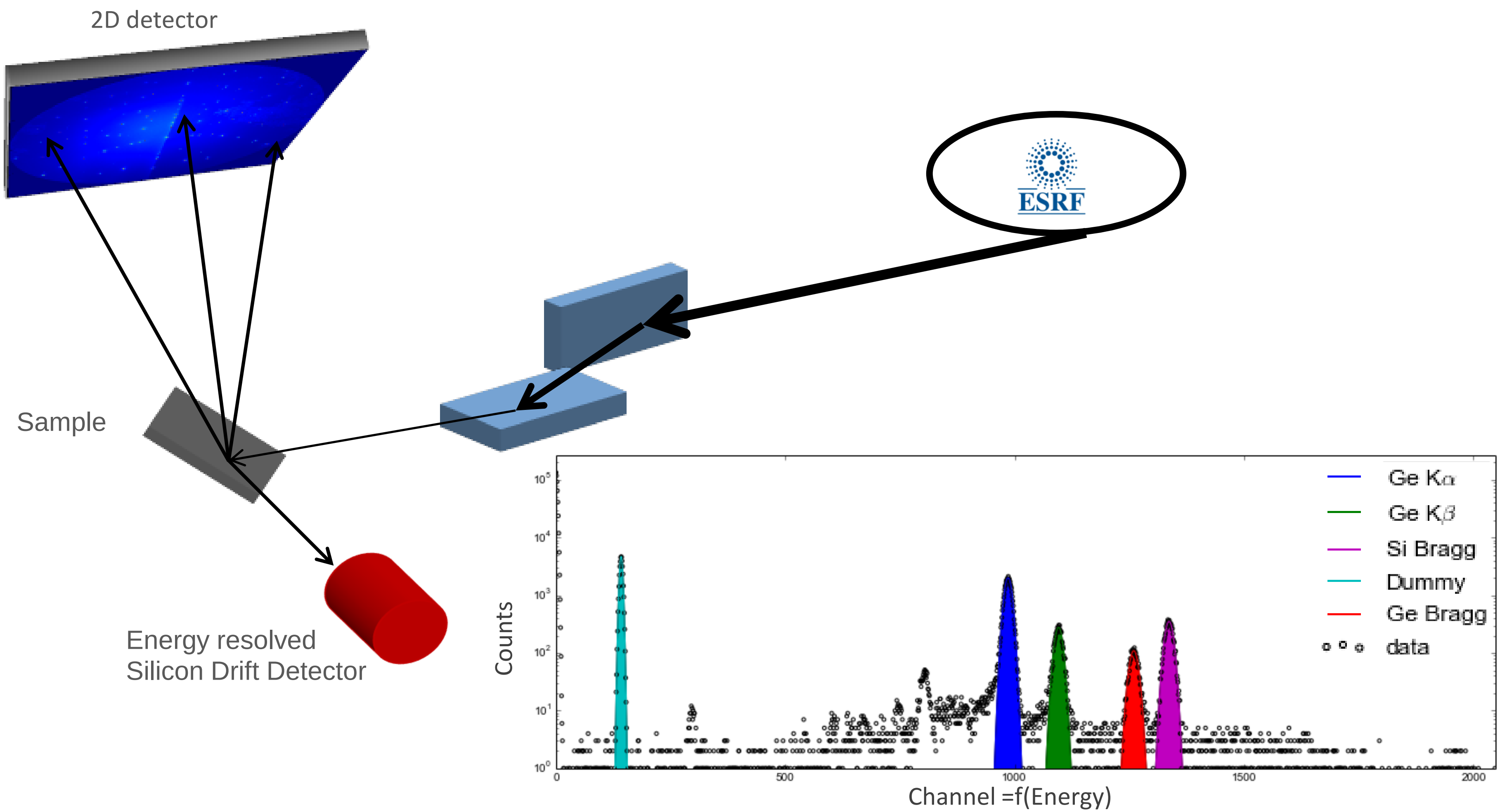
Full tensors $\sigma = C\varepsilon$

$$\sigma = \begin{pmatrix} -986 & -35 & 30 \\ -35 & -878 & 25 \\ 30 & 25 & -209 \end{pmatrix} \text{Mpa}$$

Local stress agrees with macroscopic stress (by $\sin^2\psi$)

J. Villanova et al, Nucl. Inst. Meth. Phys. B 268 p 282 (2010)
J. Villanova et al, J. Appl. Cryst. 45 (5) p 926-935. (2012)

Direct energy measurement with energy-resolved detector (SDD)

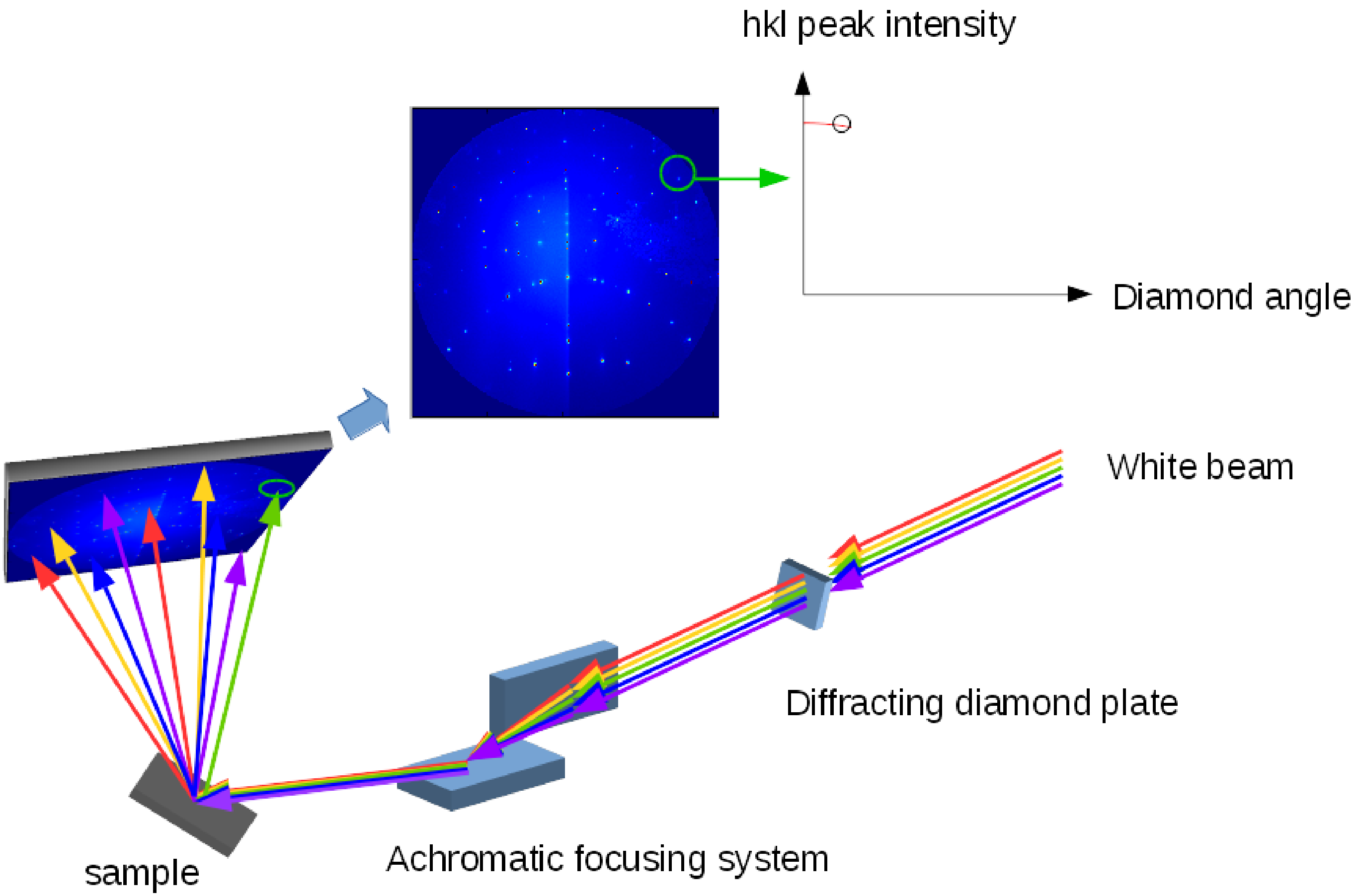


Local energy spectra acquired with the Laue diffraction pattern (no overhead)

Energy calibration required (using fluorescence or known diffraction peaks)

