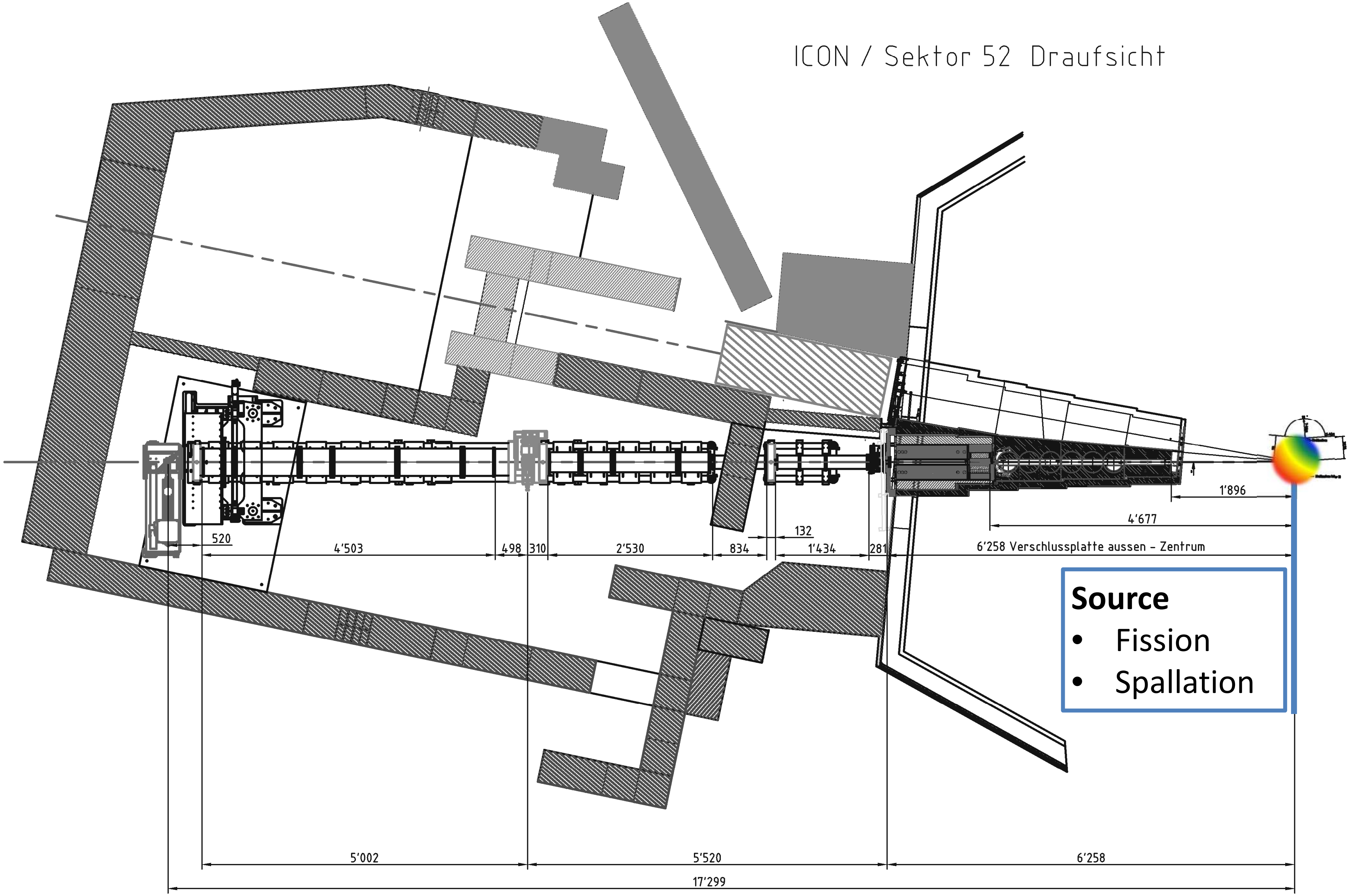
Wir schaffen Wissen – heute für morgen

Steven Peetermans

Neutron Imaging & Activation Group, Paul Scherrer Institut, Switzerland
now at *AV Controlatom, Belgium*

Energy-selective neutron imaging


ICON / Sektor 52 Draufsicht
1'896
4'677
520
4'503
498
310
2'530
834
132
1'434
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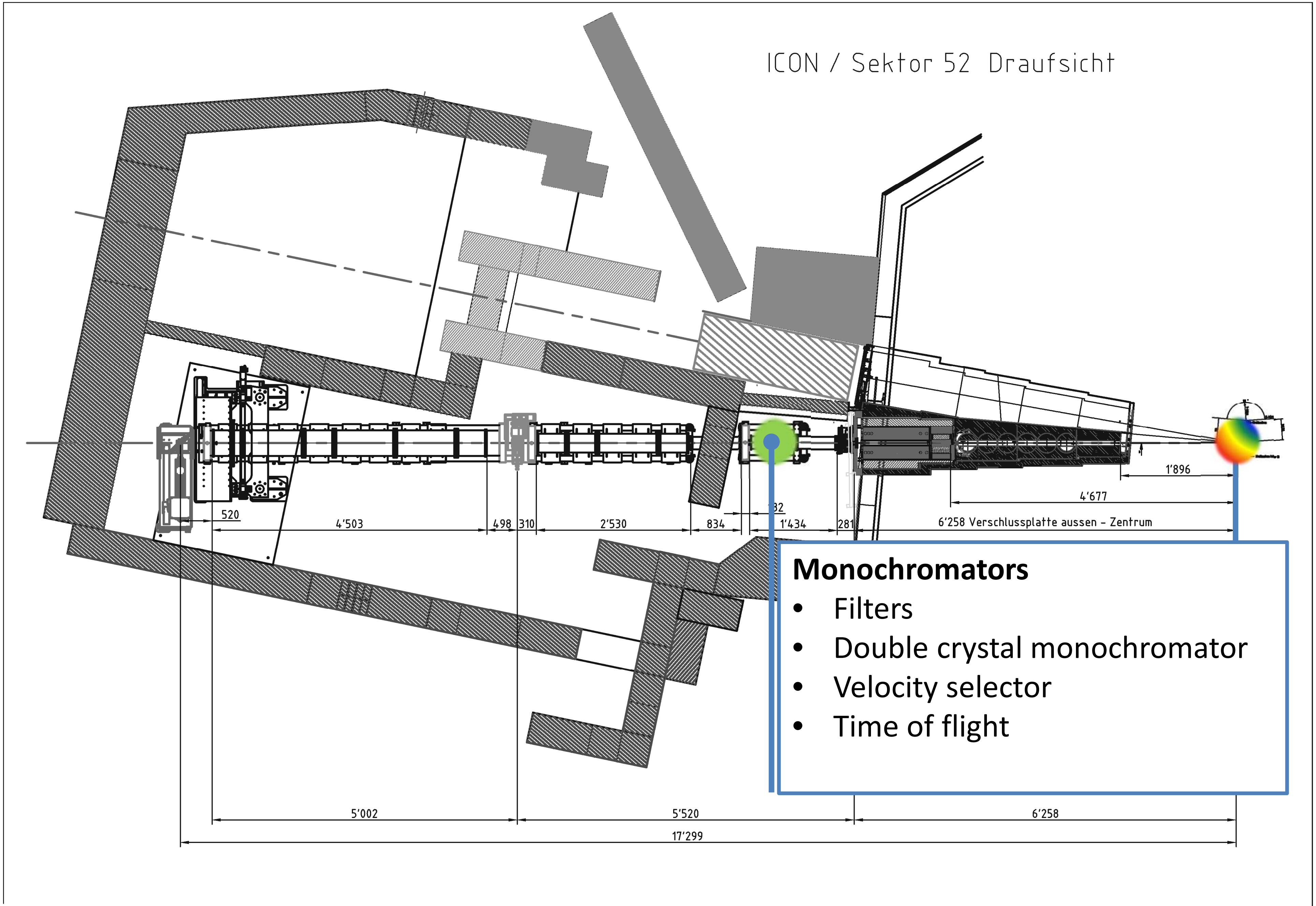
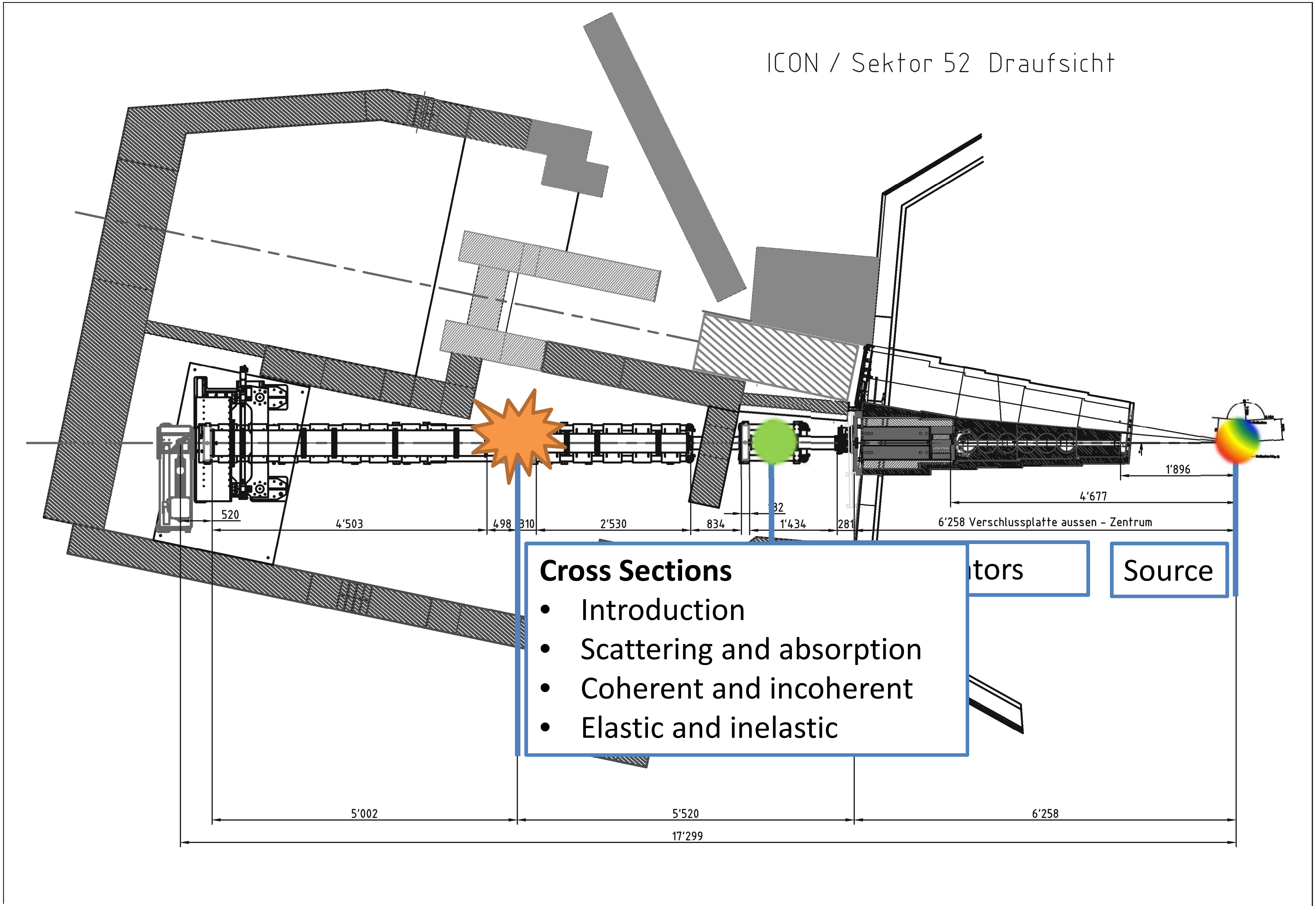
6'258 Verschlussplatte aussen – Zentrum
5'002
5'520
6'258
17'299
Source
• Fission
• Spallation

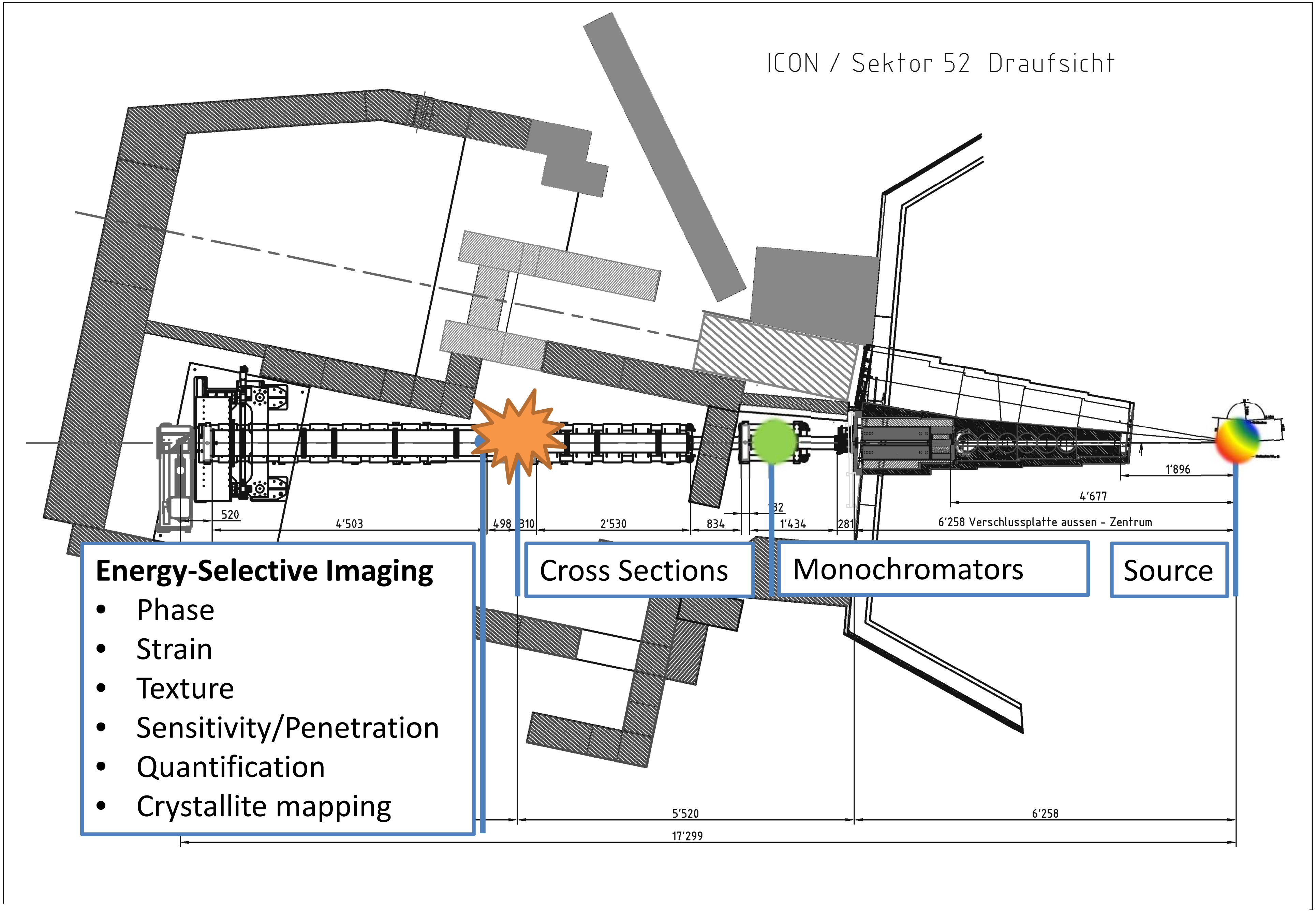


Monochromators
- Filters
- Double crystal monochromator
- Velocity selector
- Time of flight