Quest for the last parameter:
The Rainbow filter technique

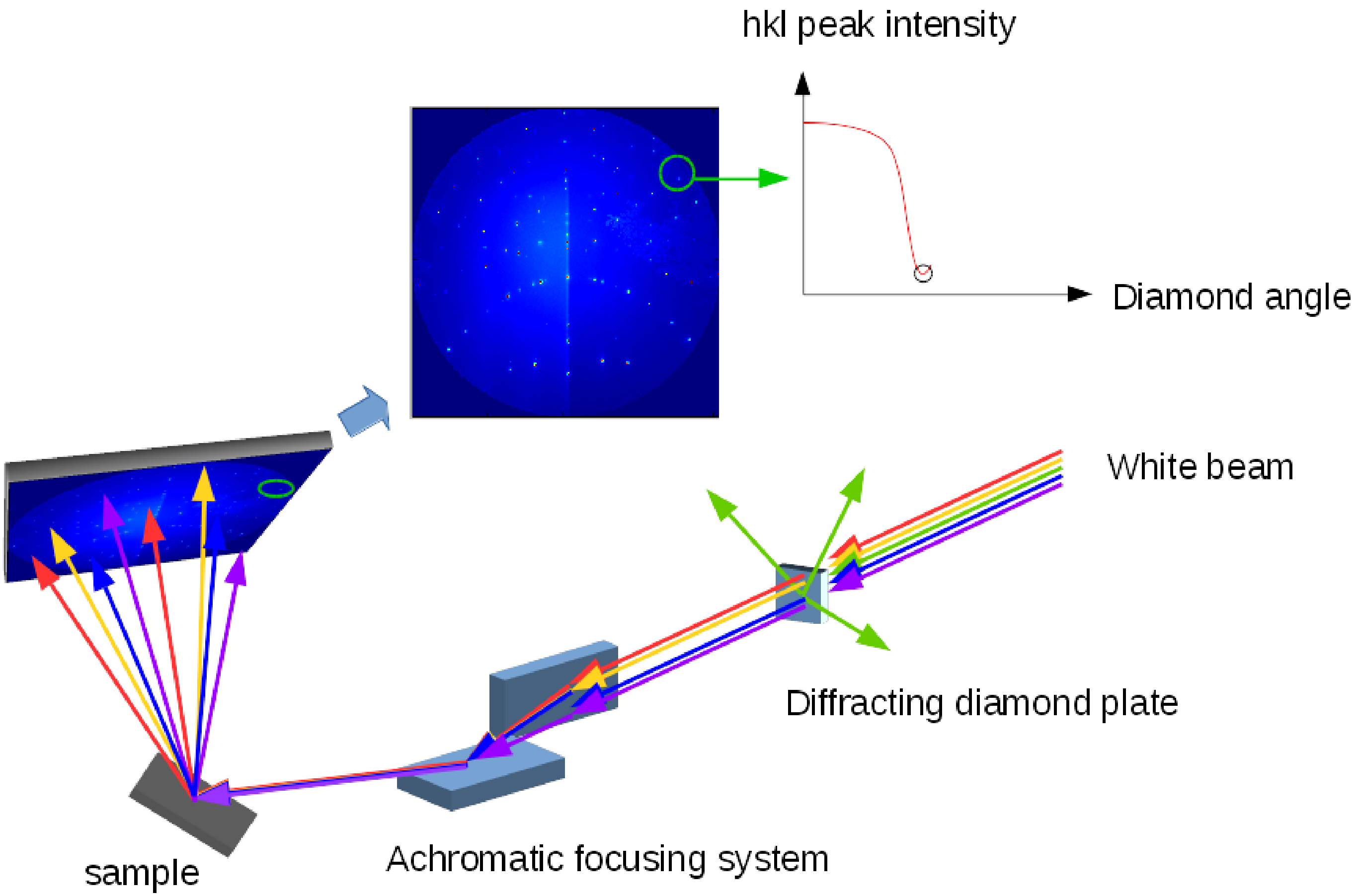
"Inverse" monochromatic diffraction



Robach *et al.*, Acta Cryst. (2013). A**69**, 164-170

Quest for the last parameter:
The Rainbow filter technique

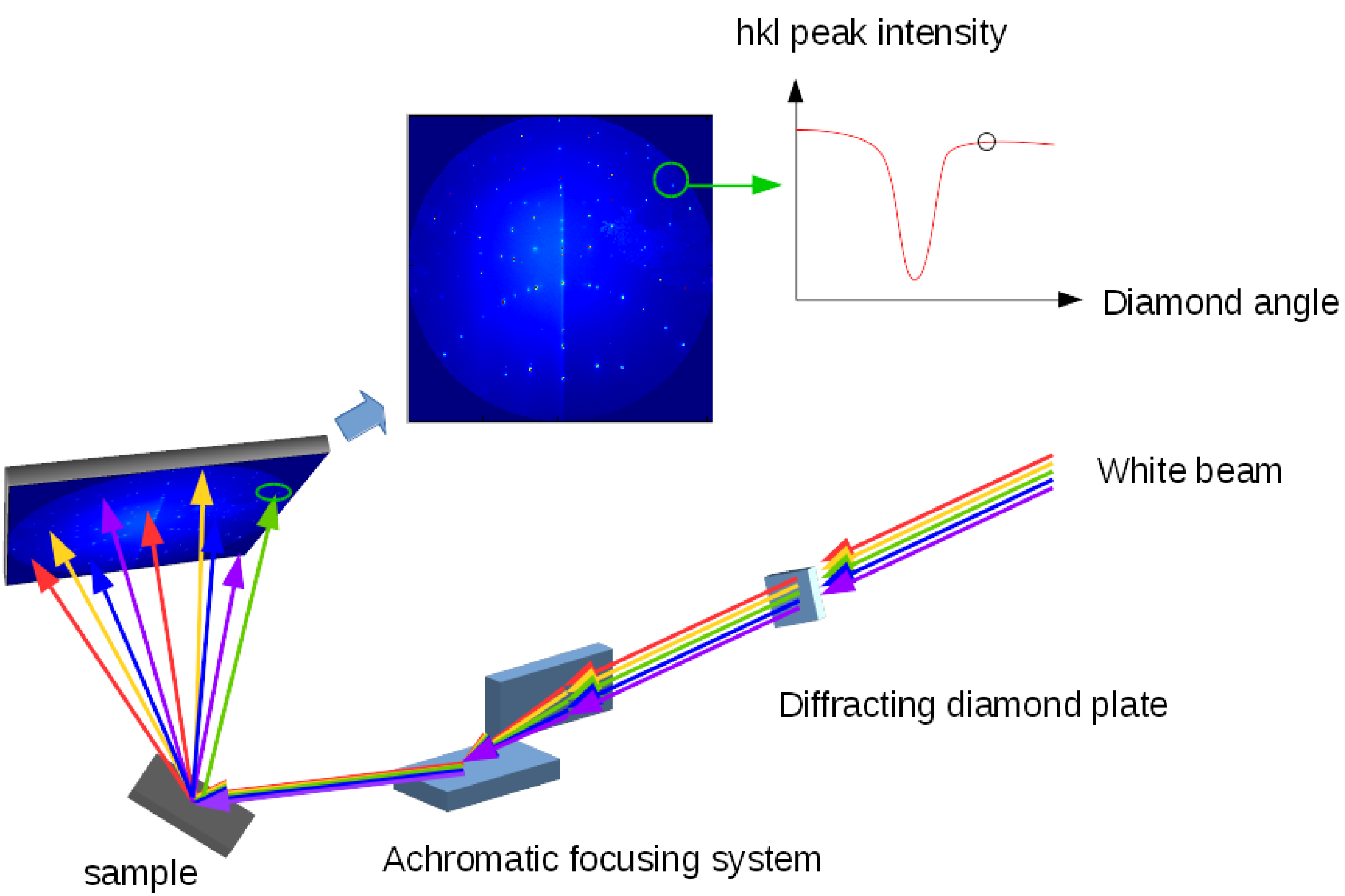
"Inverse" monochromatic diffraction



Robach *et al.*, Acta Cryst. (2013). A**69**, 164-170

Quest for the last parameter:
The Rainbow filter technique

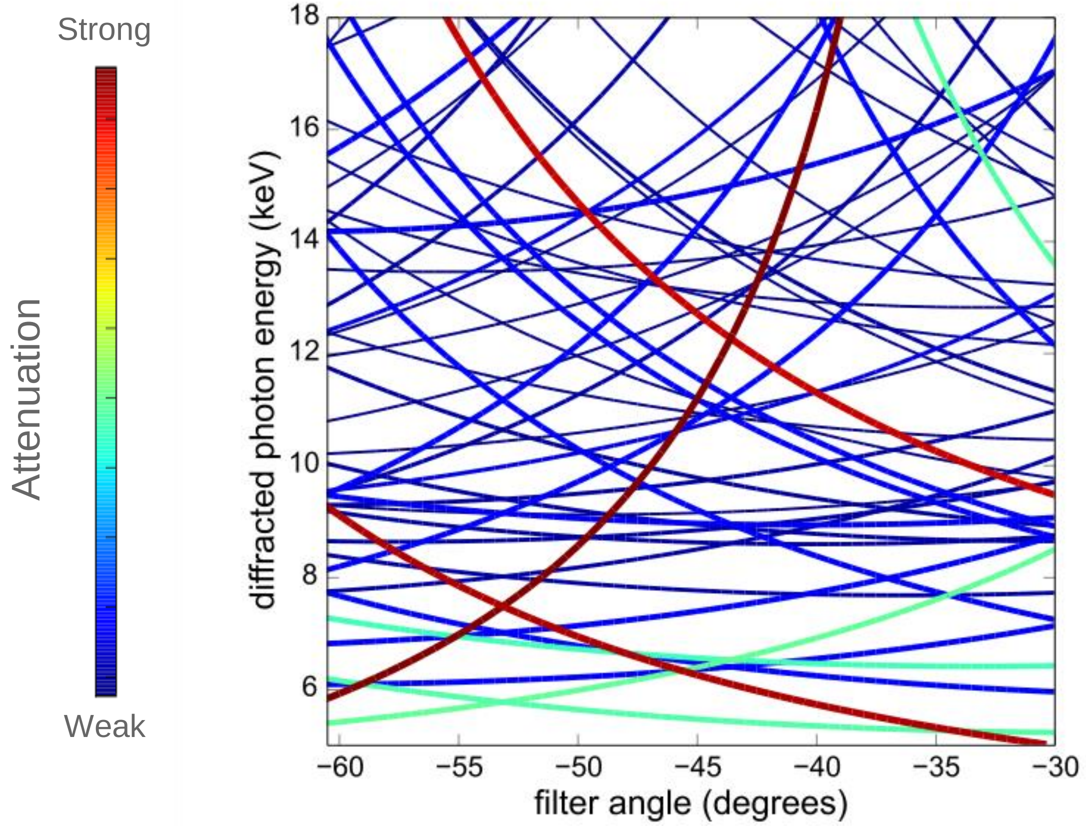
"Inverse" monochromatic diffraction



Robach *et al.*, Acta Cryst. (2013). A**69**, 164-170

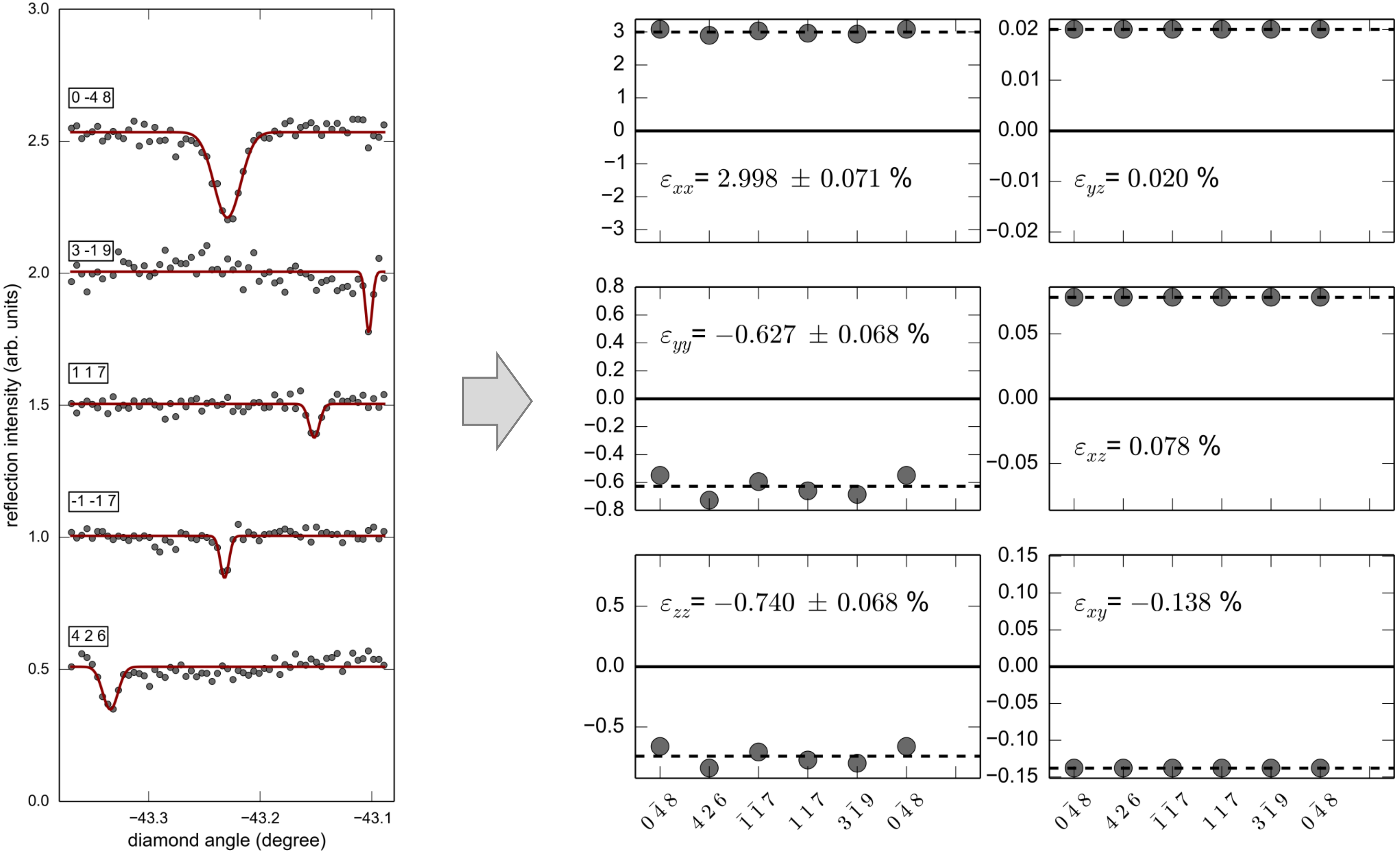
The Rainbow filter technique

Relationship between filtered out energies and diamond filter angle

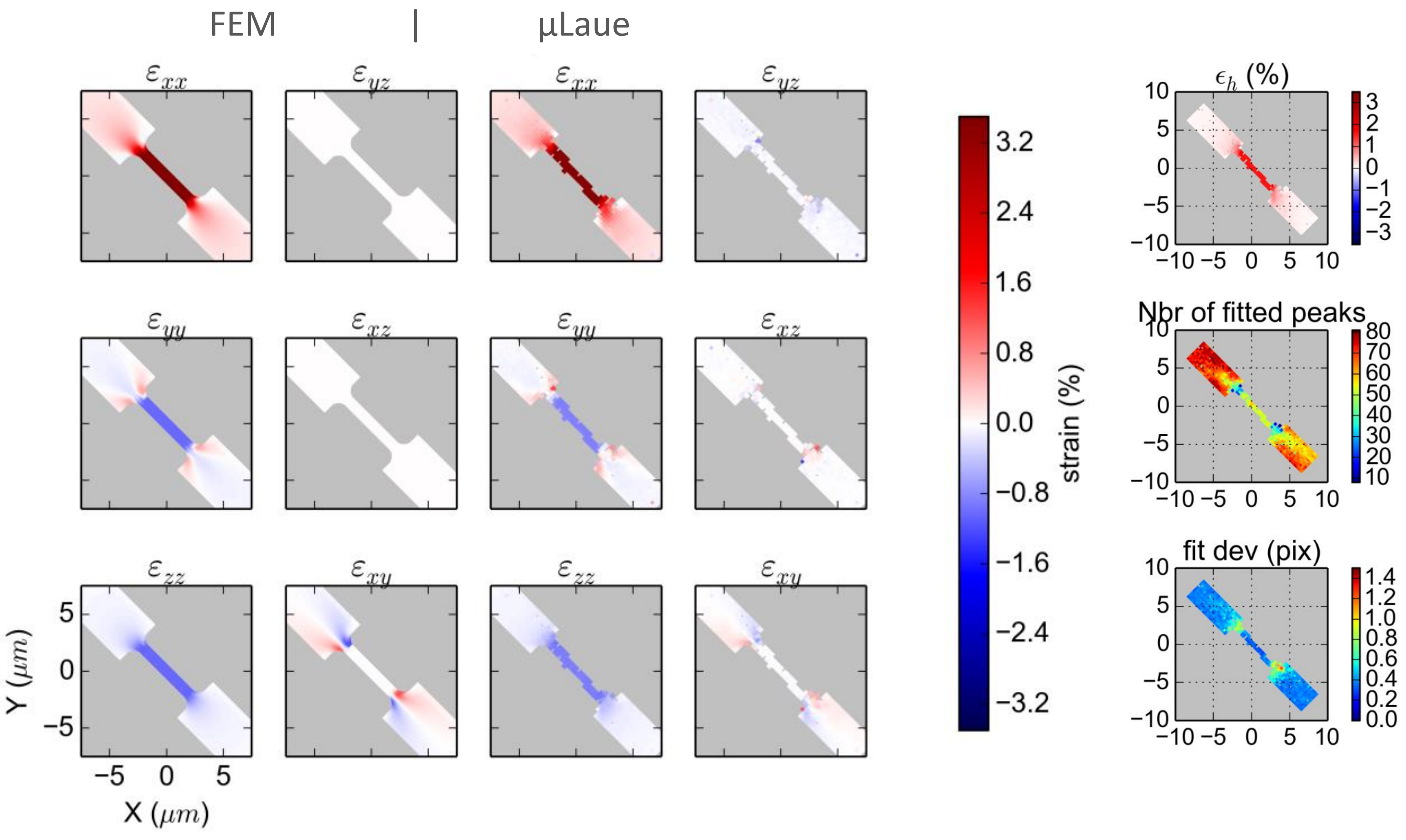


The full bandwith of the beamline (5-25 keV) can be covered

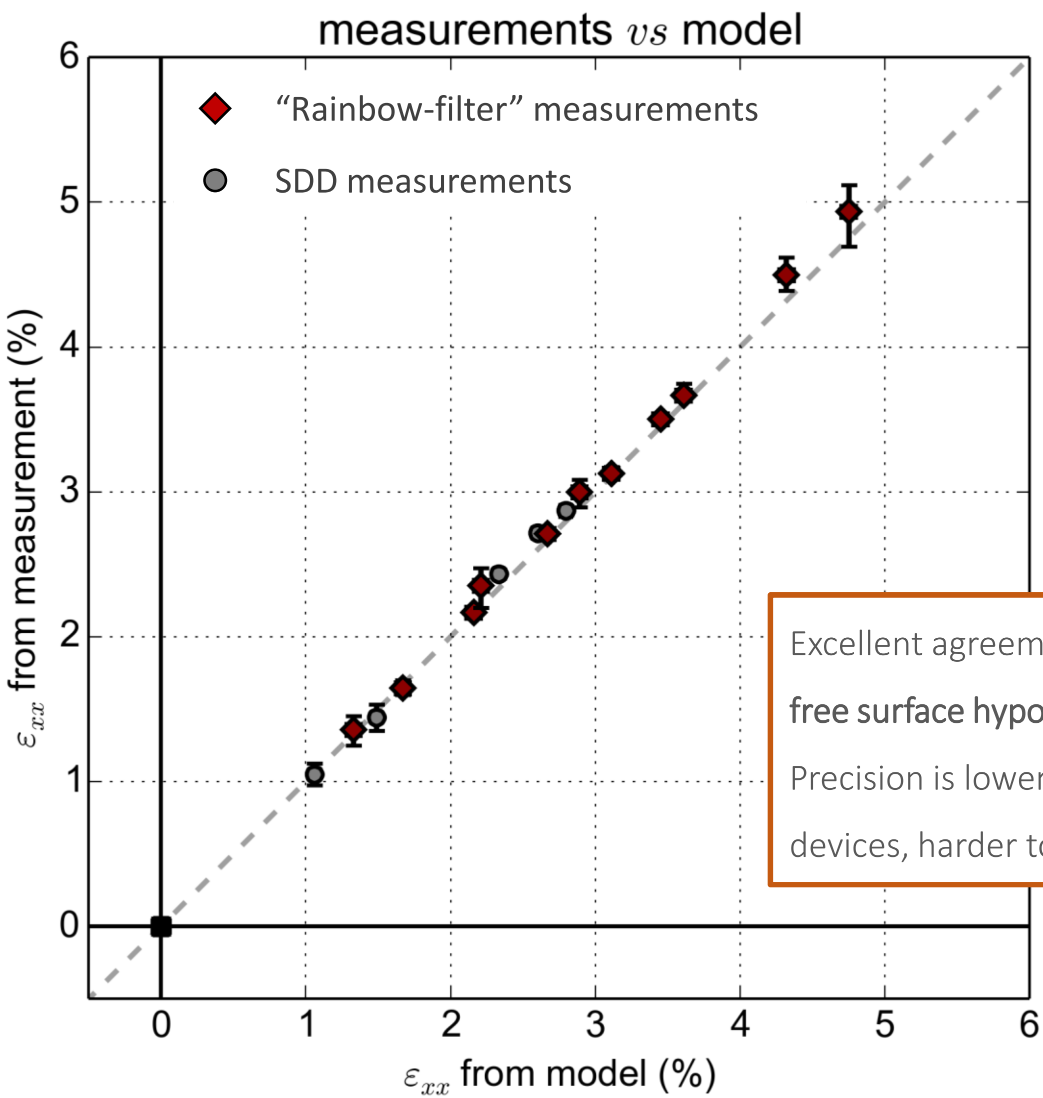
Rainbow filter technique: full strain tensor determination



XRD measurements *vs* FEM simulations



Stress free surfaces hypothesis *vs* rainbow technique

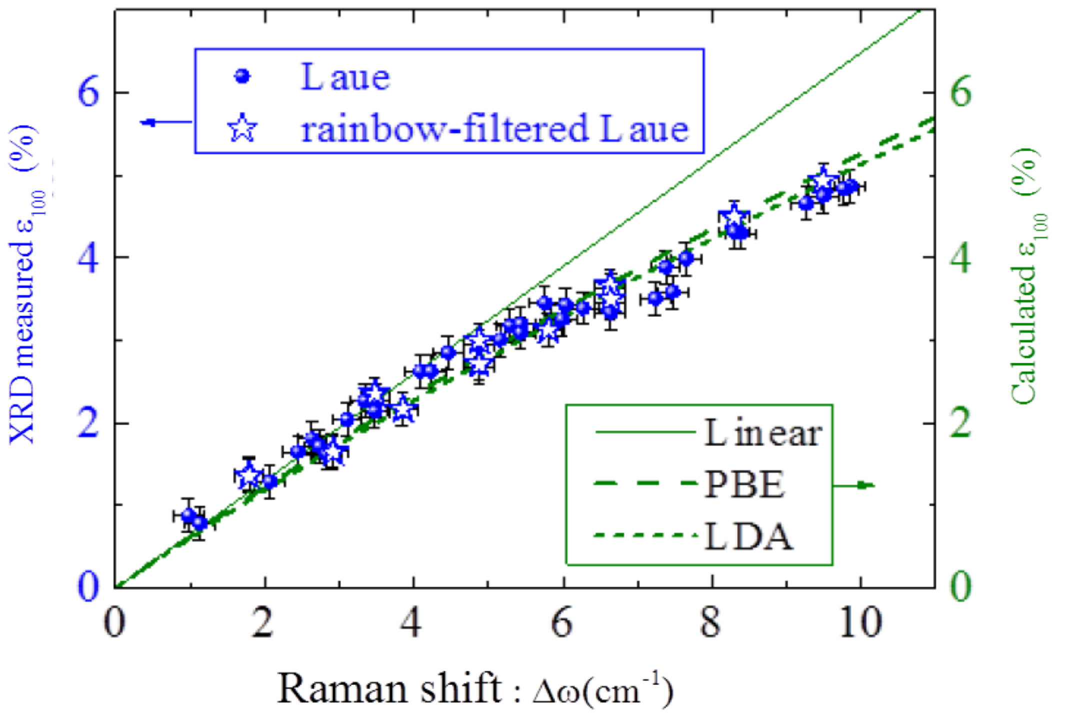


Excellent agreement up to 5 % strain, the **stress-free surface hypothesis is valid!**

Precision is lower at higher strain (smaller devices, harder to measure)

Evidencing non-linear strain effects in Ge

Raman spectral shift for <100> uniaxial stress

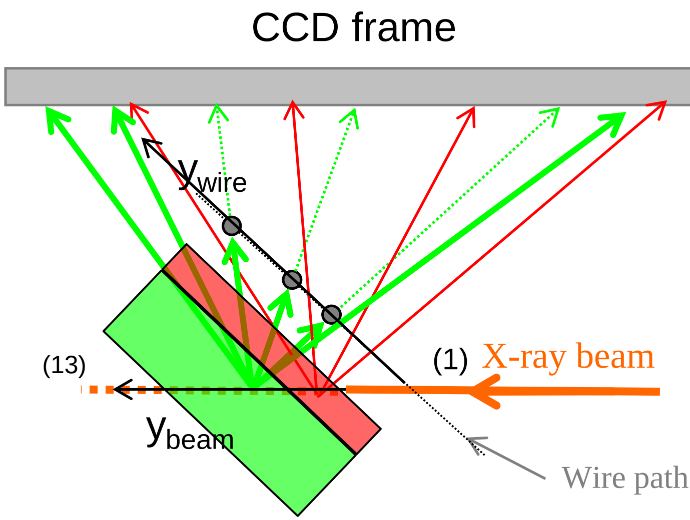