Cross Sections

- Introduction
- Scattering and absorption
- Coherent and incoherent
- Elastic and inelastic


ICON / Sektor 52 Draufsicht
Energy-Selective Imaging
• Phase
• Strain
• Texture
• Sensitivity/Penetration
• Quantification
• Crystallite mapping
Cross Sections
Monochromators
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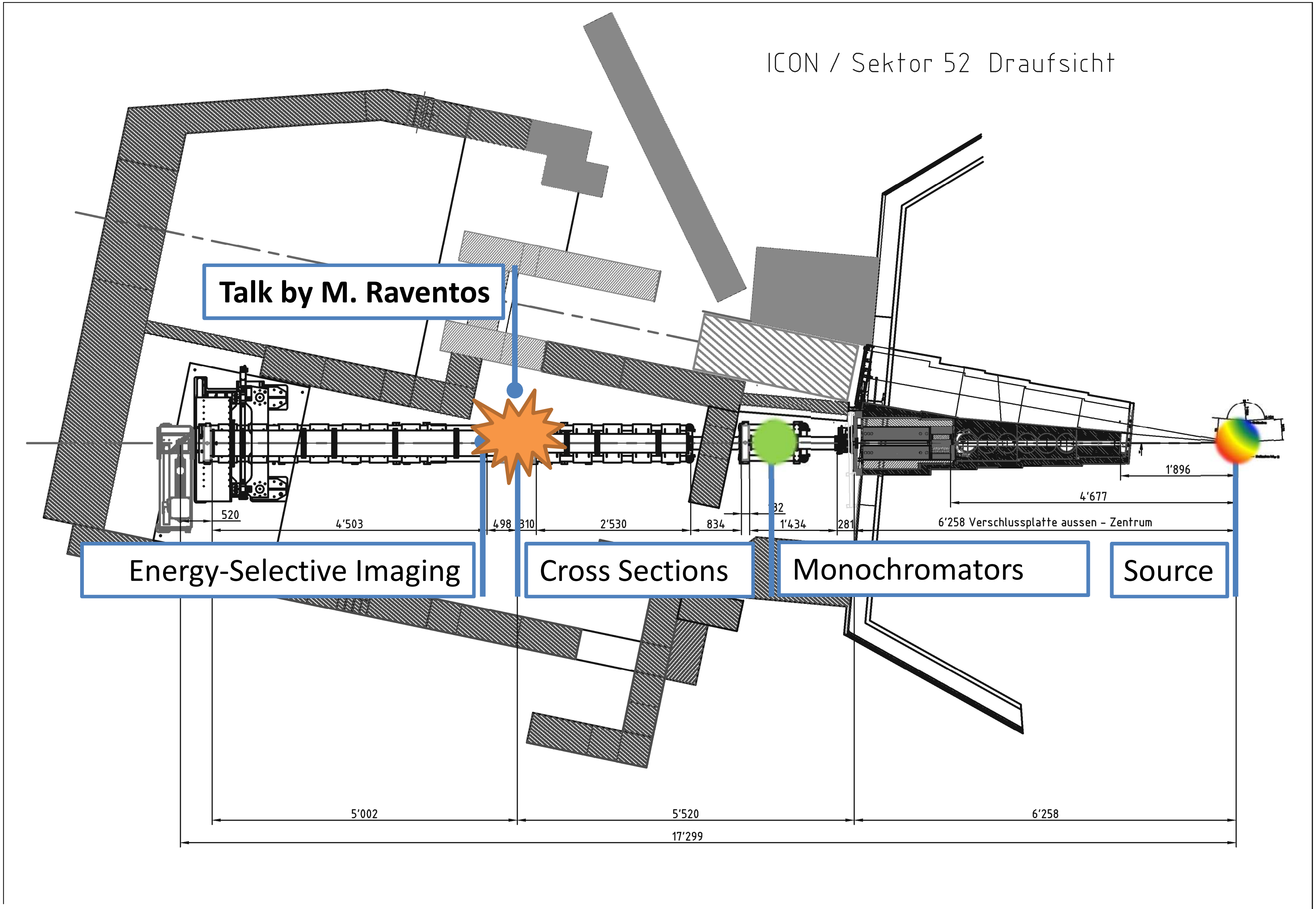
ICON / Sektor 52 Draufsicht
Talk by M. Raventos
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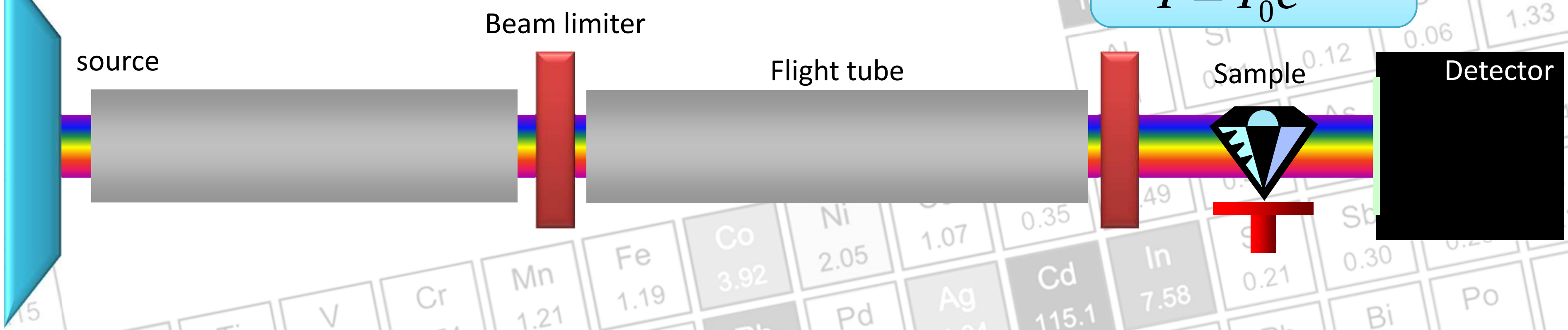
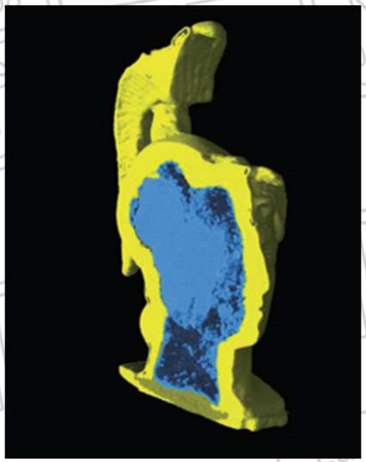
Traditional transmission imaging

Lambert-Beer law

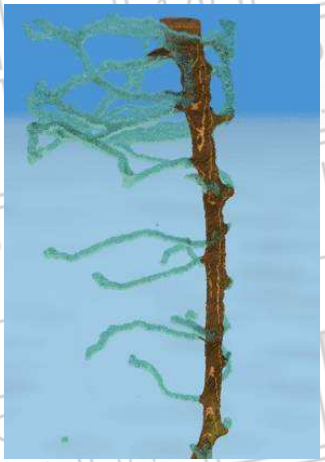
$$I = I_0 e^{-\Sigma x}$$



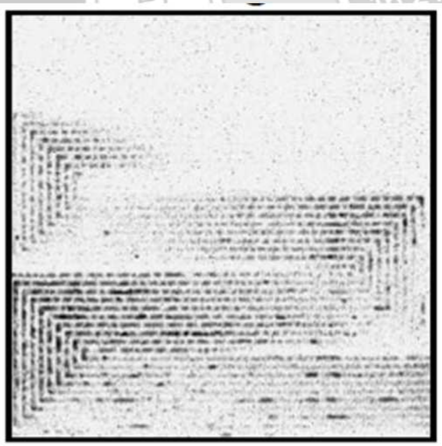
Spatial discrimination between materials of different cross-section



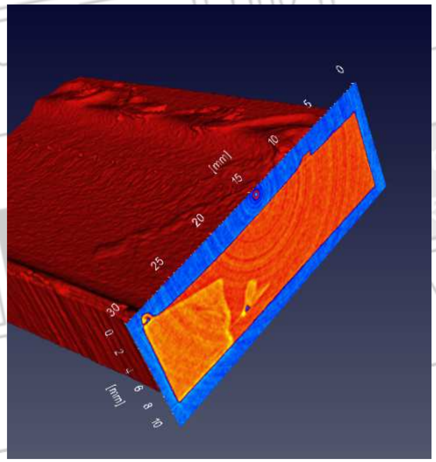
Archaeology



Biology



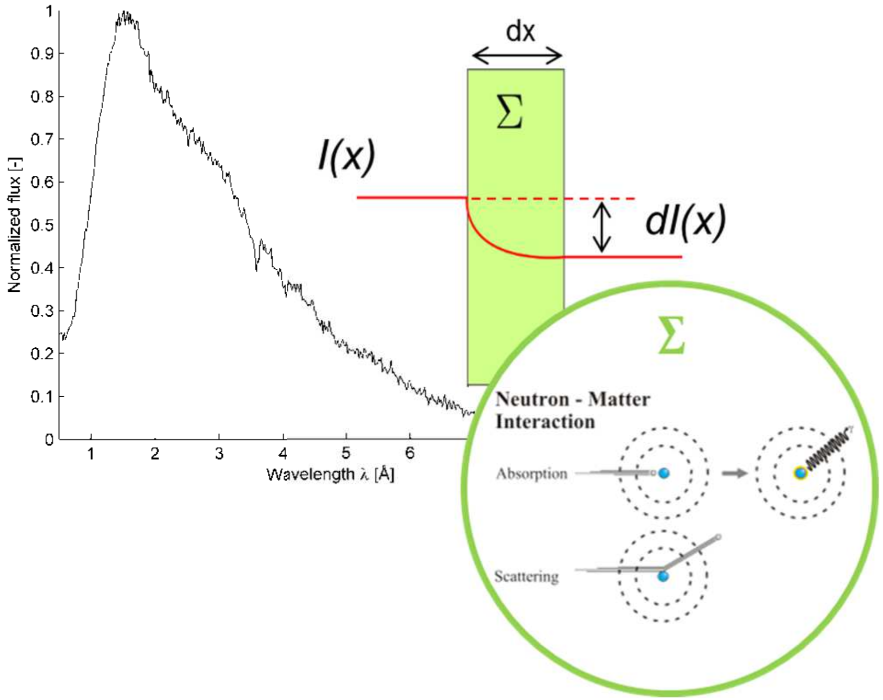
Fuel cells



Materials science

Reality is more complex

But Σ is more than just a number…



$$\int_{\lambda} I(\lambda)d\lambda = \int_{\lambda} I_0(\lambda)e^{-\Sigma(\lambda)x}d\lambda$$

Interaction (Σ) depends on the neutron energy

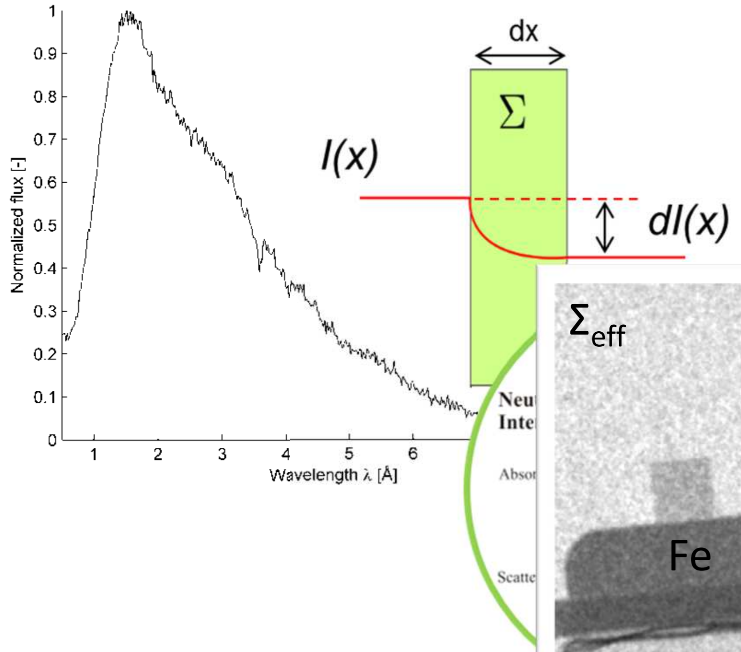
Incident beam spectrum $I_0(\lambda)$

Traditionally imaging: spectral averaged Σ_{eff}

Information is lost

Traditional neutron imaging

But Σ is more than just a number…



$$\int_{\lambda} I(\lambda)d\lambda = \int_{\lambda} I_0(\lambda)e^{-\Sigma(\lambda)x}d\lambda$$

Interaction (Σ) depends on the neutron energy

pectrum $I_0(\lambda)$

aging: spectral averaged Σ_{eff}

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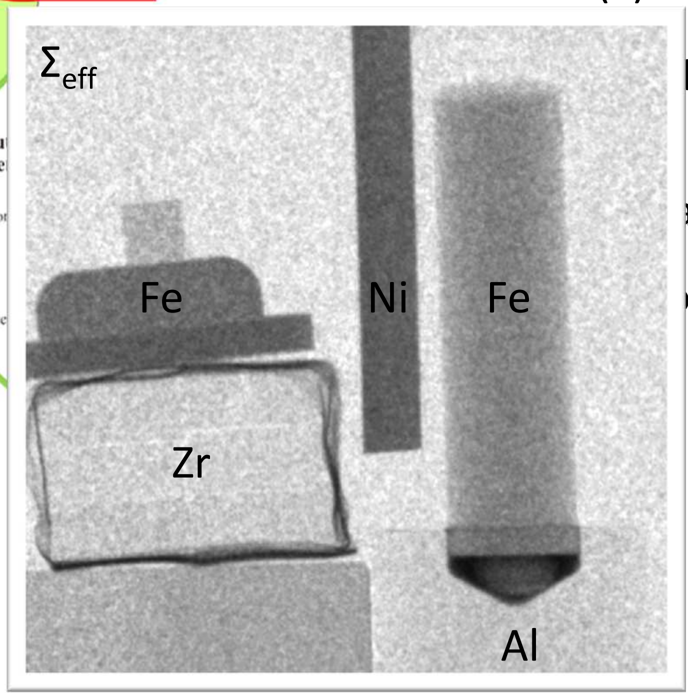
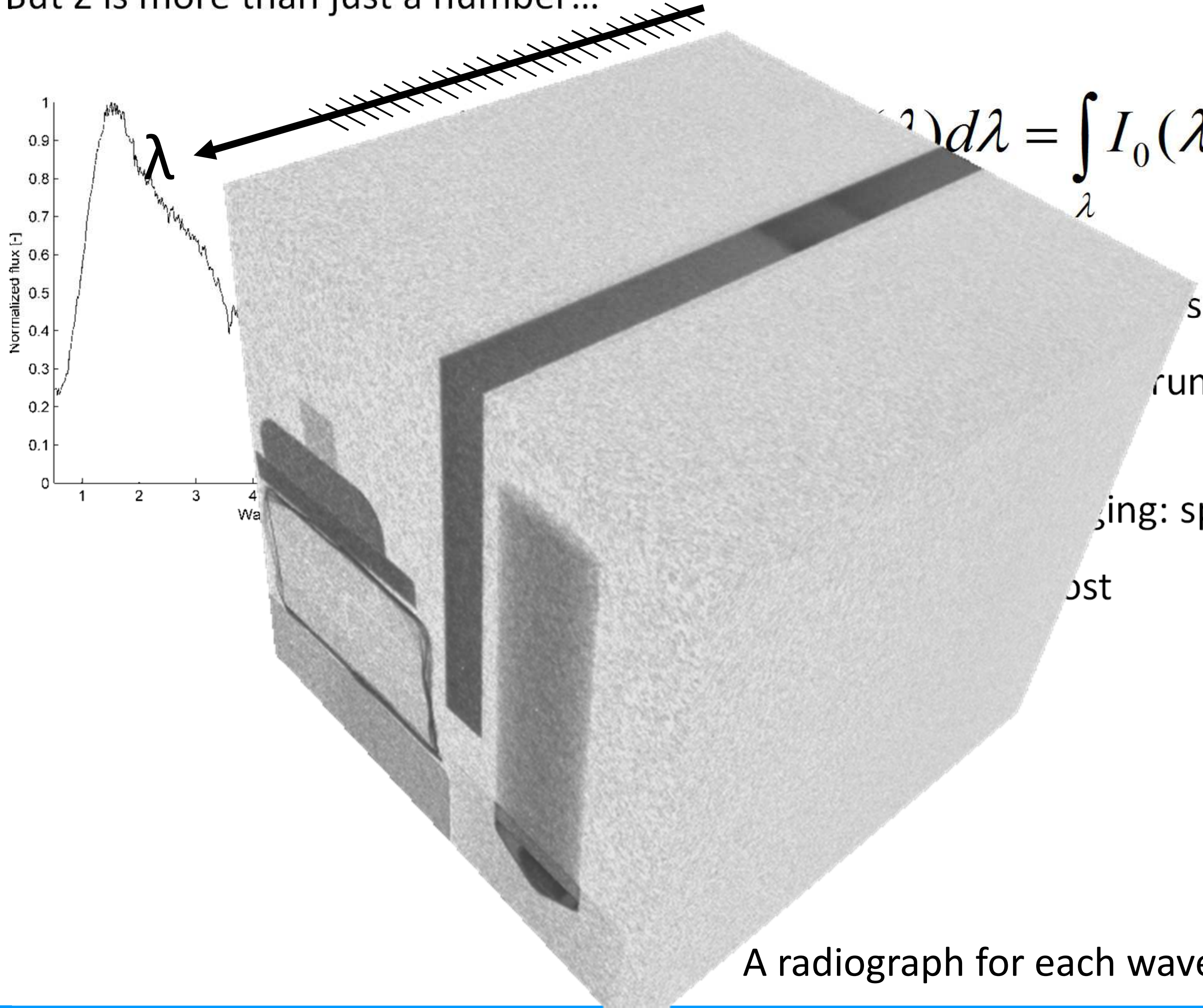


Energy-selective neutron imaging

But Σ is more than just a number…



$$\ldots)d\lambda = \int_\lambda I_0(\lambda) e^{-\Sigma(\lambda)x} d\lambda$$

…s on the neutron energy

…rum $I_0(\lambda)$

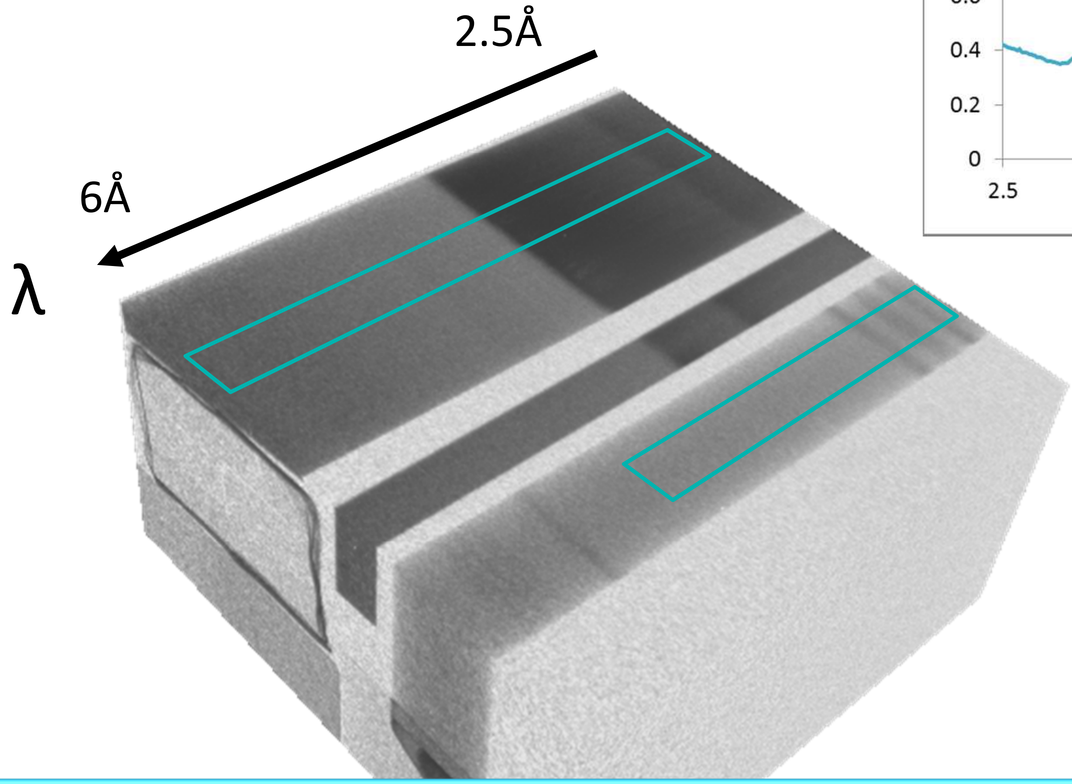
…ing: spectral averaged Σ_{eff}

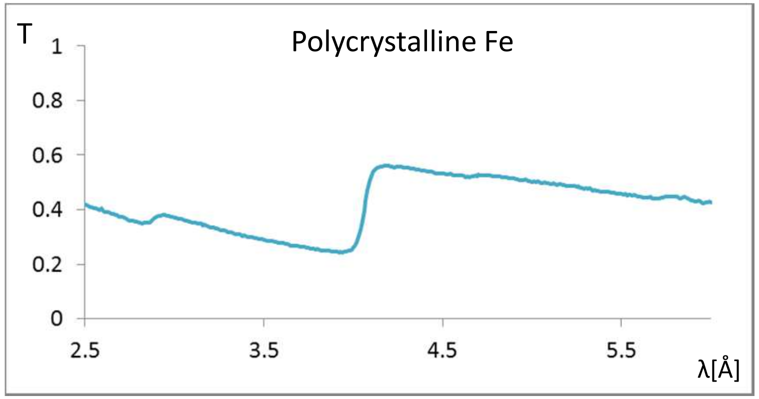
…ost

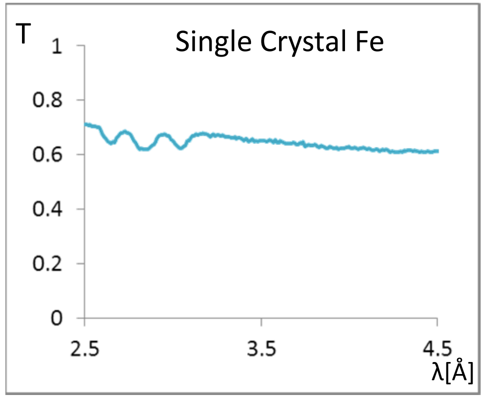
A radiograph for each wavelength

A closer look…

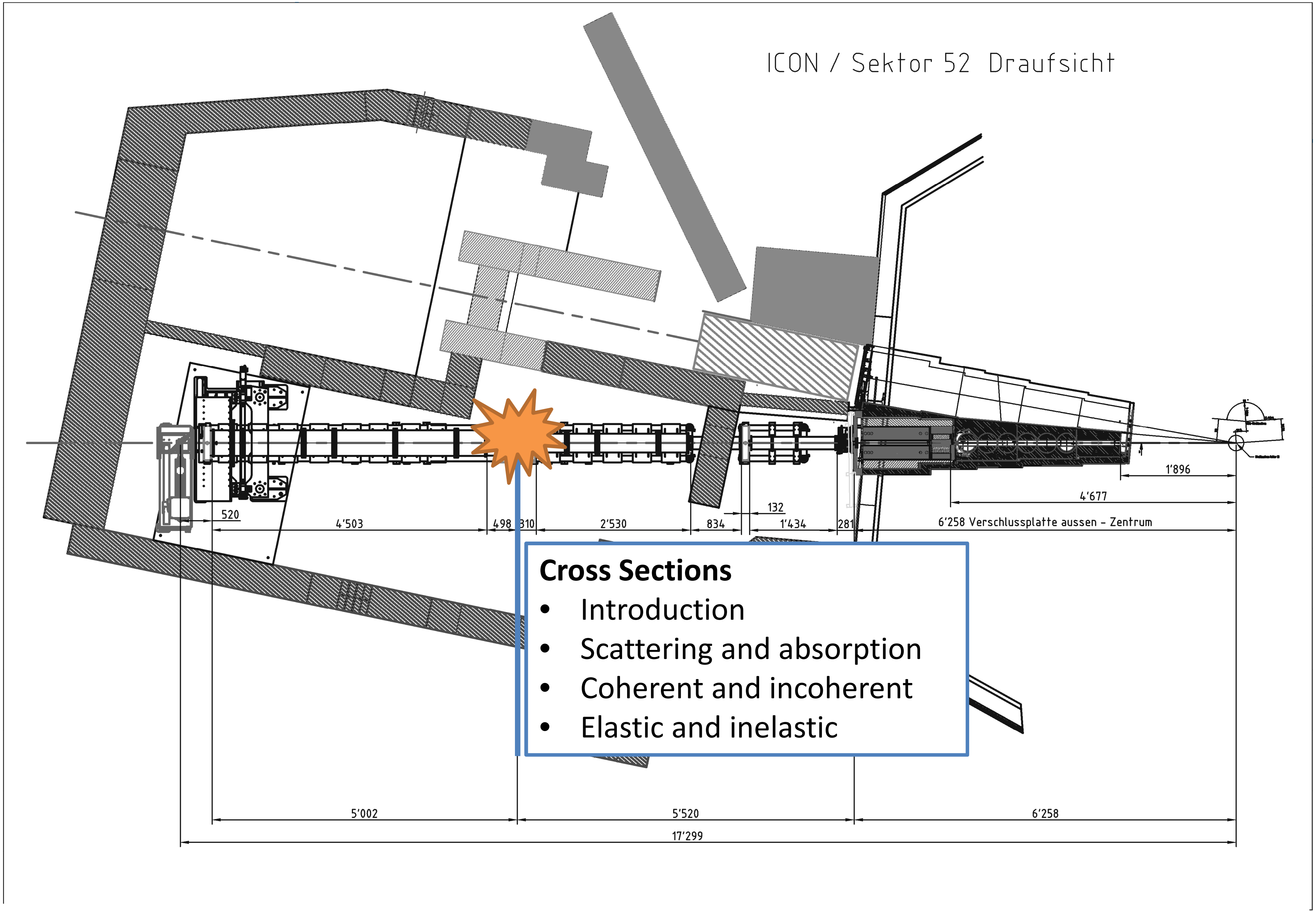
A closer look at the iron samples







This lecture: investigate the Σ(λ) to probe new sample properties



Cross Sections

- Introduction
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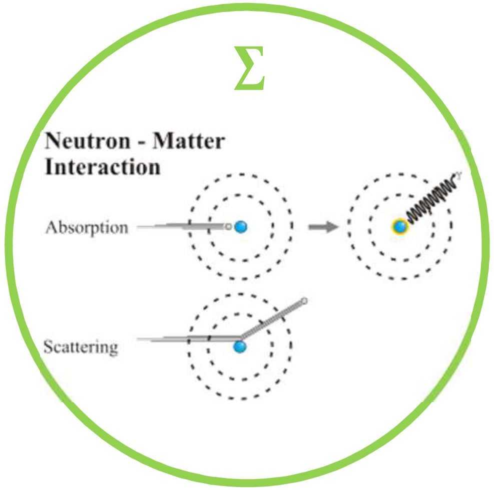
Cross section