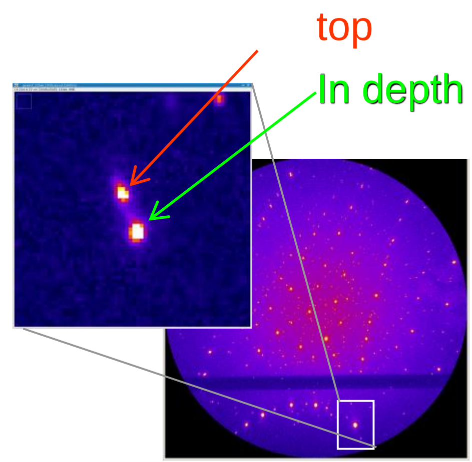
Strain measurements: µLaue as reference for Raman spectroscopy

A. Gassenq, *et al.*, APL **108**, 241902 (2016)

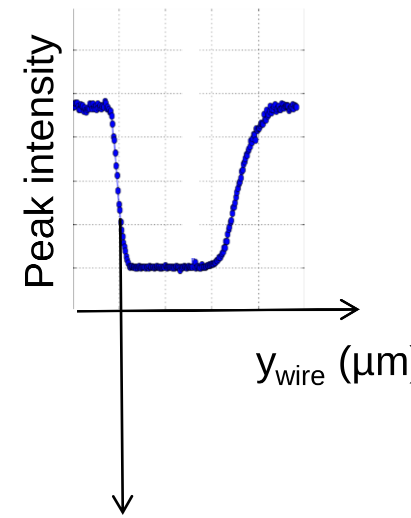
μLaue Diffraction | towards 3D

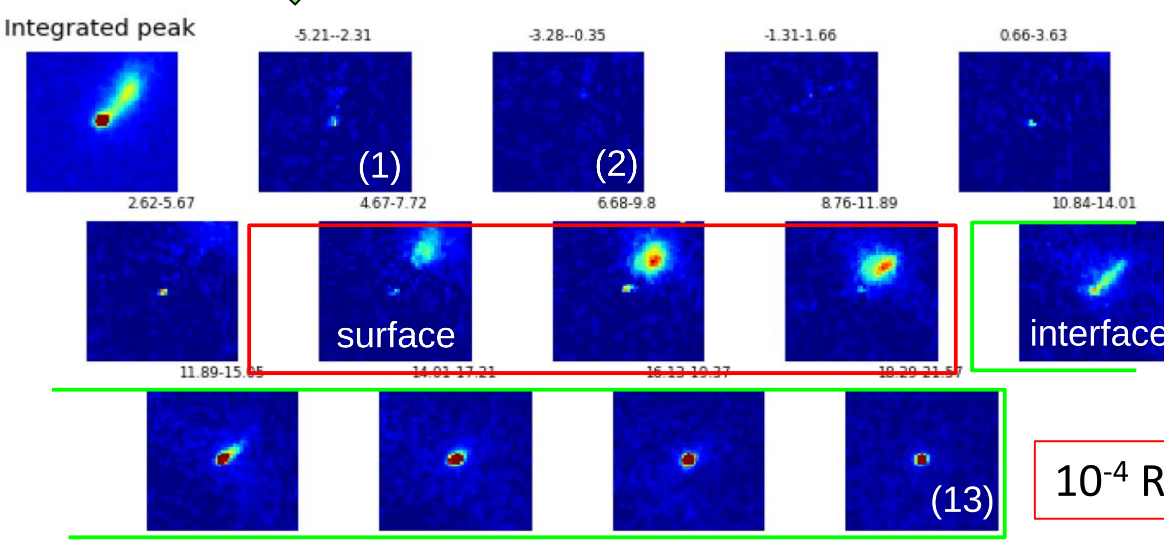
Determine orientation & strain gradients in depth





Depth resolution: 0.3μm





1- Ybeam position of scattering source
Pixel intensity = f(y_{wire})

2- Reconstruction:
Laue pattern = f(y_{beam})

10^{-4} Resolution preserved in depth

µLaue Diffraction | Getting more & more accurate

Combine µLaue with

DIC
Combine local and macro strain fields

EBSD
EBSD- based selection & Laue Measurements

Improve µLaue with

2D energy resolved detector
Faster and more accurate Full stress measurements

Laue DIC
Enhance relative strain accuracy

Beam scan
Keeping sample free during in situ experiments

µLaue Diffraction | Conclusions

X-ray Laue Microscopy

- Intense, narrow beam (<500x500 nm²)
- Complementary x-ray white and energy resolved (5 – 23 keV)
- Dedicated installation :

for basic 2D orientation map to advanced quantitative structural data

- 'simple', 'fast', non destructive, *in situ* experiments

Open to a **large scientific community**

- Metallurgy, structure and functional materials
- µ-electronics & -device, µ & nano-object mechanics
- mineralogy, cultural heritage