Cross section = interaction probability

Microscopic cross section σ:

$$\sigma = \frac{\text{\# neutrons undergoing interaction per target nucleus}}{\text{incident neutron flux}}$$



Macroscopic cross section ∑:

$$\Sigma = N\sigma$$

$$\Sigma = \frac{\rho N_A}{M}\sigma$$

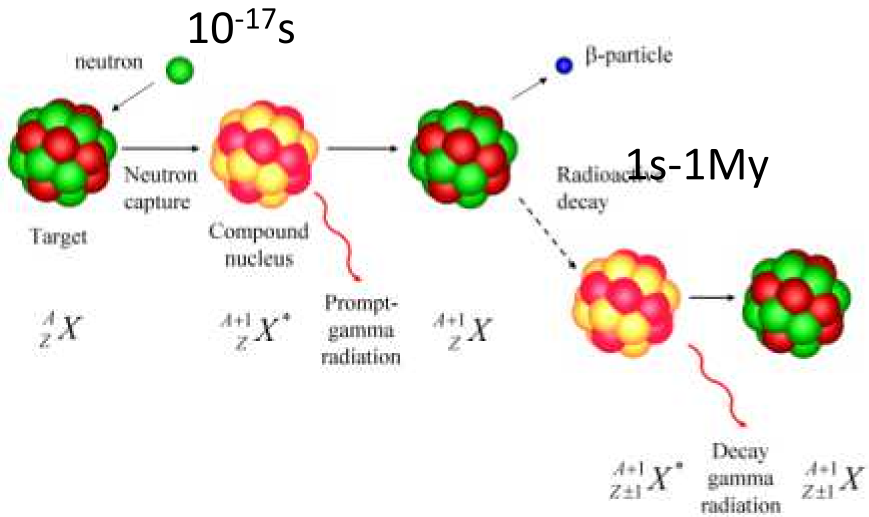
N = density of nuclei
ρ = mass density
M = molar mass
N_A = Avogadro's constant (6.022e23#/mol)

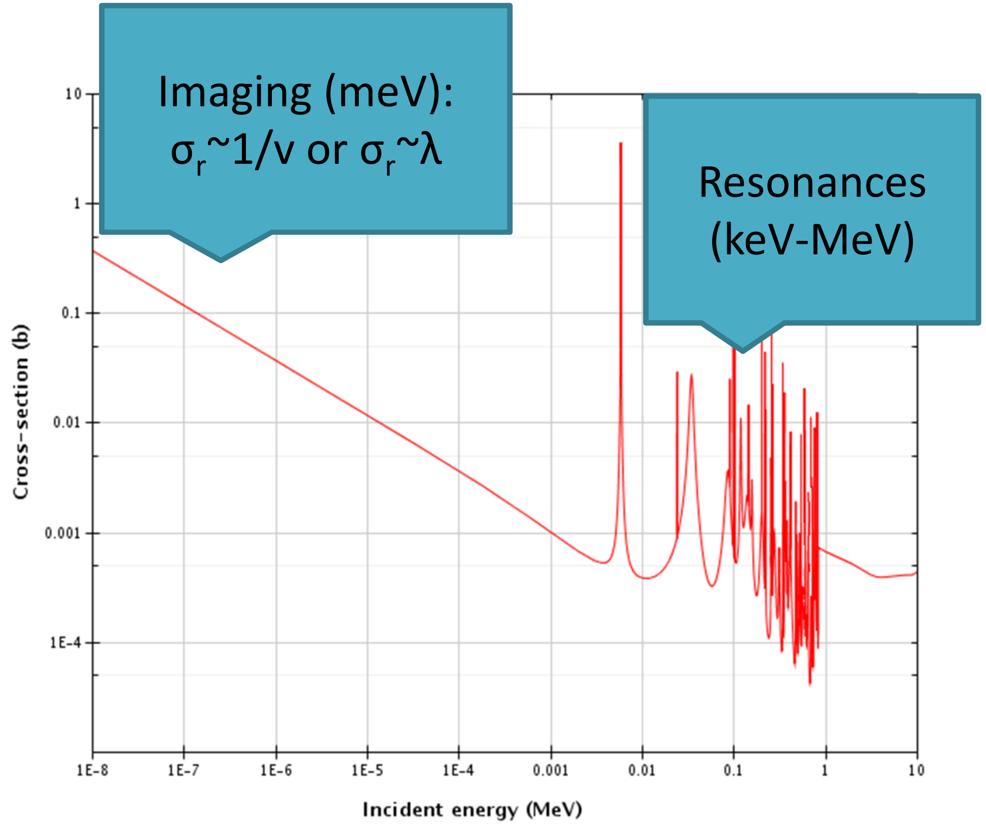
Clearly the interaction (cross-section) is wavelength dependent. What interactions are there possible?

Neutron capture (absorption)

An incident neutron can be absorbed by the nucleus:

- Formation of an excited compound nucleus
- Prompt γ-emission
- Radioactive decay (β,γ)

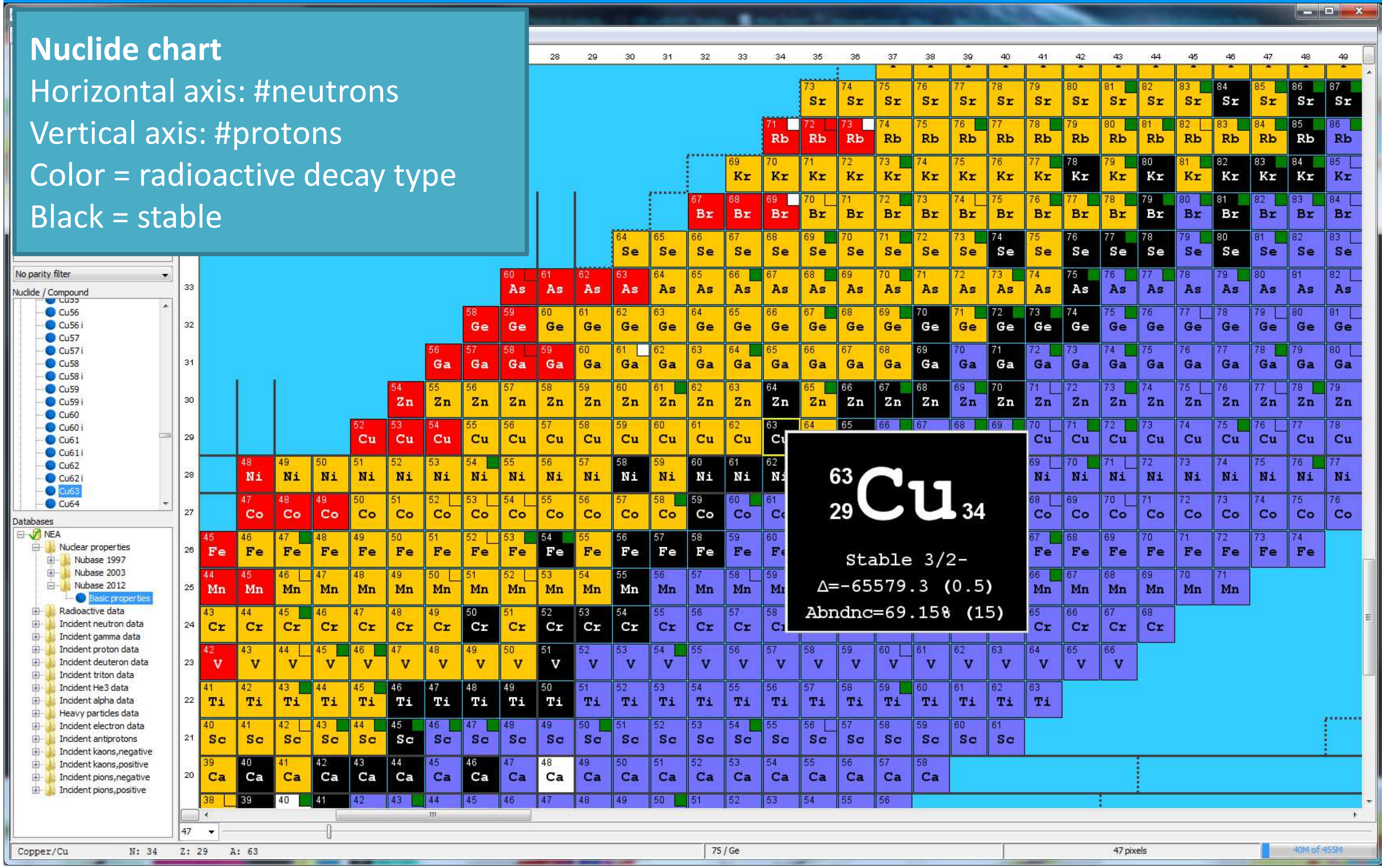




"The slower the neutron, the more time it has to interact and be absorbed"
Or Limit of the Breit-Wigner equation for describing resonances

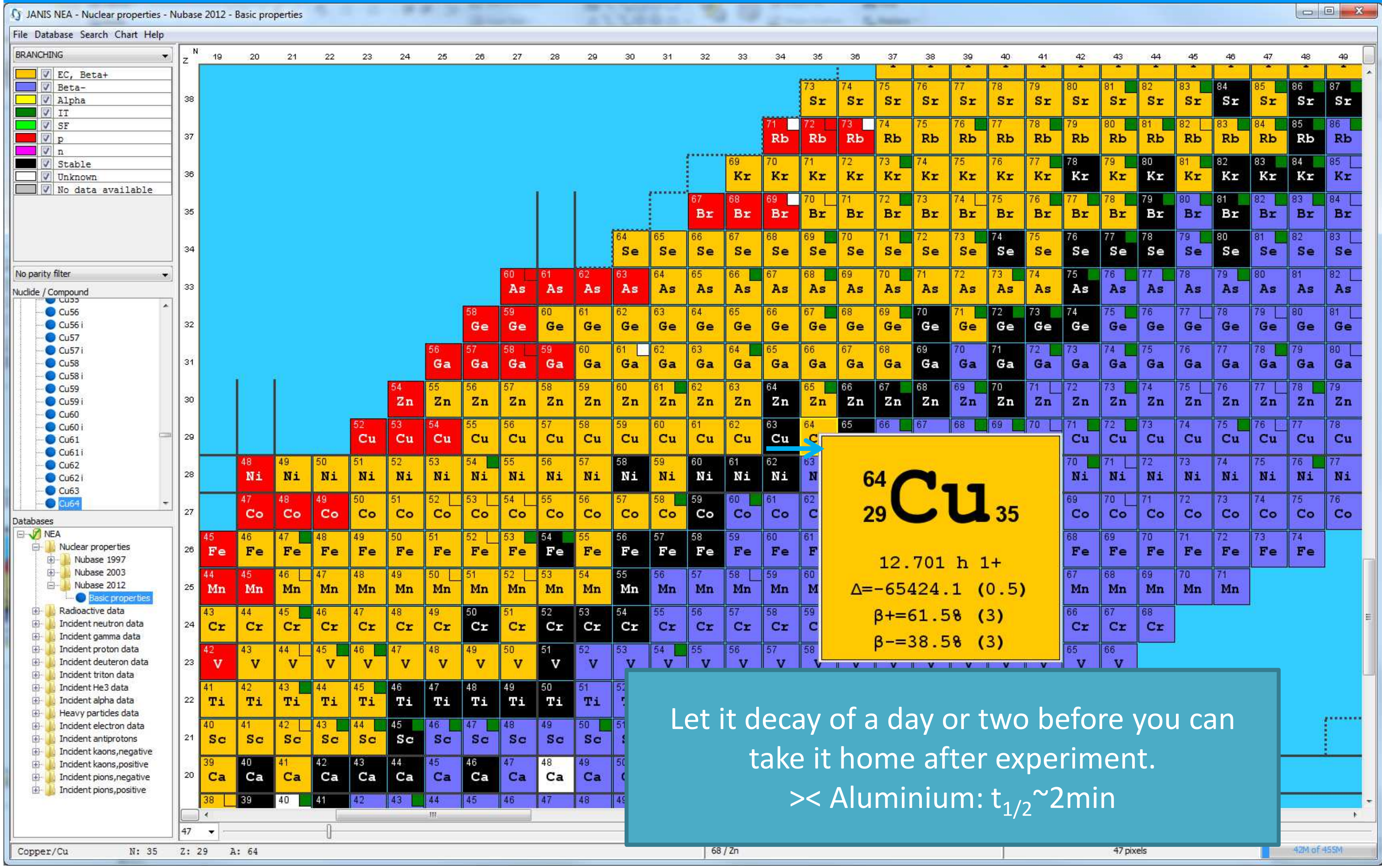
Activation of samples: example Cu

Nuclide chart
Horizontal axis: #neutrons
Vertical axis: #protons
Color = radioactive decay type
Black = stable



http://www.oecd-nea.org/janis/ Check the nuclide chart to see what you can expect.

Activation of samples: example Cu

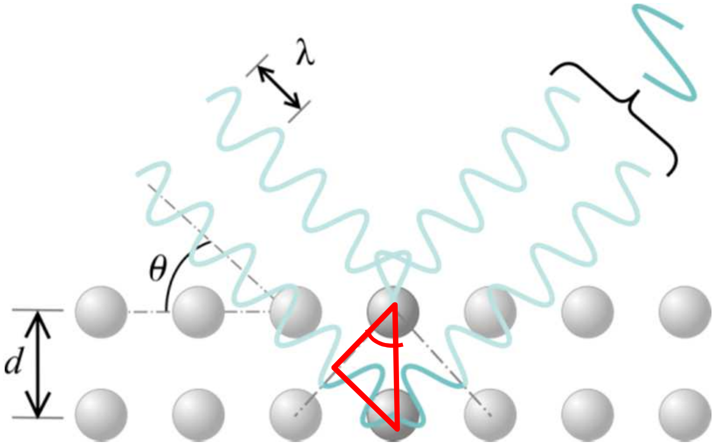


Let it decay of a day or two before you can take it home after experiment.
>< Aluminium: $t_{1/2}$~2min

http://www.oecd-nea.org/janis/

Coherent and incoherent scattering

In many materials the nuclei form an ordered, periodic structure.