Fundamental and **applied sciences**

- local structure
- metrology, control and reliability

µLaue Diffraction | Conclusions

µLaue @ ESRF

Next proposals submissions round September 2020

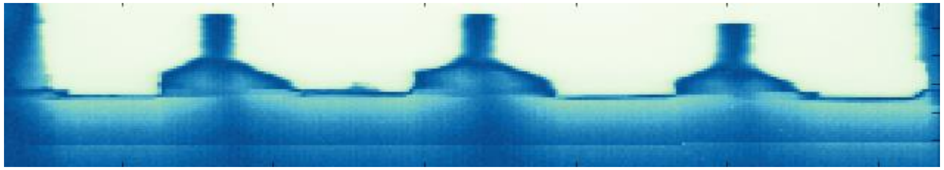
Access through CRG (SOLEIL) and program committees

http://sunset.synchrotron-soleil.fr/sun/

https://www.esrf.eu/UsersAndScience/UserGuide/Applying

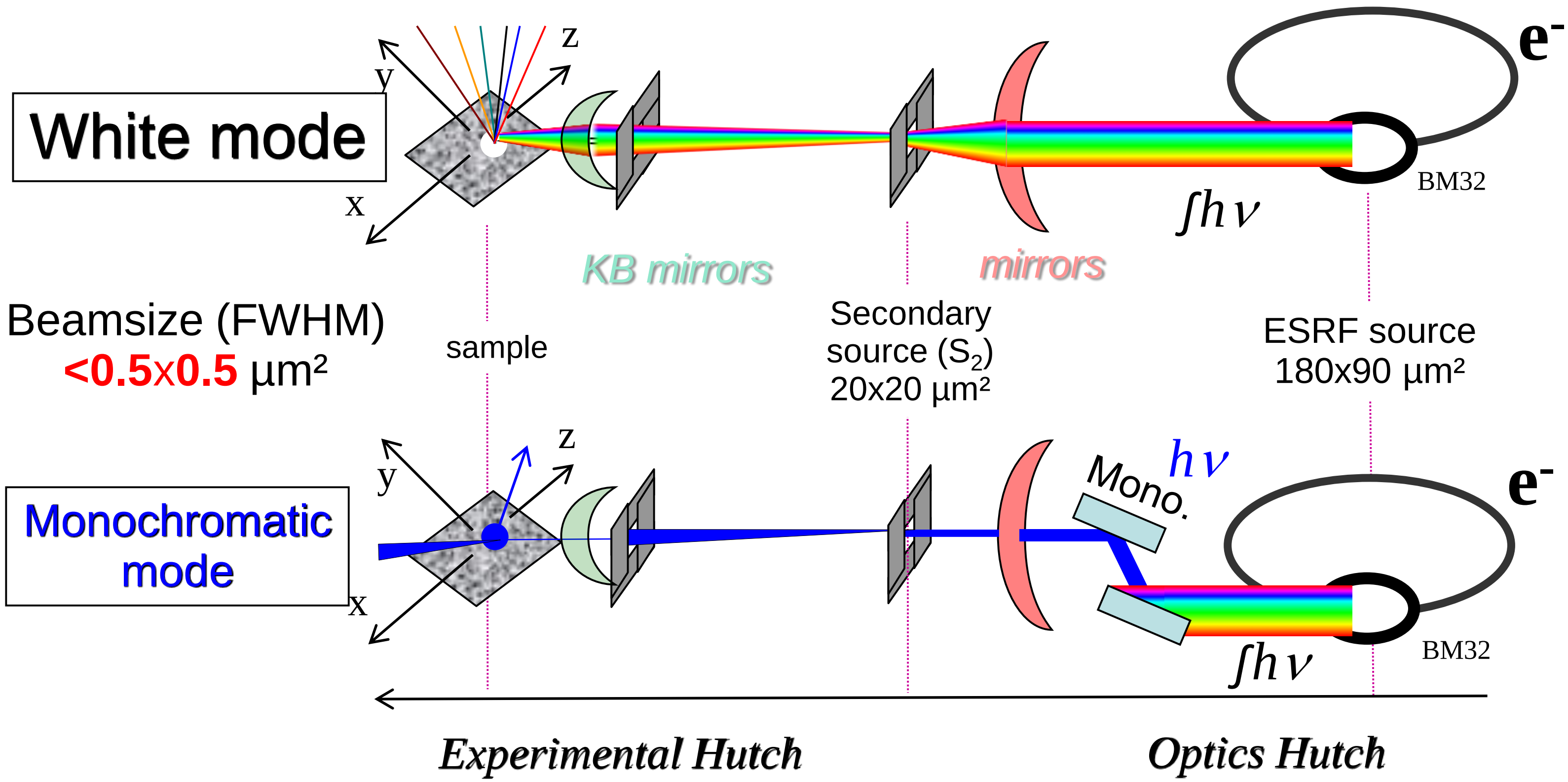
Open to free tests and collaborating developments !

Thank you for your attention



Appendix : µLaue | Optics setup

2 steps demagnification with secondary source

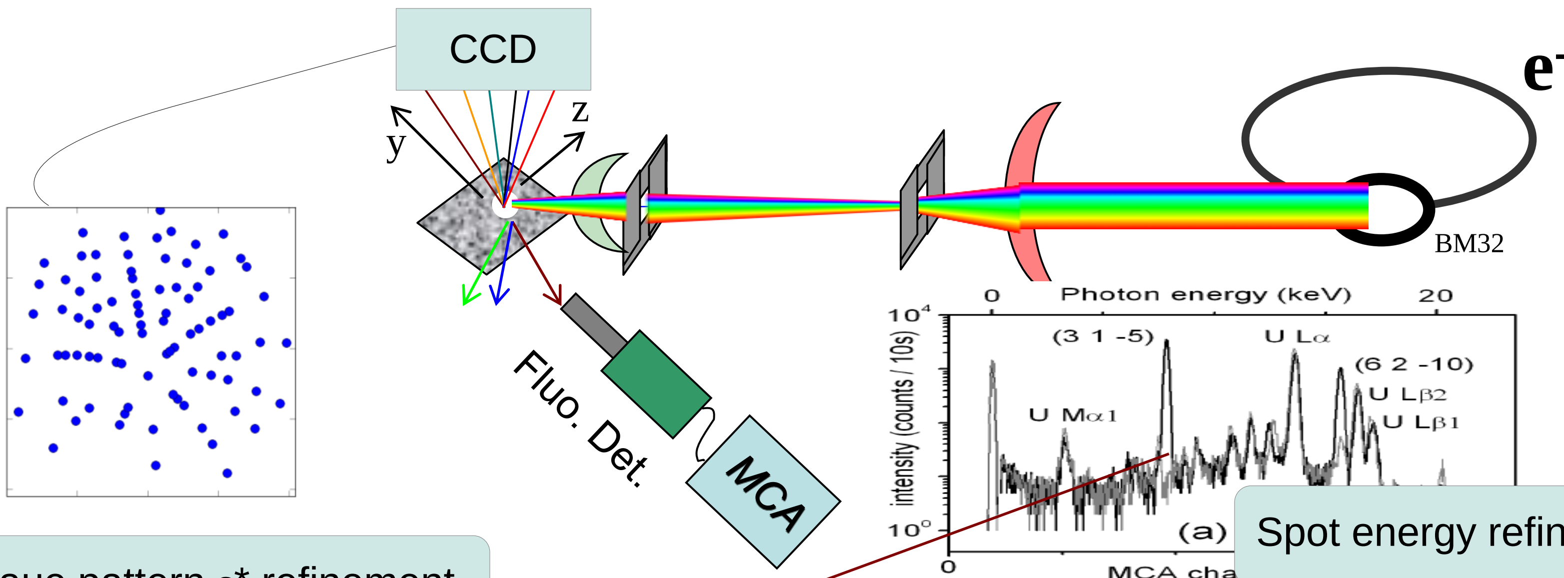


Fixed microbeam position and incident angles = Image of S_2

Appendix : µLaue Diffraction | Energy measurements

Movable Energy dispersive detector (SDD)

Beamsize (FWHM) **<0.5x0.5** µm²



Laue pattern ε* refinement

Spot energy refinement

$$\epsilon_{hkl} = \bar{\epsilon} + (\epsilon^* \tilde{\mathbf{G}}).\tilde{\mathbf{G}}$$

=> Determine $\bar{\varepsilon}$ then ε

Advantage:
Measurements of ε* and ε_{hkl} at same (**r**,t)
=> better accuracy
Several spot energy measurements
=> better statistics

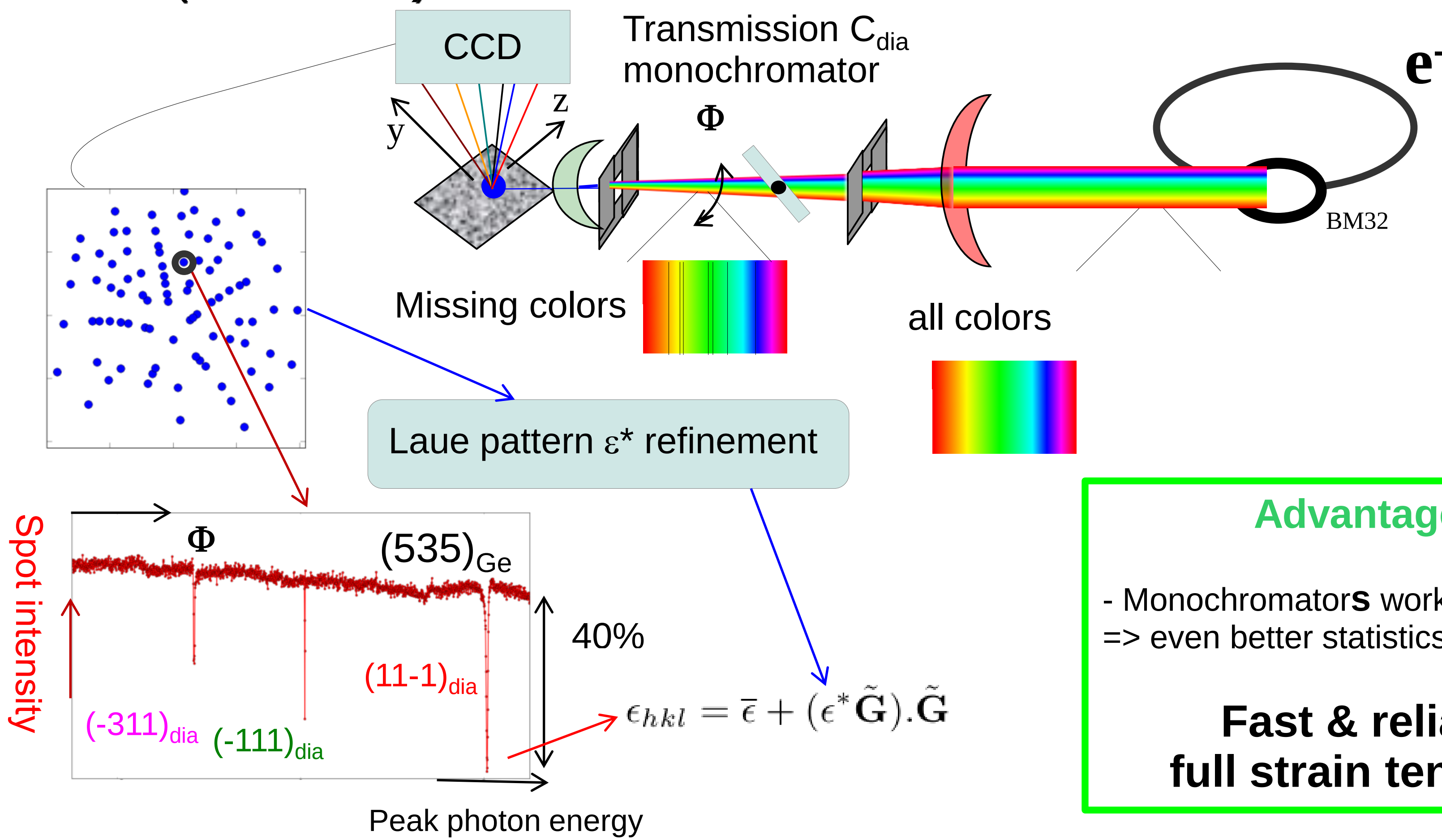
Fast & reliable
full strain tensor ε

O. Robach *et al*, J. Appl. Cryst. 44 (2011) 688

Appendix : µLaue Diffraction | Energy measurements

Monochromator Transmission Mode (Rainbow)

Beamsize (FWHM) **<0.5x0.5** µm²



Advantages:

- Monochromator**s** working in parallel => even better statistics

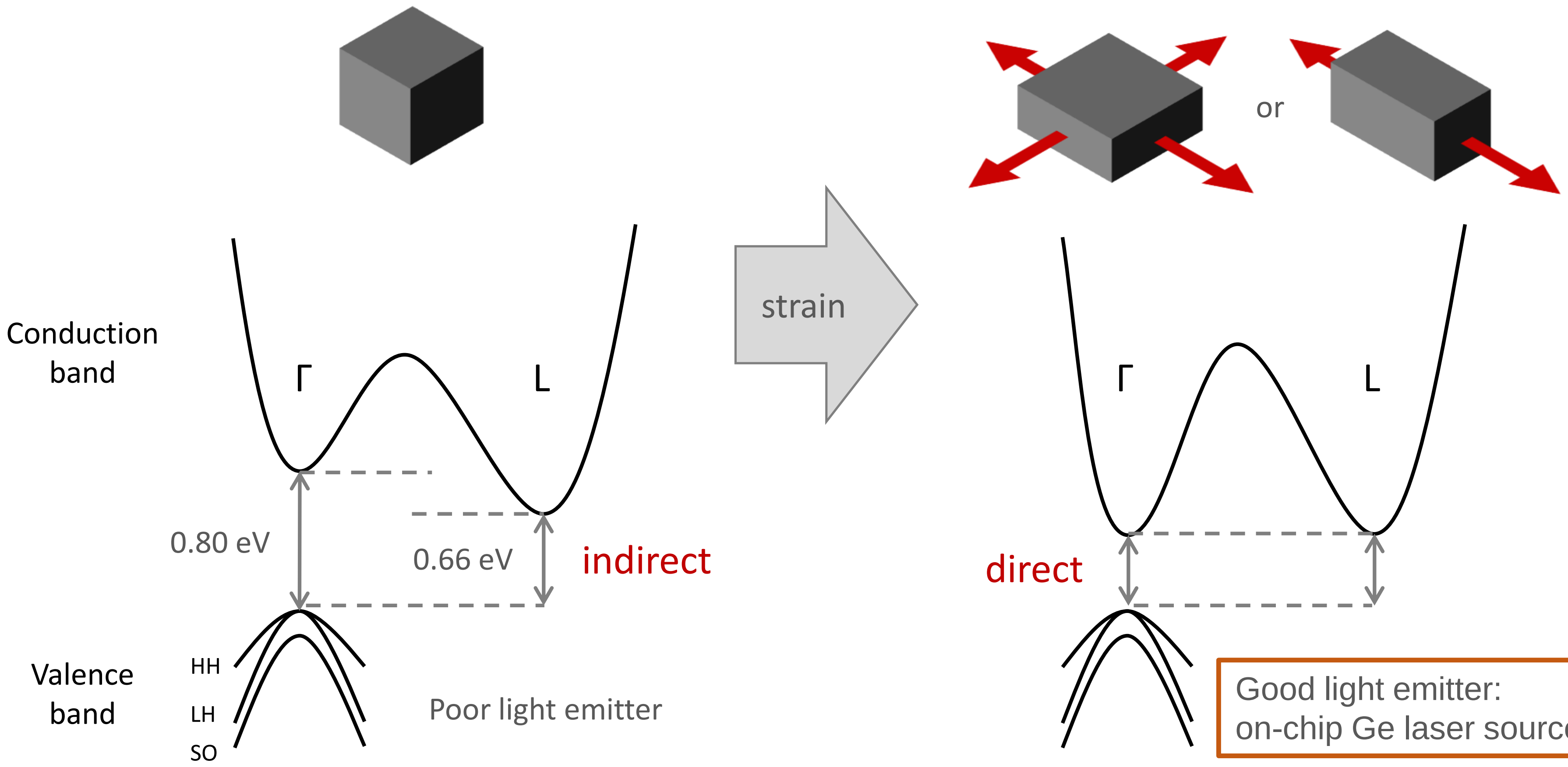
Fast & reliable full strain tensor ε

Appendix: Fine structural metrology for applied physics

Works of Samuel Tardif, CEA , CRG-IF BM32 beamline

Why straining Ge ?