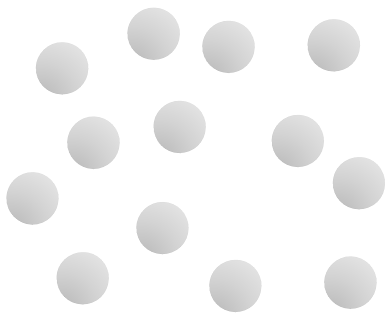


If phase difference equals *nλ*, with *n= 1,2,3,…* reflected waves in phase (constructive interference):

Scattering is *Coherent*

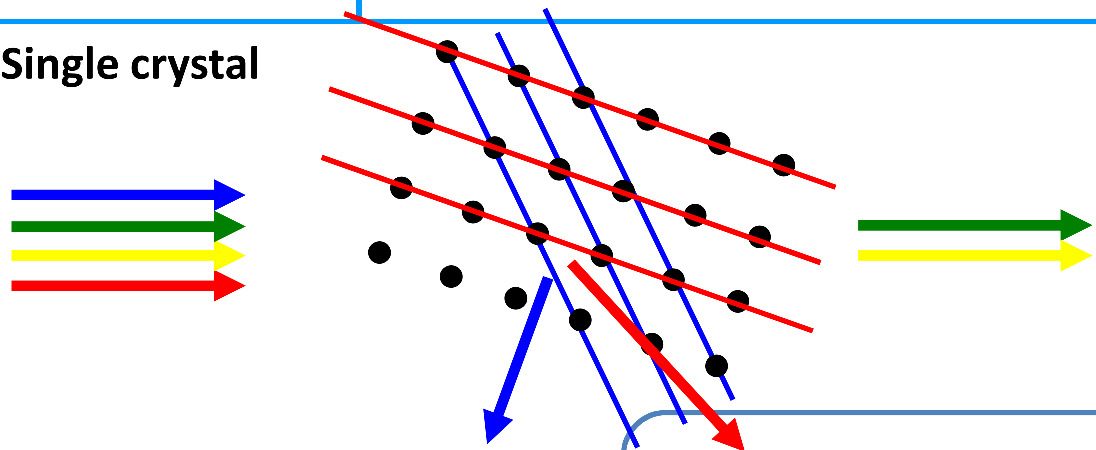
Condition: $2d\sin\vartheta = n\lambda$ **Bragg Law**

Amorphous materials:
no periodic structure is present
scattering is *Incoherent*



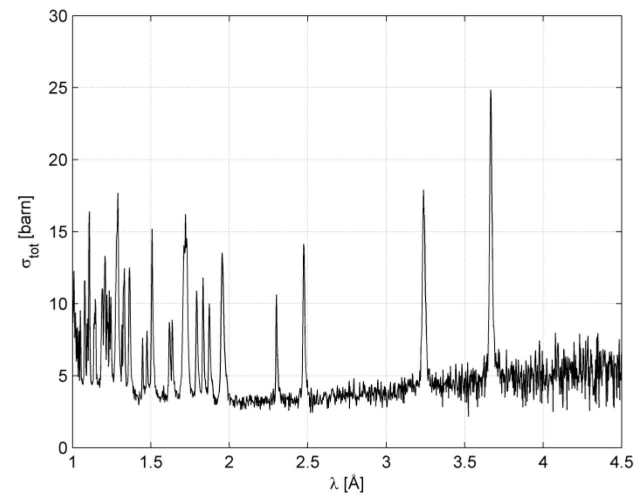
Single Crystals and Pollycrystaline material

Single crystal

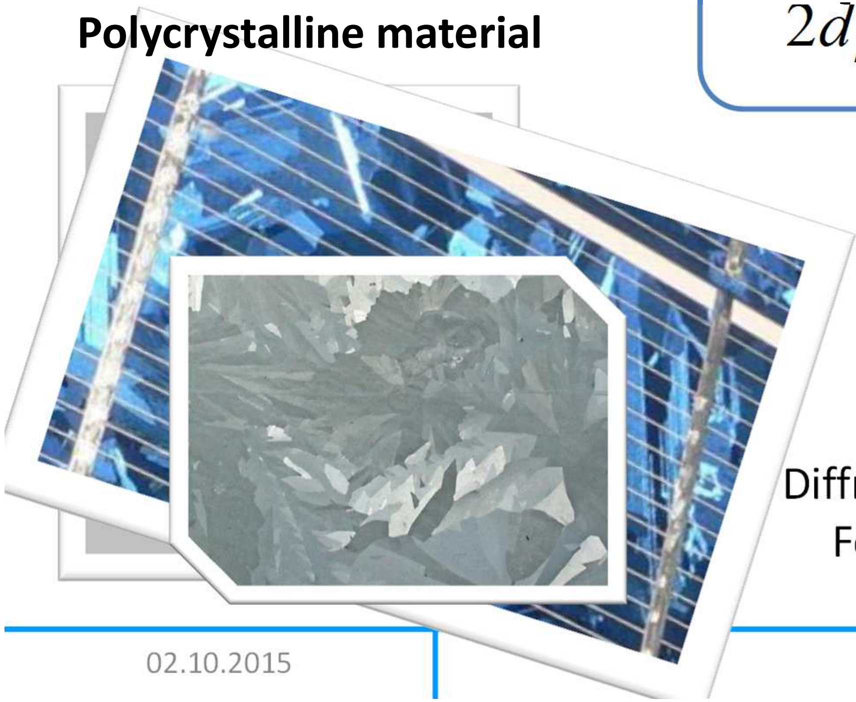


Bragg's law

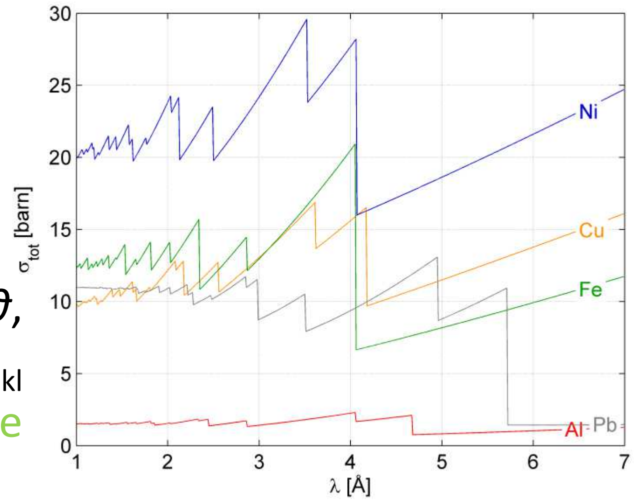
$$2d_{hkl} \sin\theta = \lambda_{hkl}$$



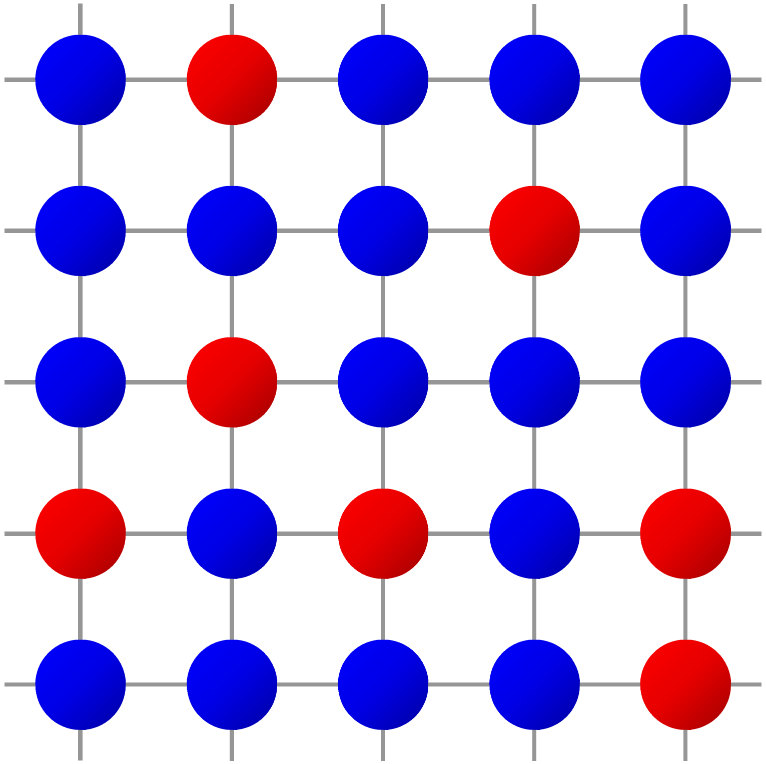
Polycrystalline material



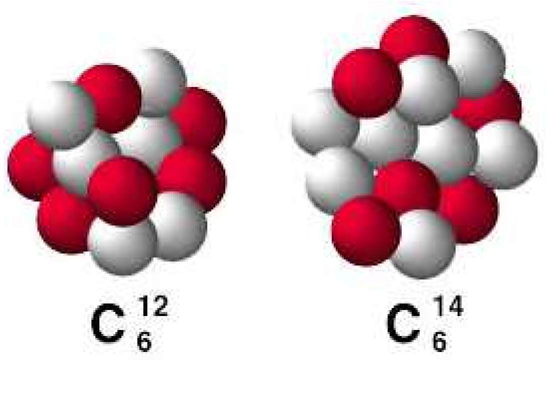
All orientations ϑ,
Diffraction until $\lambda_{hkl} = 2d_{hkl}$
Followed by Bragg Edge



Periodic (?) structure



Different isotopes,
Interaction with nuclear spin I
combines as $J^{\pm} = I \pm ½$
→ neutrons see this difference

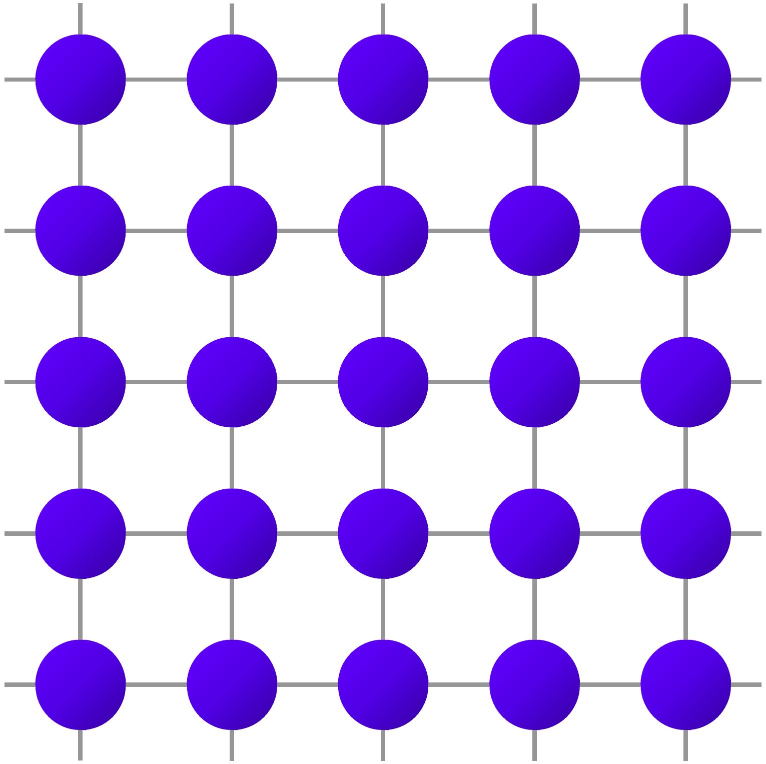


Not truely a periodic structure for our incident neutrons

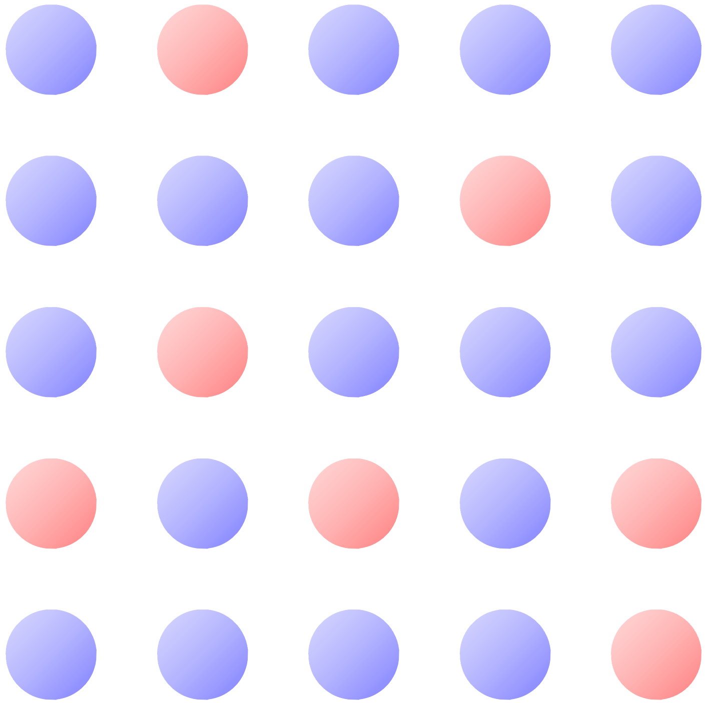
Often you have both coherent and incoherent scatt.

Periodic structure of
average scattering length $\bar{b}$

Corrections to it (random)



+



Coherent scattering

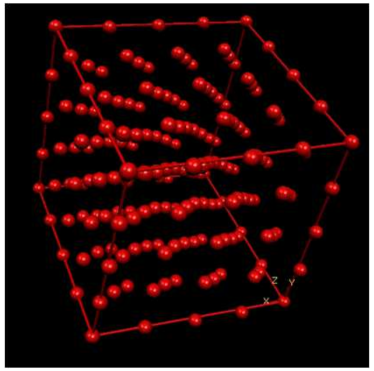
Incoherent scattering

Elastic vs. Inelastic scattering

Crystal at room temperature; Atoms feature thermal motion

The energy associated with atomic vibrations is quantized: phonons

http://www.phonon.fc.pl

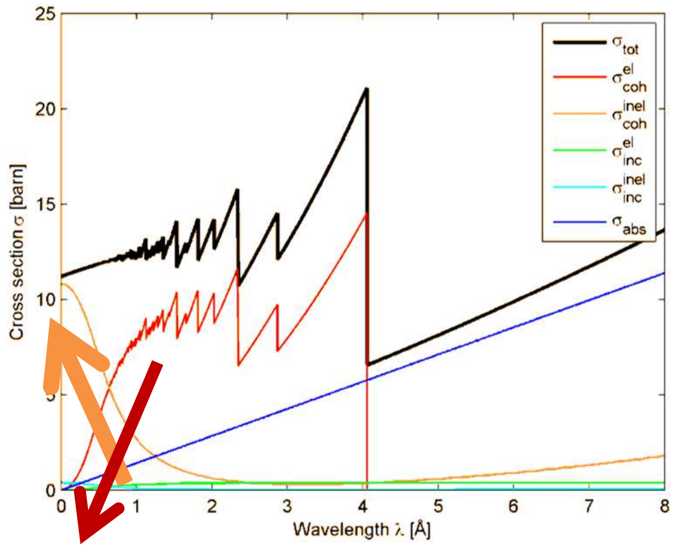


Incident neutron can

- Transfer energy to the crystal (phonon creation)
- Get energy from the crystal (phonon annihalition).

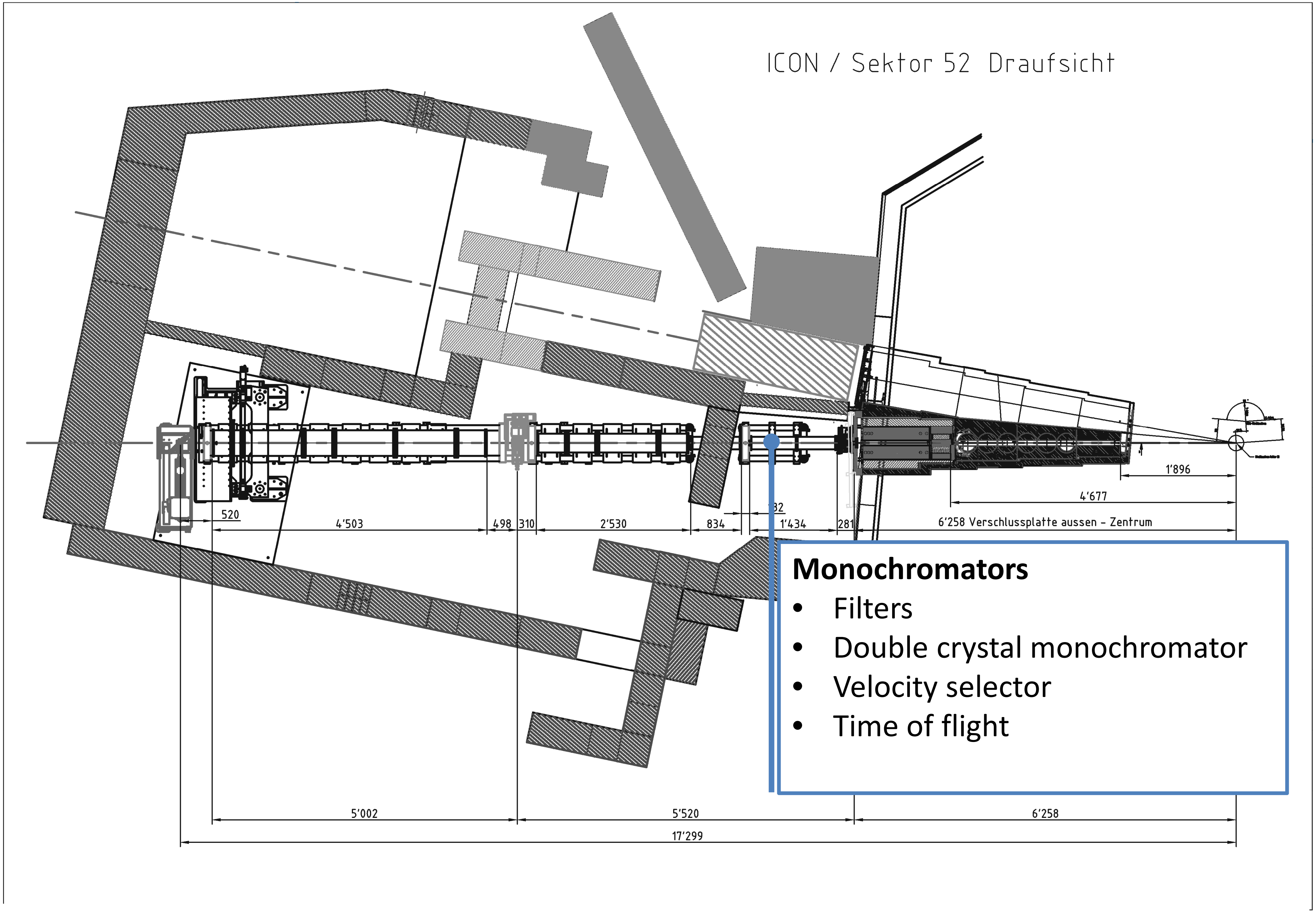
Elastic scattering: no energy transfer

Inelastic scattering: energy transfer



The higher the neutron energy (lower λ),
The more phonons can be involved

σ_{inc} ↗ σ_{coh} ↘

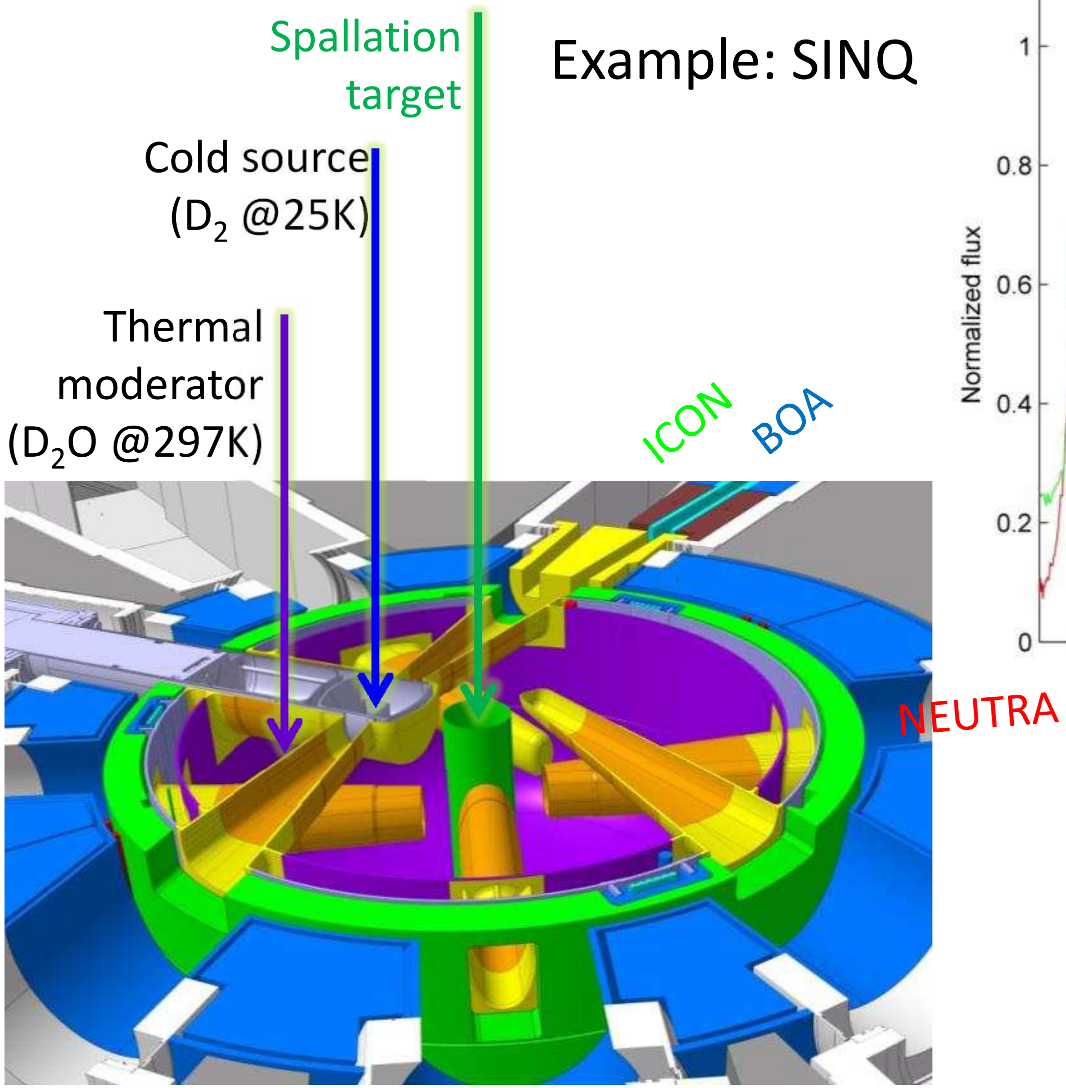


Monochromators

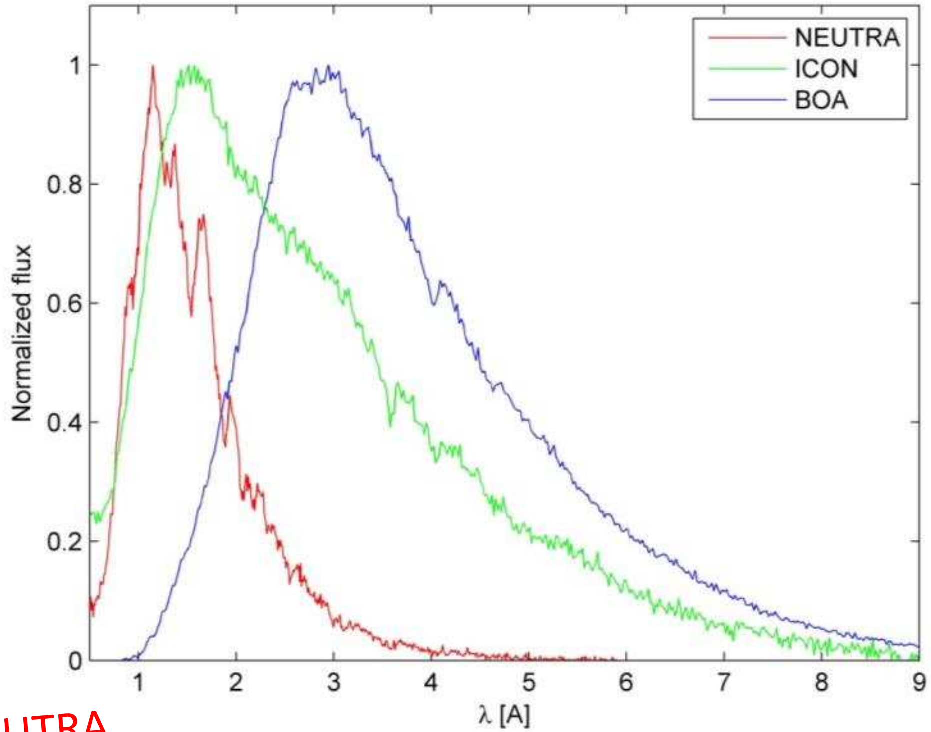
- Filters
- Double crystal monochromator
- Velocity selector
- Time of flight

Different moderators, different average energy

Example: SINQ



SINQ neutron source

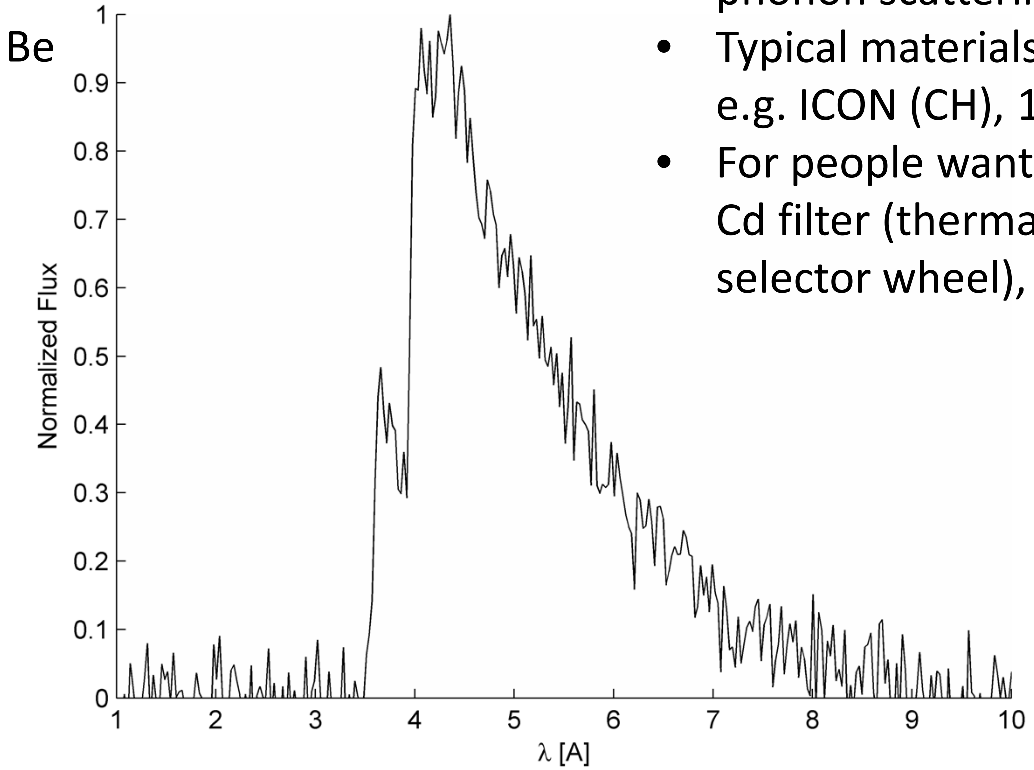


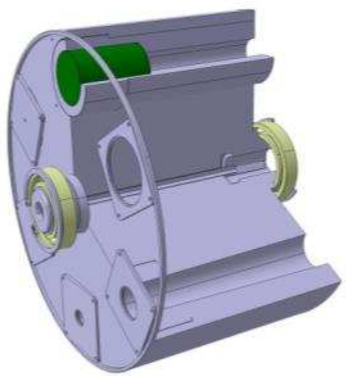
$$\int_{\lambda} I(\lambda)d\lambda = \int_{\lambda} I_0(\lambda)e^{-\Sigma(\lambda)x}d\lambda$$

If we use the full beam spectrum, we get the average **thermal** or **cold** cross-section