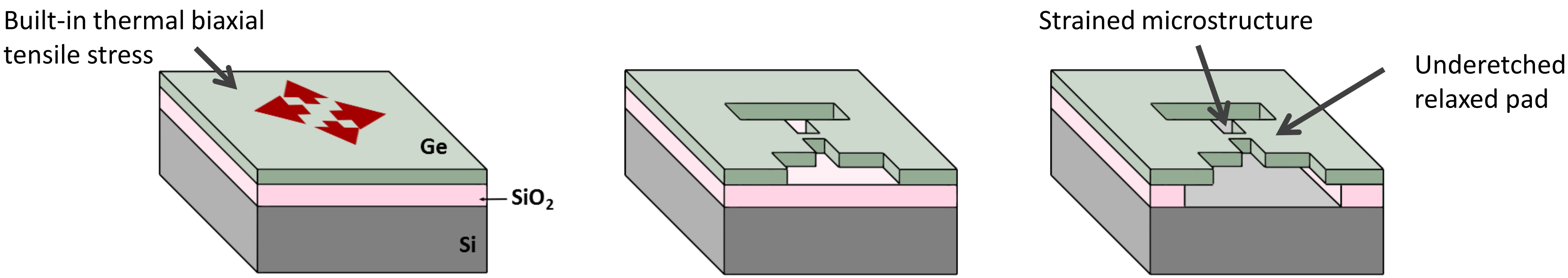
Strain engineering: from indirect to direct bandgap

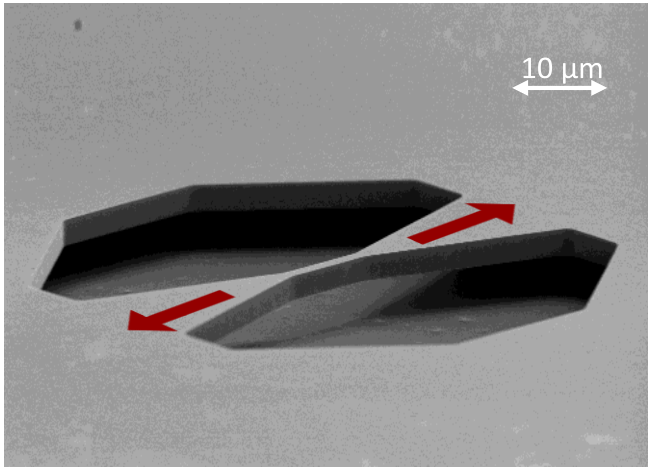


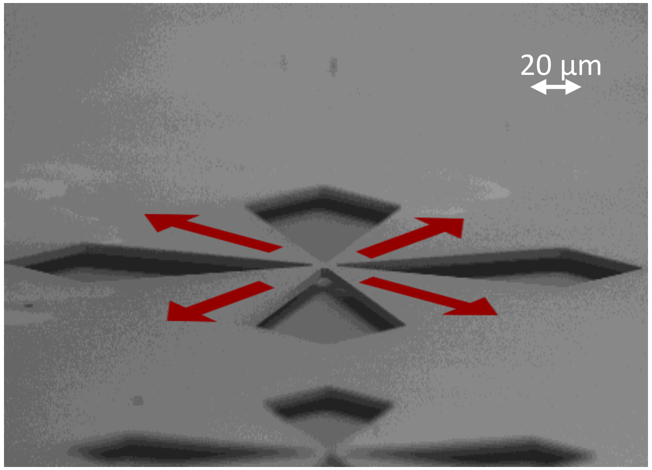
Good light emitter:
on-chip Ge laser source!

How to strain Ge ?

Strain concentration in GeOI membranes [Alban Gassenq]





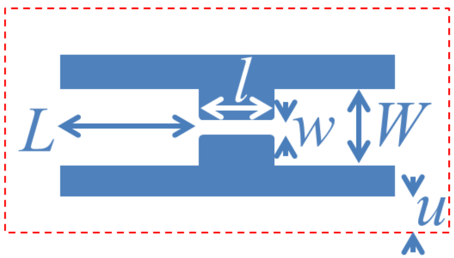


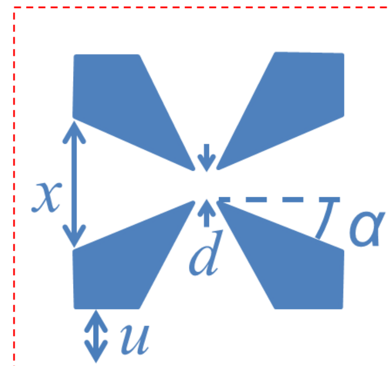
Vincent Reboud *et al., Proc. SPIE* 9367, Silicon Photonics X, 936714, 2015

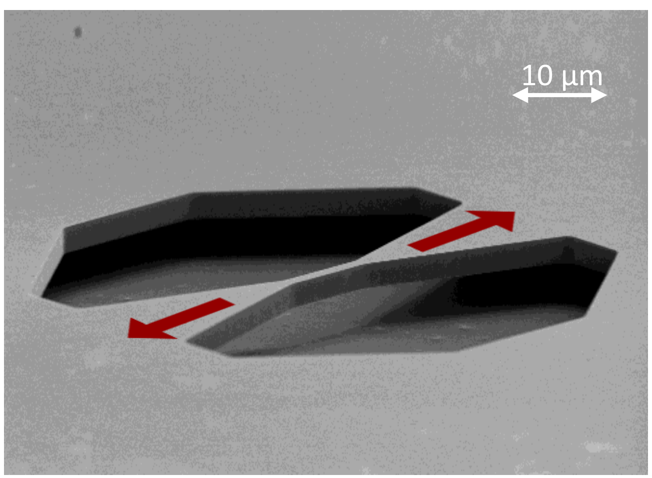
How to strain Ge ?

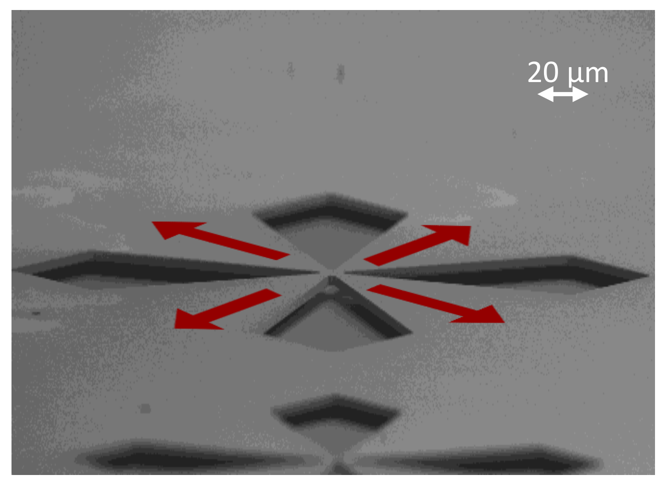
Model objects to study any arbitrary uniaxial or biaxial stress

Strain can be statically tuned by changing the design parameters (width, length, …)



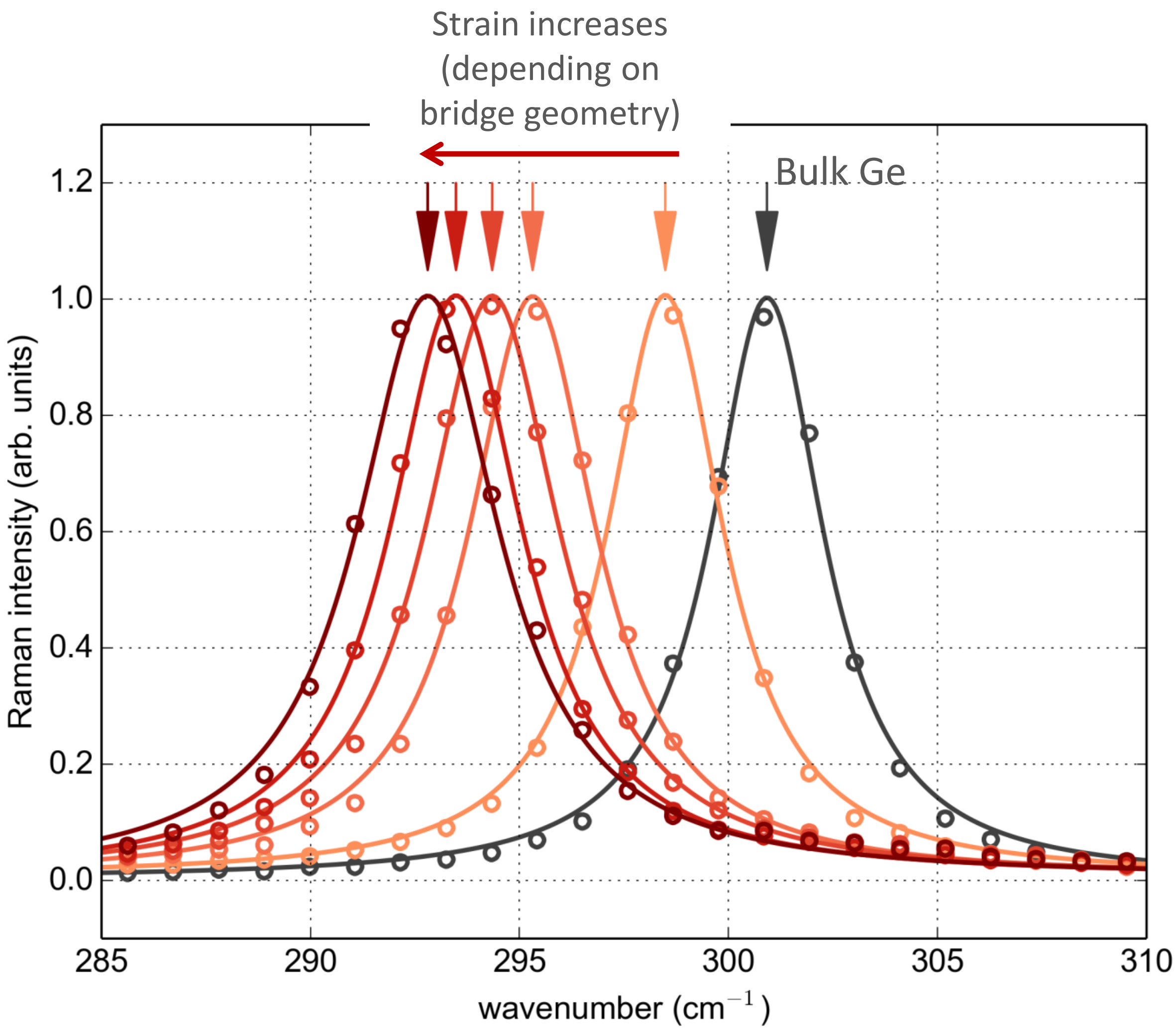


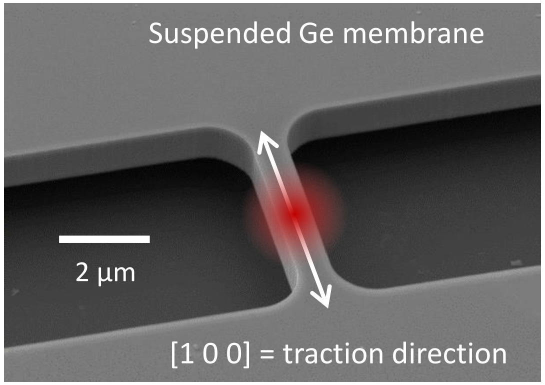




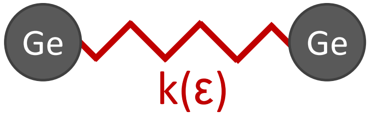
Vincent Reboud *et al., Proc. SPIE* 9367, Silicon Photonics X, 936714, 2015

Measuring the strain at the µm scale : Raman spectroscopy





Probe the Ge-Ge resonance



shift to **strain** conversion:

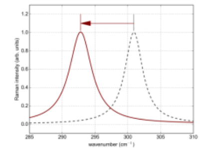
0.649 % per cm^{-1} (uniaxial)

0.236 % per cm^{-1} (biaxial)

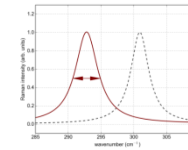
Raman spectroscopy measurements with Guilherme Osvaldo Dias, SEM image by Alban Gassenq

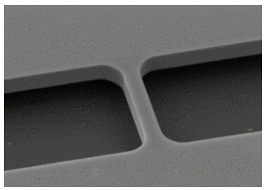
Raman spectroscopy mapping

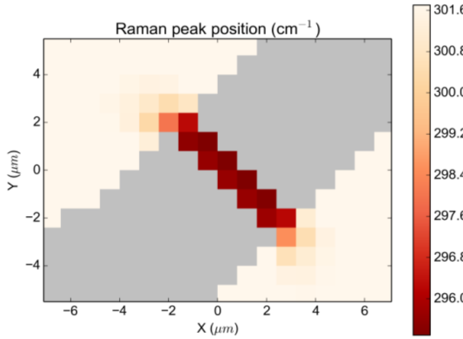
local strain **amplitude**

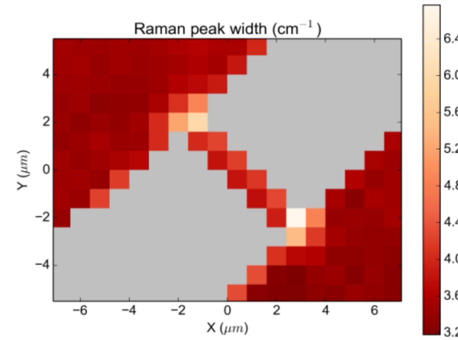


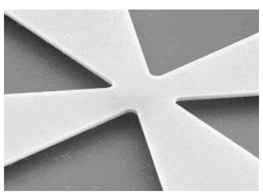
local strain **distribution**

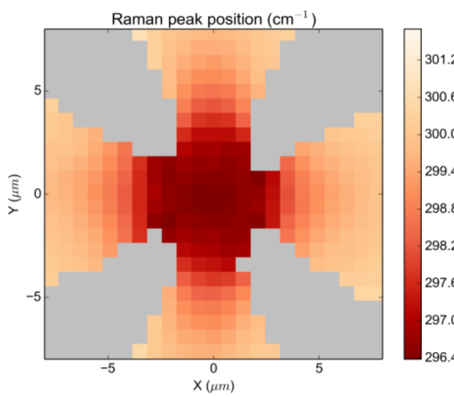


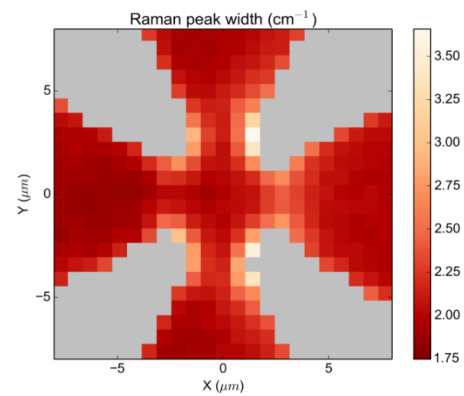




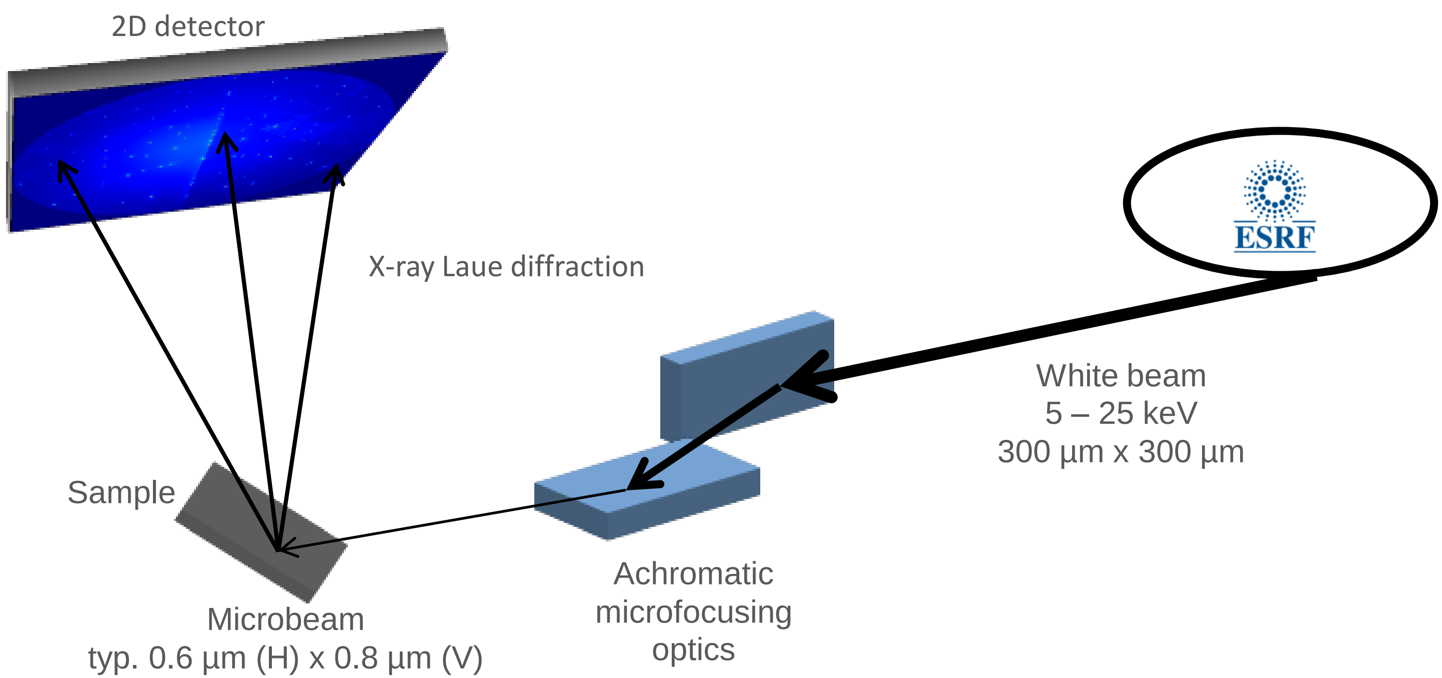






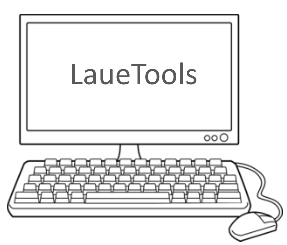


Good check of the homogeneity of the strain but analysis is difficult when the strain is not pure bi- or uniaxial



Diffraction peaks

2θ	χ	Intensity
…	…	…
…	…	…
…	…	…



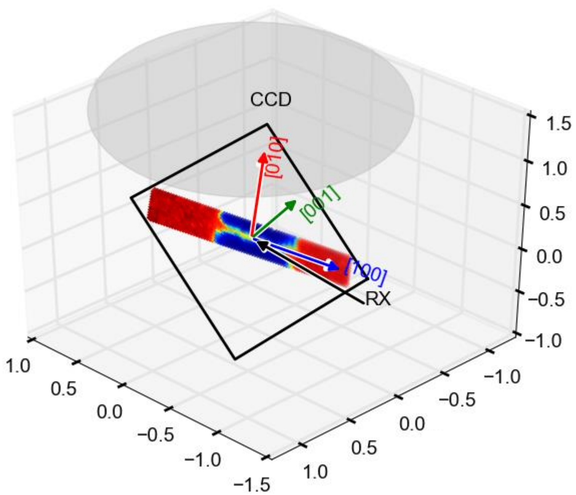
Analysis software (developed at BM32 by J.S. Micha, O. Robach)

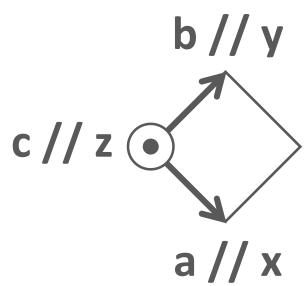
Orientation + Deviatoric strain tensor

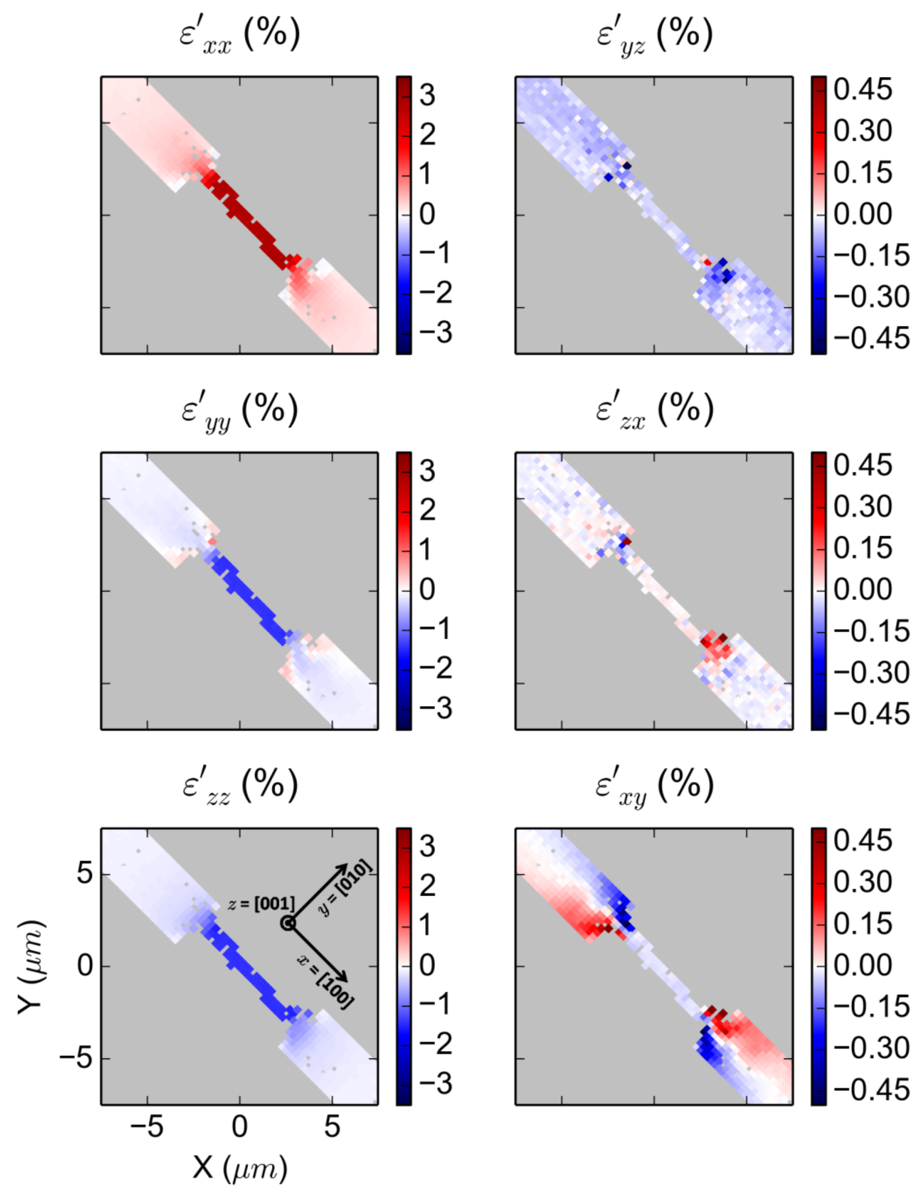
(= sensitive to **symmetry changes** but insensitive to **homothetic transformations**)

$$\begin{pmatrix} Full \\ Strain \end{pmatrix} = \begin{pmatrix} Hydrostatic \\ Strain \end{pmatrix} + \begin{pmatrix} Deviatoric \\ Strain \end{pmatrix}$$