Filters

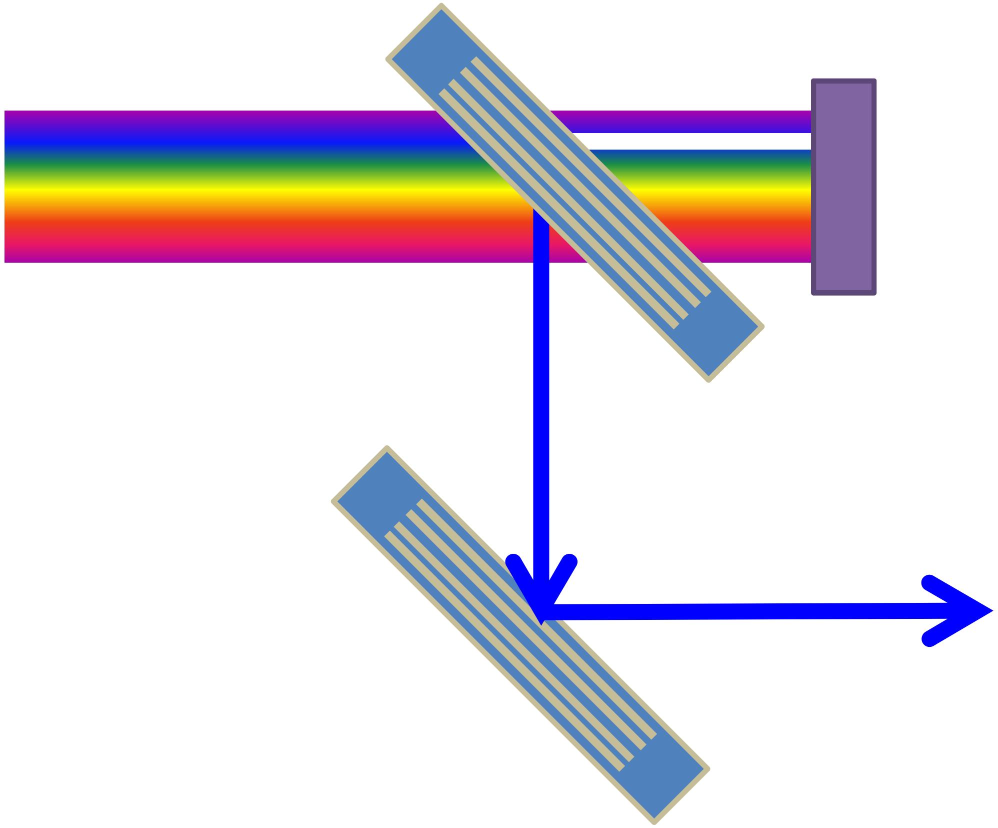
- Cross section high before the Bragg cut-off, low after
- Take a block thick enough, only neutrons $\lambda > \lambda_{Br}$ get through
- (Cool it to limit losses above λ_{Br} : reduce inelastic phonon scattering)
- Typical materials: Be (~4Å), C(~6.7Å)
  e.g. ICON (CH), 100mm in selector wheel)
- For people wanting only epithermal neutrons:
  Cd filter (thermal resonance). E.g. Antares II (D), 2mm in selector wheel), NPP control rods





ICON selector wheel

Double crystal monochromator



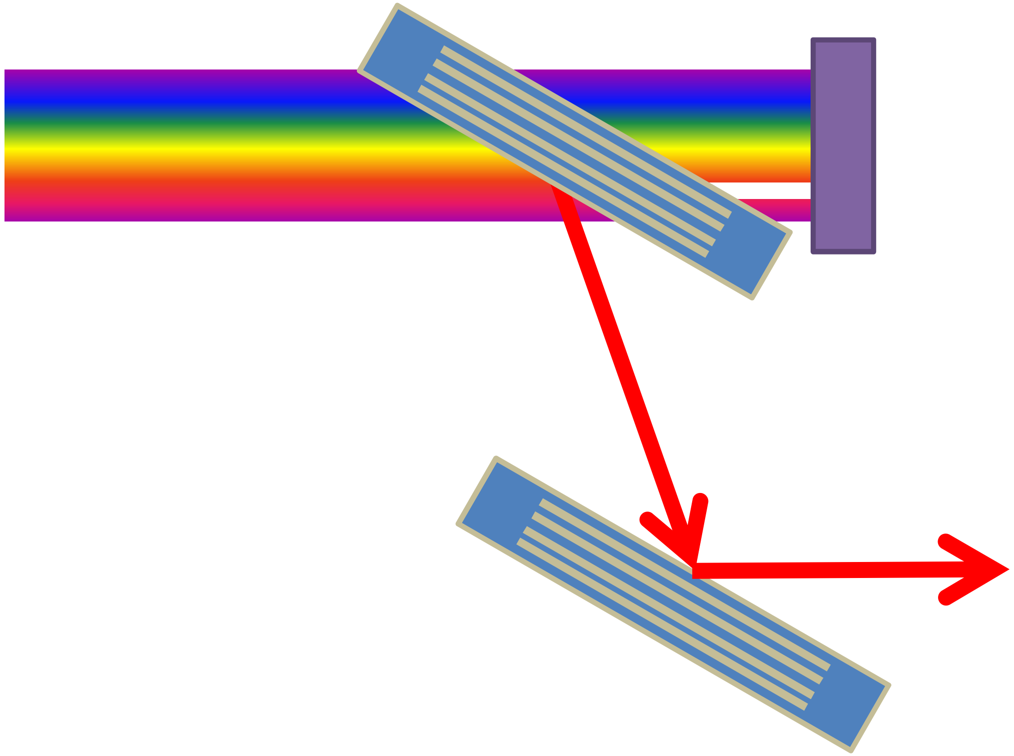
Bragg Law

$$2d \sin\theta = n\lambda$$

Set crystal angle to $\theta_1 \rightarrow \lambda_1$

Second crystal parallel to bring it back in the original direction

Double crystal monochromator



Bragg Law

$$2d\sin\theta = n\lambda$$

Set crystal angle to $\theta_1 \rightarrow \lambda_1$

Second crystal parallel to bring it back in the original direction

Set crystal angles to $\theta_2 \rightarrow \lambda_2$, + translate

Typical crystals: PG002, Ge, Si

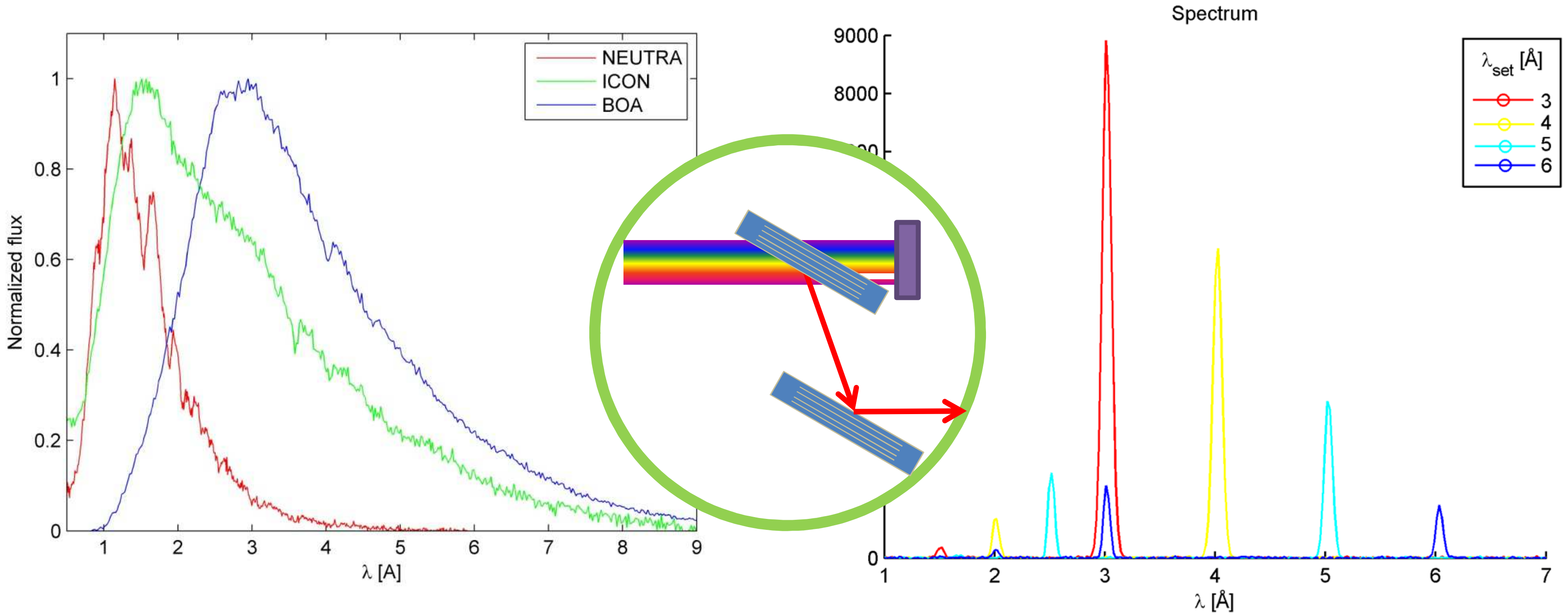
Spectrum

Higher order contributions (n=2,3,… reflections)

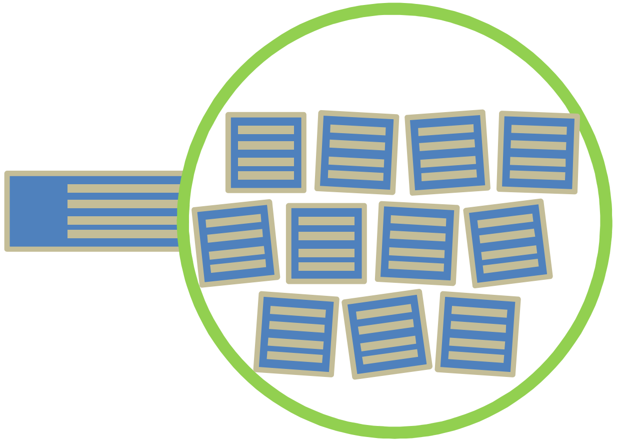
$$2d \sin\theta = n\lambda$$

Crystal set to θ→λ, also λ/2 (n=2), λ/3 (n=3)…

Solution: PG002 + Be-filter / Ge, Si: 2nd order forbidden (F=0)



Spectral width



Single crystal
= small blocks (~100µm) of small misorientation

This is called mosaicity

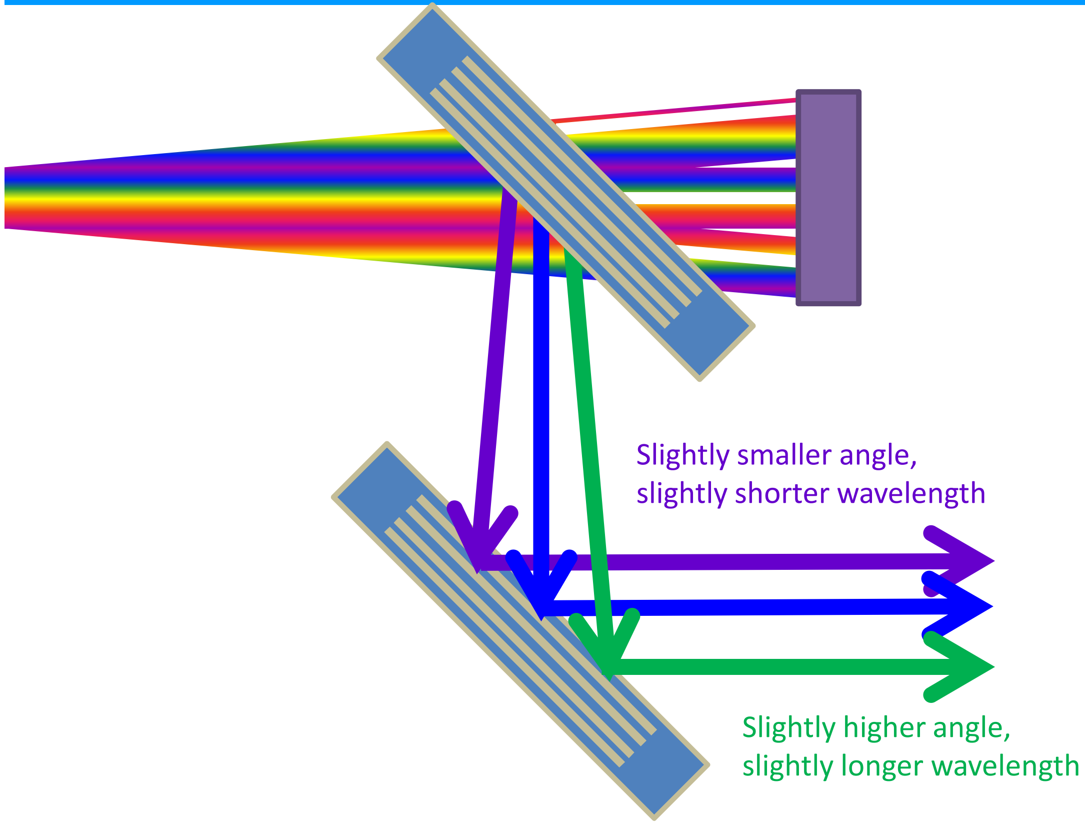
So θ and hence λ ar no δ-function

The better the monochromaticity, the less neutrons you get on you’re detector!
(So the longer the exposure time for one image, let alone a tomography)

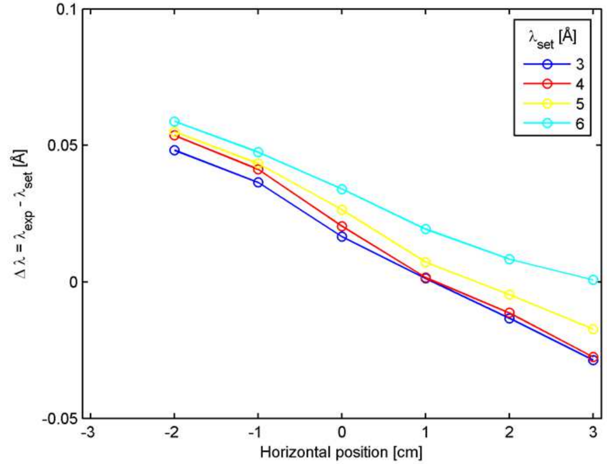
Si, Ge: too perfect ($\Delta\lambda/\lambda$ <0.01%), hot bending to introduce defects
PG002: Commercially available, only ordered in the [002] direction, the choice in imaging