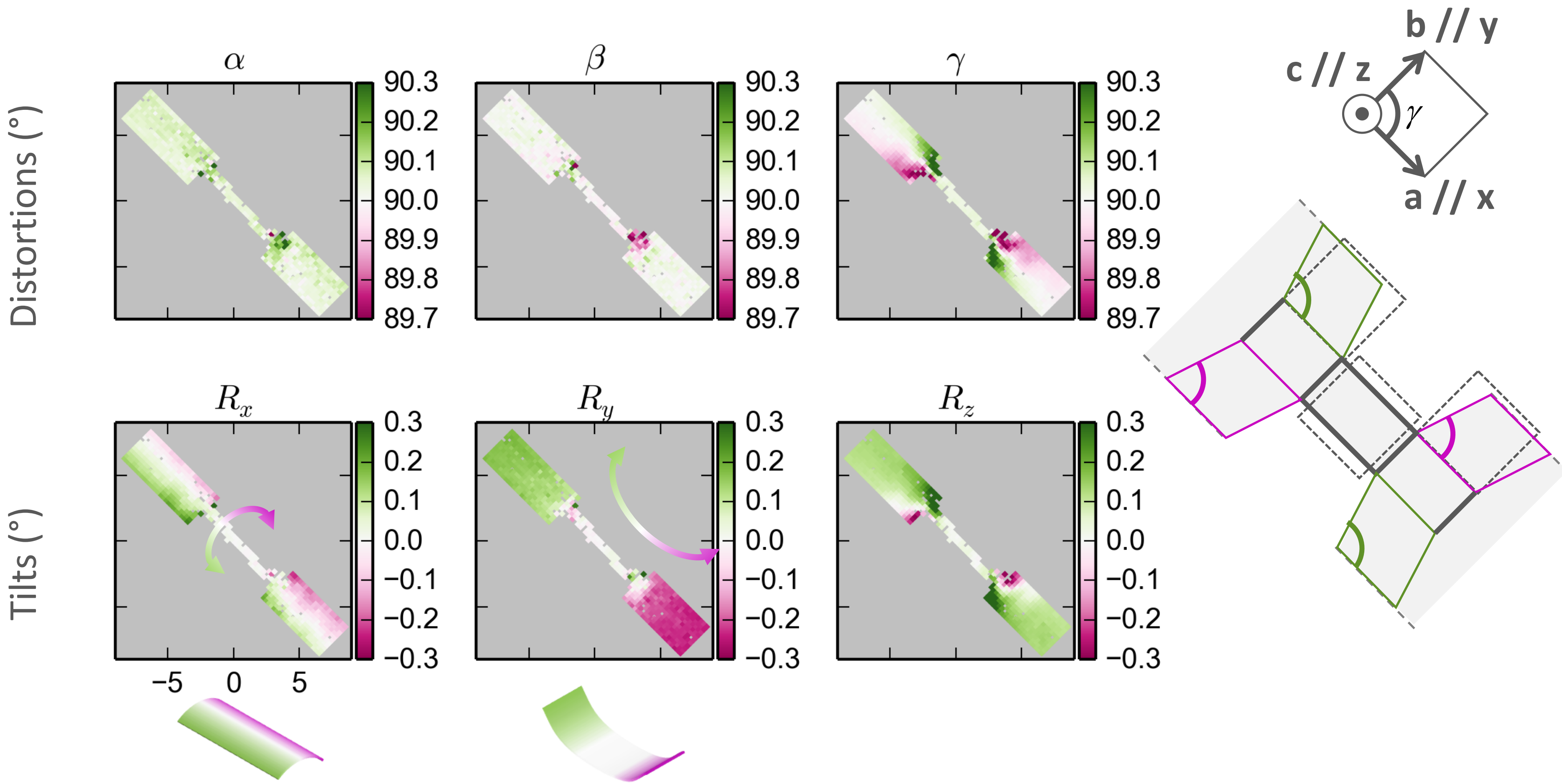
Direct measurement of the deviatoric strain tensor





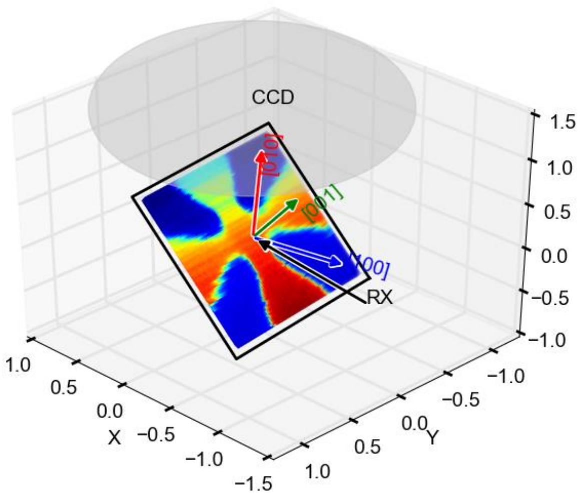


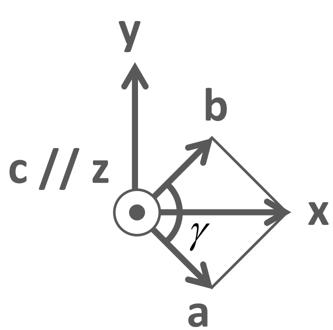
Distortions and rotations of the lattice (uniaxial stress)

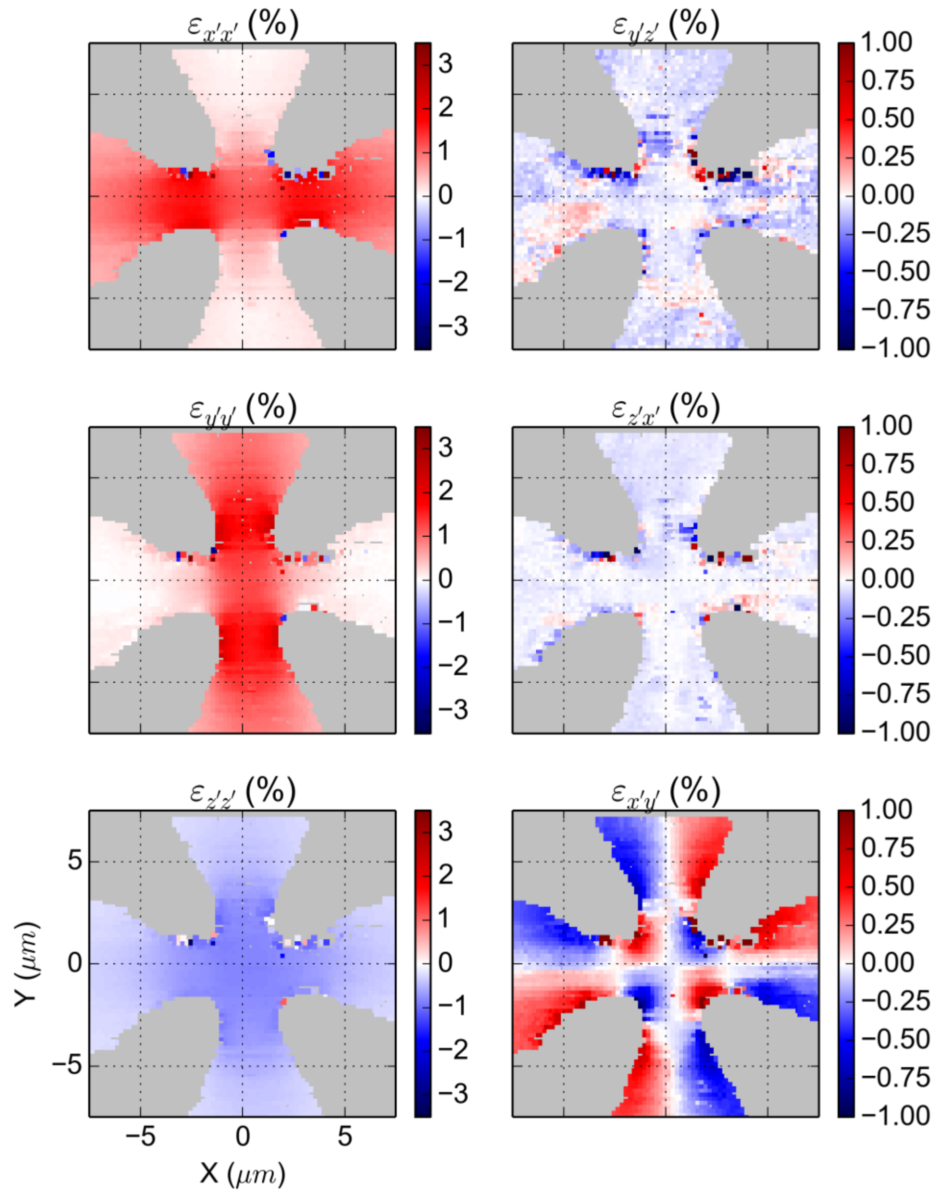


Small anticlastic relaxation of the stretching arms and strong distortion + rotation near the fillets (max shear stress)

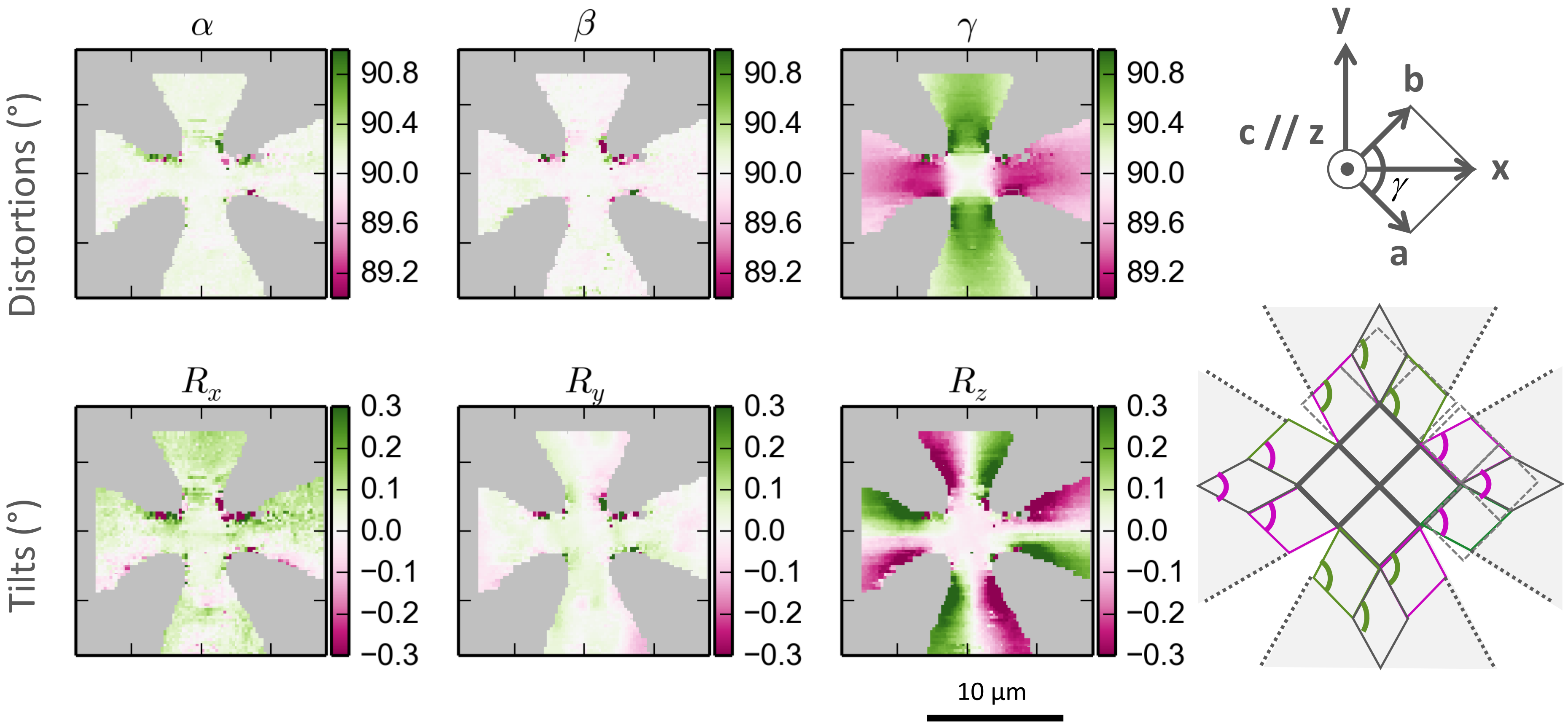
Direct measurement of the deviatoric strain tensor







Distortions and rotations of the lattice (biaxial stress)



Out-of-plane rotations are negligible (no anticlastic or synclastic relaxations) Strong distortions and rotations near the fillets

From the deviatoric to the full strain tensor

Without measuring Laue spots energy

Hypothesis : No stress perpendicular to the free surfaces ($\sigma_{33} = 0$)

Hooke's law **F** = k**x** … *with tensors and Voigt's notation:*

$$\begin{pmatrix} \sigma_{11} \\ \sigma_{22} \\ \sigma_{33}=0 \\ \sigma_{23} \\ \sigma_{13} \\ \sigma_{12} \end{pmatrix} = \begin{pmatrix} c_{11} & c_{12} & c_{12} & 0 & 0 & 0 \\ c_{12} & c_{11} & c_{12} & 0 & 0 & 0 \\ c_{12} & c_{12} & c_{11} & 0 & 0 & 0 \\ 0 & 0 & 0 & c_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & c_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & c_{44} \end{pmatrix} \begin{pmatrix} \varepsilon'_{11} + \varepsilon_h/3 \\ \varepsilon'_{22} + \varepsilon_h/3 \\ \varepsilon'_{33} + \varepsilon_h/3 \\ 2\varepsilon'_{23} \\ 2\varepsilon'_{13} \\ 2\varepsilon'_{12} \end{pmatrix}$$

$1,2,3 = x, y, z = [100], [010], [001]$

σ = stress (unknown)

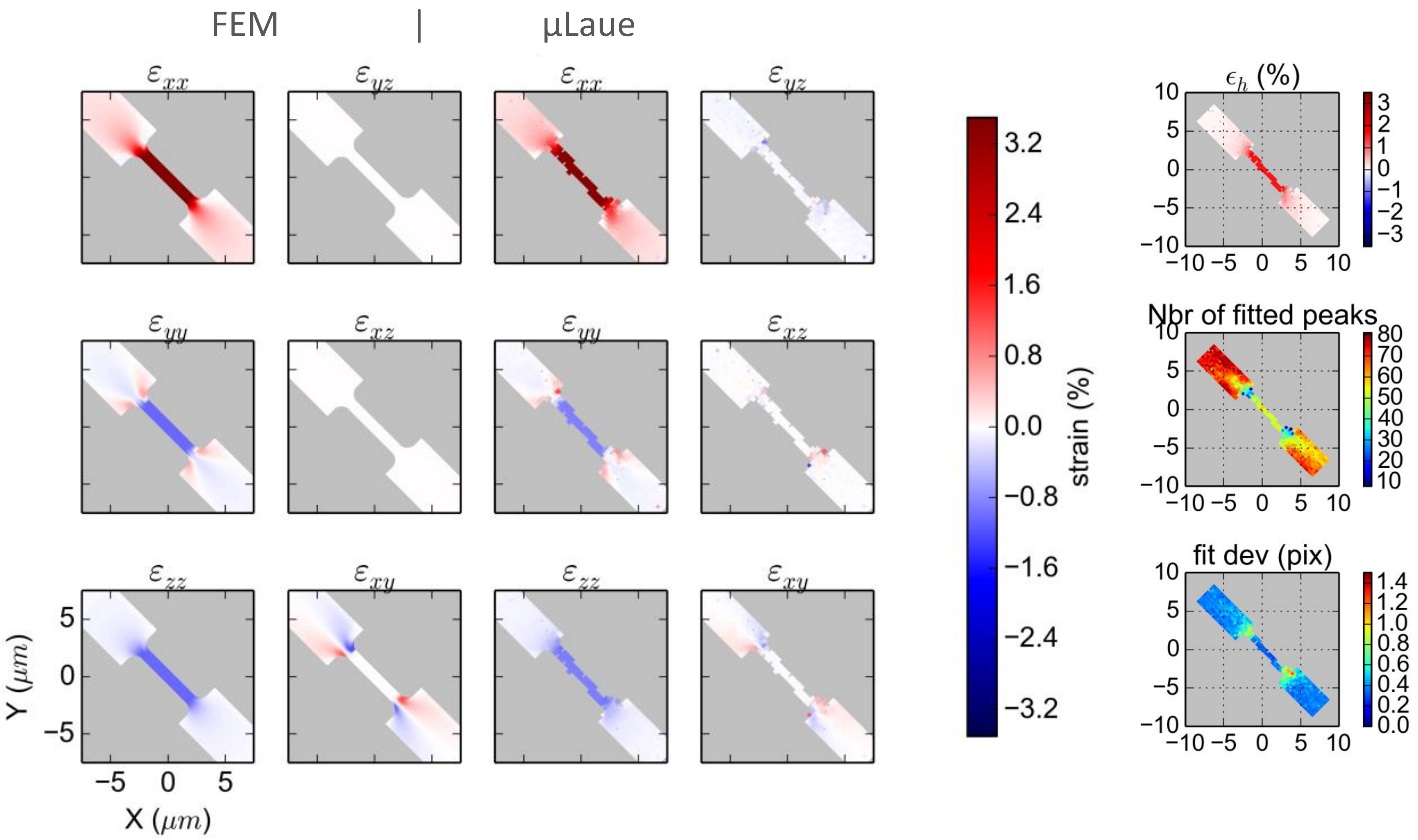
c_{11}, c_{12}, c_{44} = 126, 44.0, 67.7 GPa = elastic coefficients of Ge

ε' = deviatoric strain (measured)

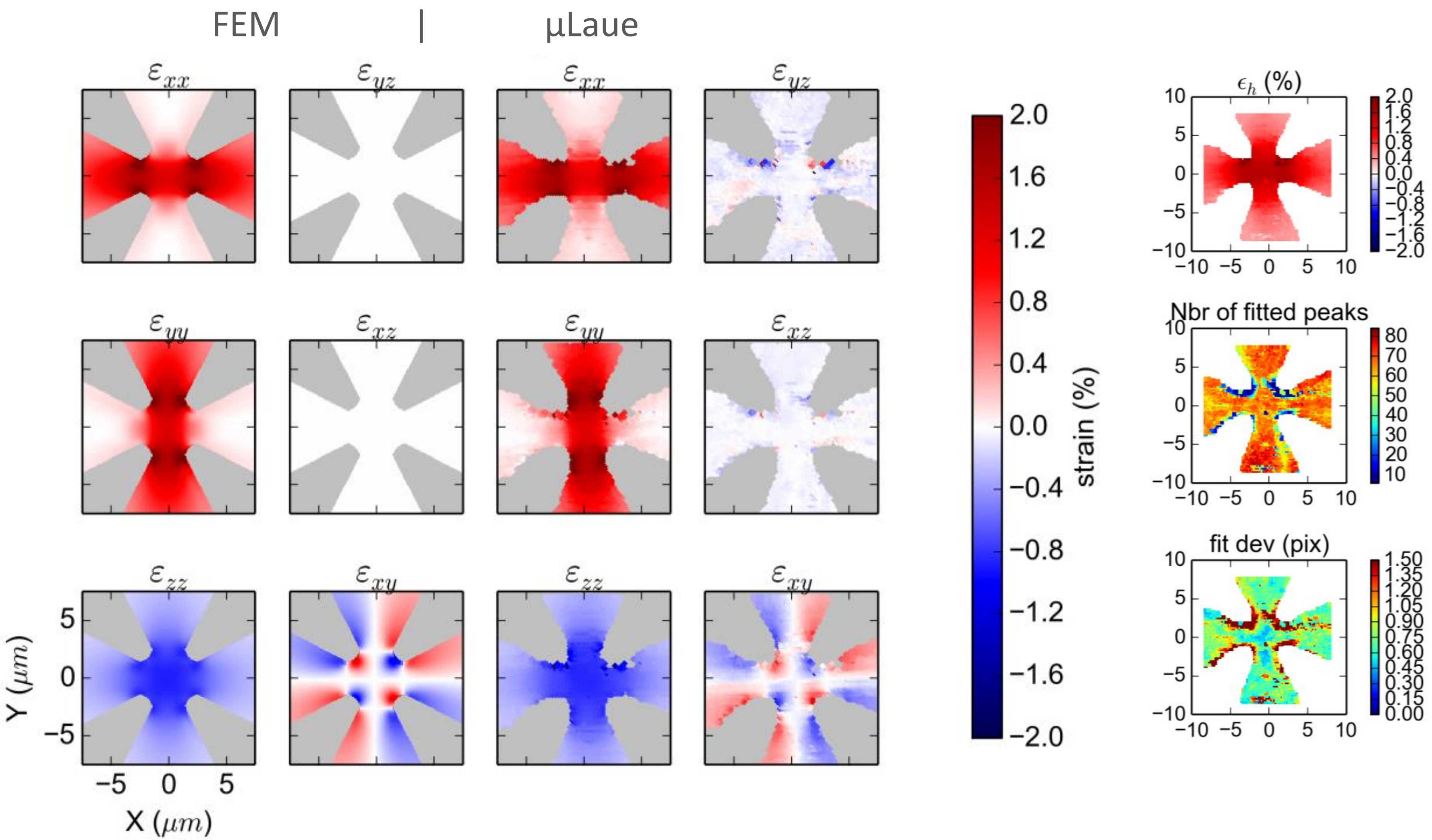
ε_h = hydrostatic strain (unknown)

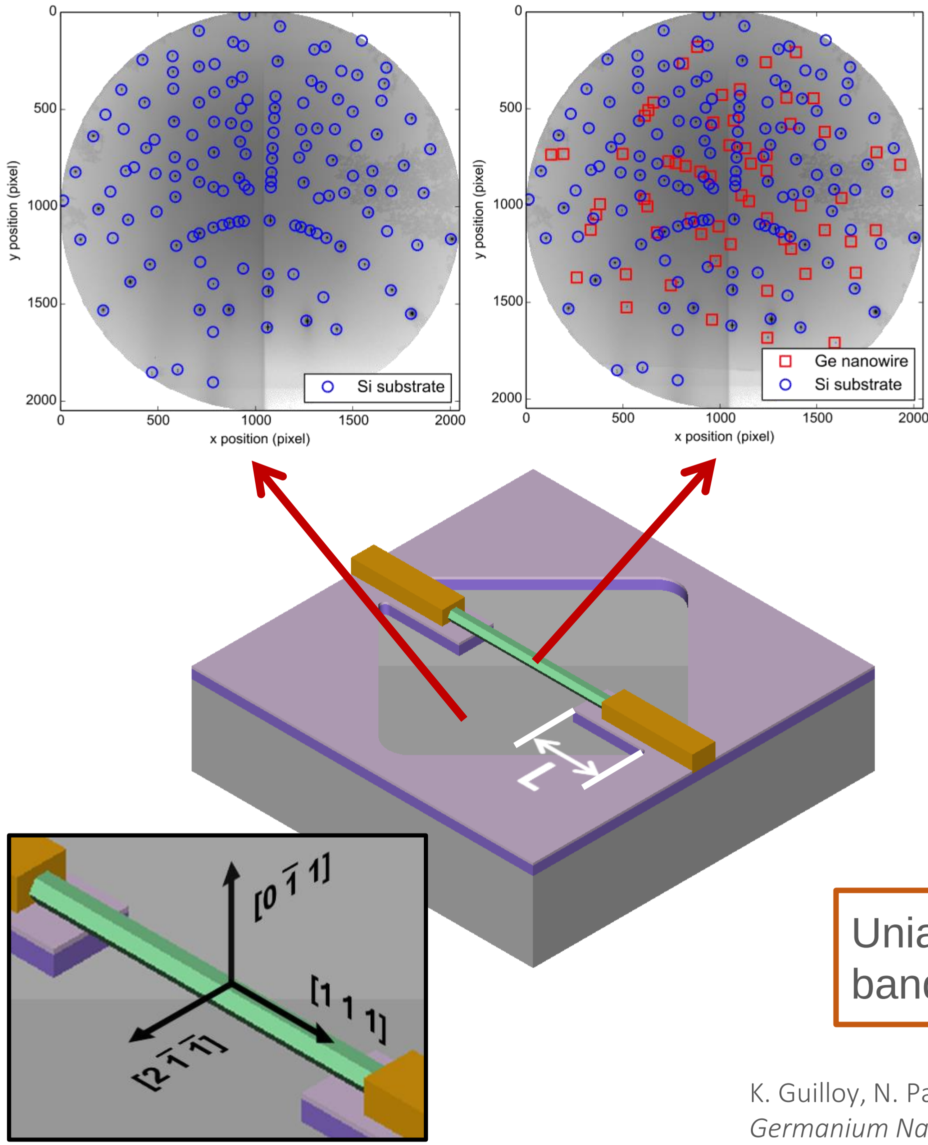
Solve the 6 equations for the 6 unknowns to get the full strain tensor

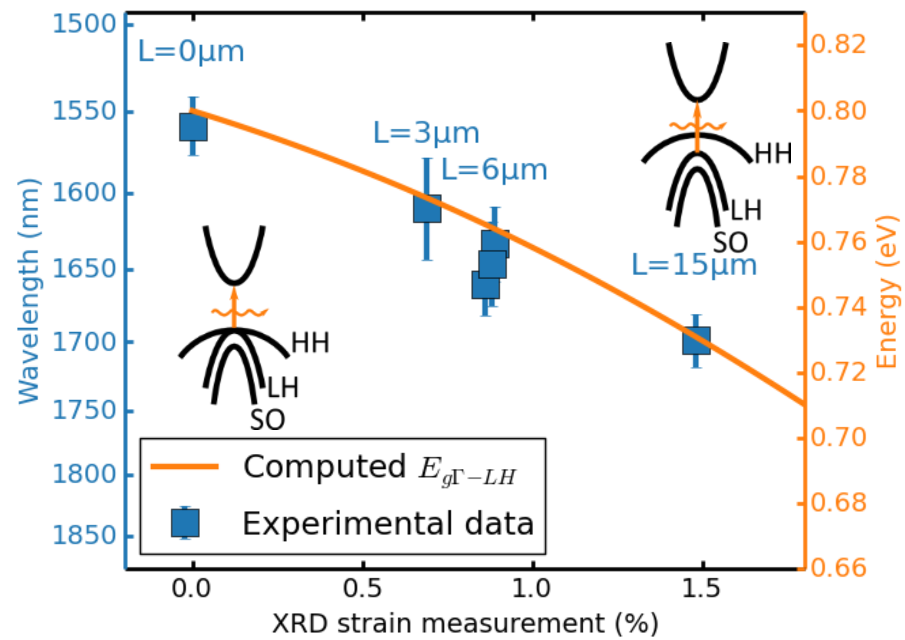
XRD measurements *vs* FEM simulations



XRD measurements *vs* FEM simulations







Uniaxial [111] strain in the range 0 % – 1.5 %, bandgap energies consistent with theoretical model

K. Guilloy, N. Pauc, A. Gassenq, P. Gentile, S. Tardif, F. Rieutord, V. Calvo, *Tensile Strained Germanium Nanowires Measured by Photocurrent Spectroscopy and X-ray Microdiffraction*, Nano Lett., **2015**, 15 (4), pp 2429–2433

Describing how the lattice is strained (deformed) under stress (~force)

General case

$$\begin{pmatrix} \varepsilon_{xx} & \varepsilon_{xy} & \varepsilon_{xz} \\ \varepsilon_{xy} & \varepsilon_{yy} & \varepsilon_{yz} \\ \varepsilon_{xz} & \varepsilon_{yz} & \varepsilon_{zz} \end{pmatrix}$$

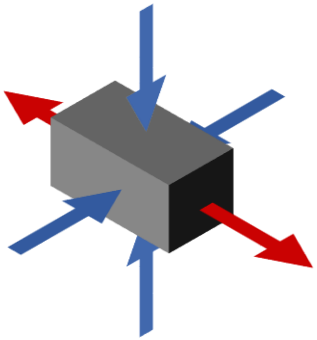
$$\varepsilon_{ij} = \frac{\Delta i}{j} = \frac{\Delta j}{i}$$

$\varepsilon_{xx}, \varepsilon_{yy}, \varepsilon_{zz}$ = axial strain

$\varepsilon_{xy}, \varepsilon_{xz}, \varepsilon_{yz}$ = shear strain

Pure uniaxial tensile stress

$$\begin{pmatrix} \varepsilon_{xx} & 0 & 0 \\ 0 & -\nu\varepsilon_{xx} & 0 \\ 0 & 0 & -\nu\varepsilon_{xx} \end{pmatrix}$$



Pure biaxial tensile stress

$$\begin{pmatrix} \varepsilon_{xx} & 0 & 0 \\ 0 & \varepsilon_{xx} & 0 \\ 0 & 0 & -2\nu\varepsilon_{xx} \end{pmatrix}$$