Imaging:
- Mosaic spread ~ 0.4°
- Monochromaticity $\Delta\lambda/\lambda$ ~1-3%

Spectral uniformity



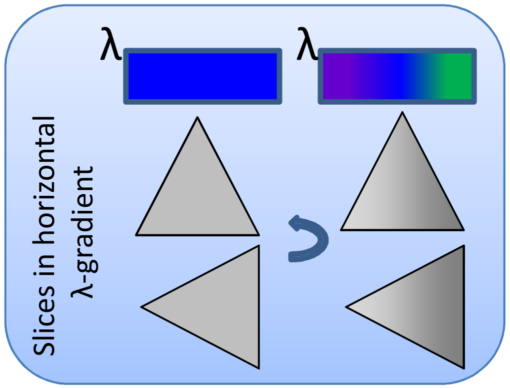
Beam hitting left vs right side FOV:
Δθ ~ 0.5° (+5 to -5cm FOV @ 5m)



Morgano, M.; Peetermans, S.; Lehmann, E.; Panzner, T. & Filges, U.
Neutron imaging options at the BOA beamline at Paul Scherrer Institut
Nuclear Instruments and Methods, section A, **2014**, *754*, 46-56

Imaging implication:

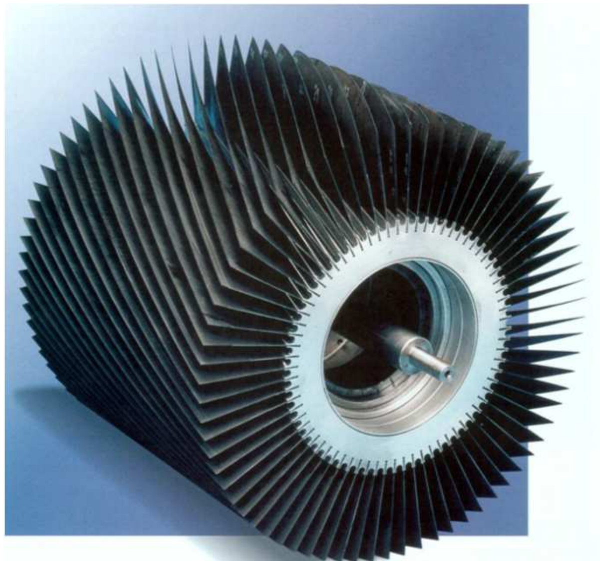
- Crystals horizontal: same wavelength within the slice, vertical gradient accross sample
- Crystals vertical: broader effective wavelength band, vertically homogeneous



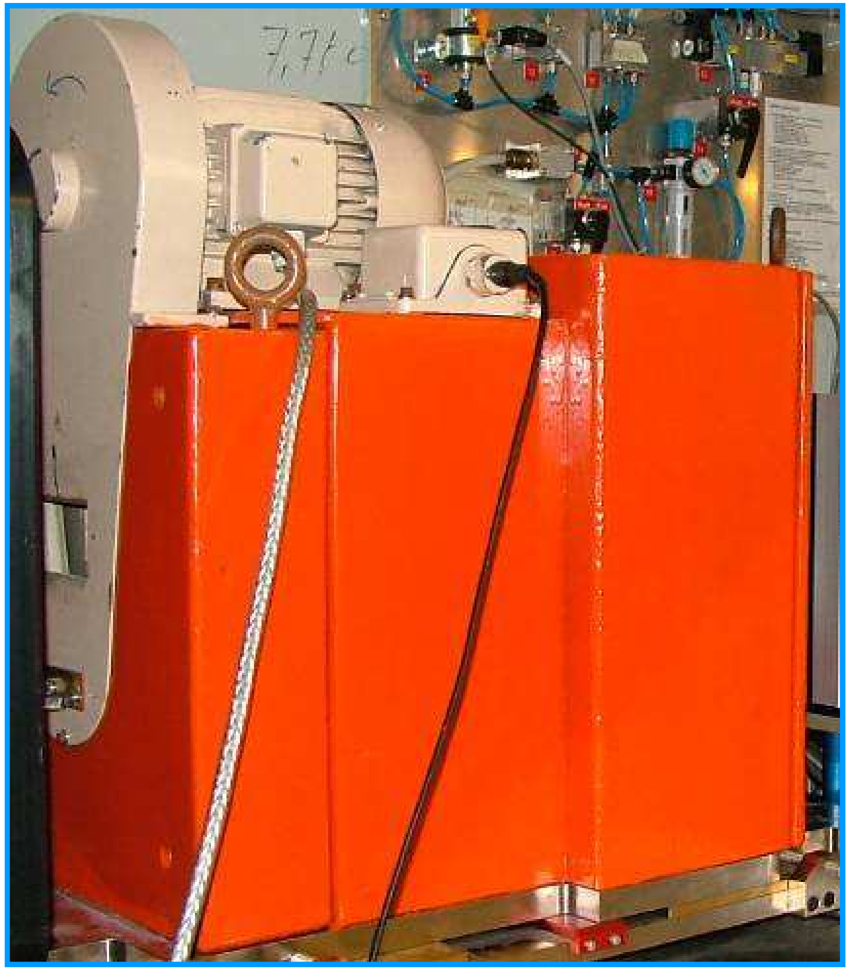
Neutron velocity selector

Δλ/λ=15%

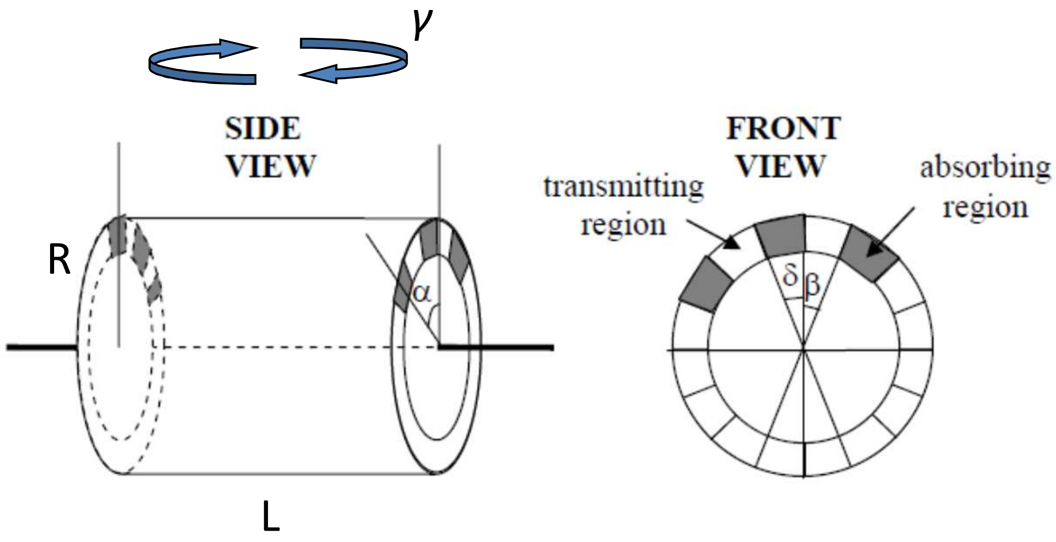
+ Relatively high count rates
+ ES Imaging past Bragg cut-off
- Sharp Bragg edge imaging

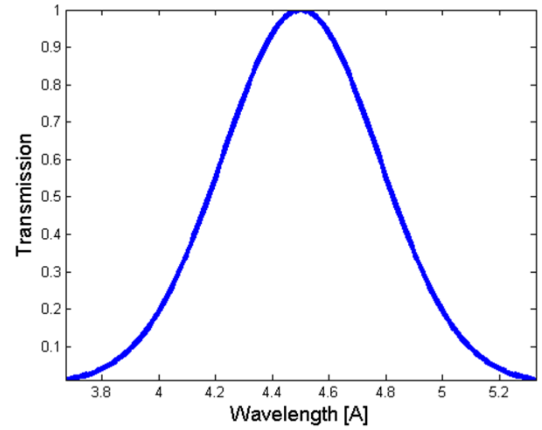


http://www.sii.co.jp/jp/segg/files/2013/03/file_PRODUCT_MASTER_1381_GRAPHIC02.pdf



Theory





Central wavelength

$$\lambda = \frac{\alpha h}{L m_n \omega}$$

Wavelength Spread

$$\frac{\Delta\lambda}{\lambda} = \frac{\beta}{\alpha}$$

Tilt w.r.t. the beam

$$\alpha_{eff} = \alpha + \gamma \frac{L}{R}$$

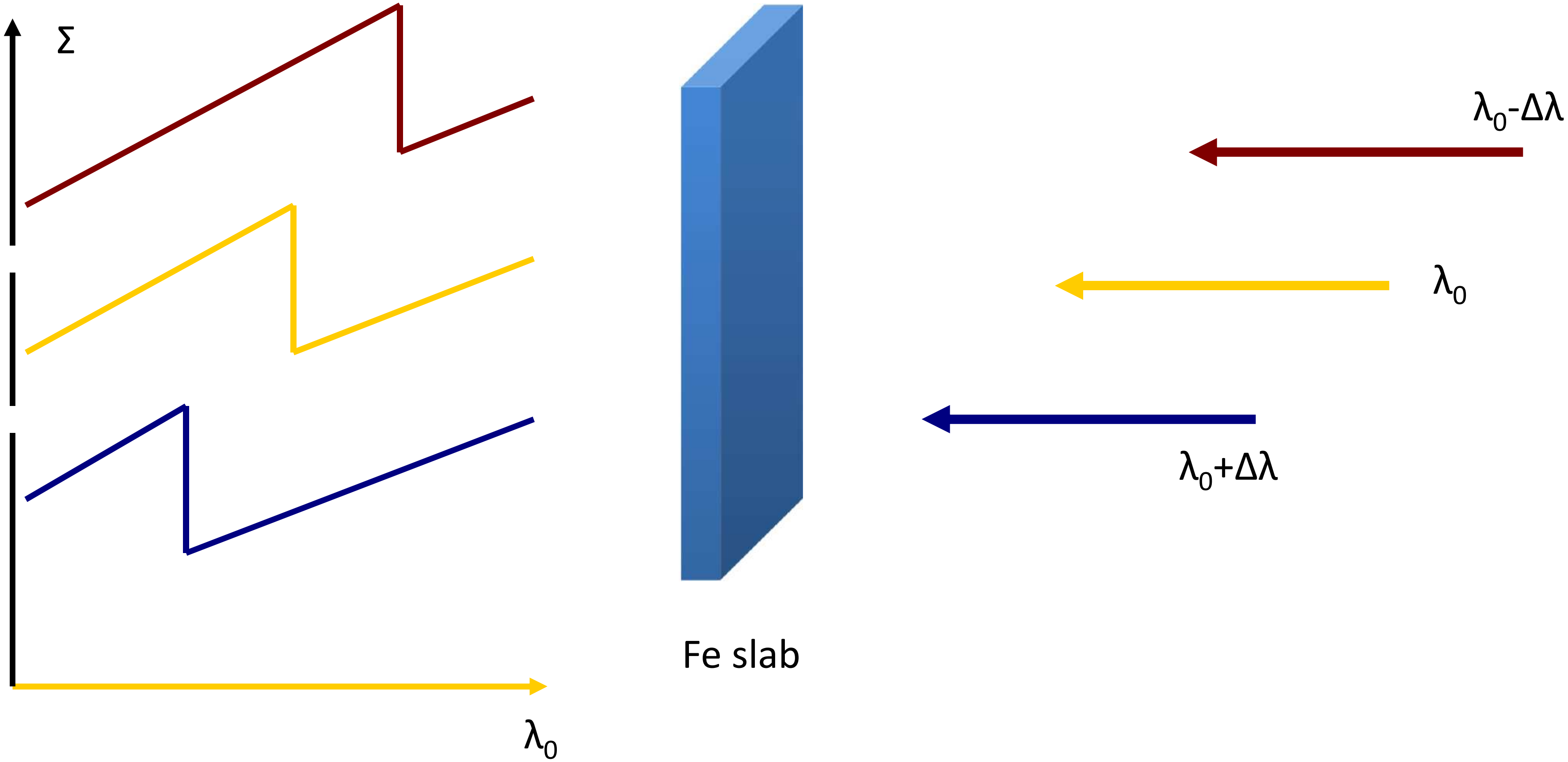
Beam divergence ⟷ Velocity selector tilt ⟶ Spectral shift over the FOV!

Case of ICON: L=400mm, R_c=82.5mm, α=12.5°, β=1.6°, γ=0.5°, L/D=343

Shift up to 1Å over the Field Of View!

Image taken from B. Hammouda, Probing nanoscale structures – the SANS toolbox

Measuring the wavelength shift

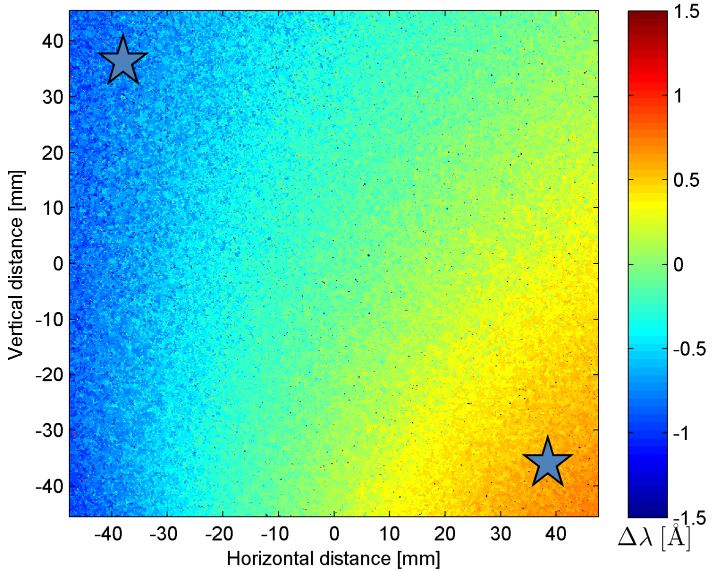


Calibration energy-scan of an iron plate (5mm)

A shift in the incoming spectrum will also induce a shift in the observed Bragg edge position

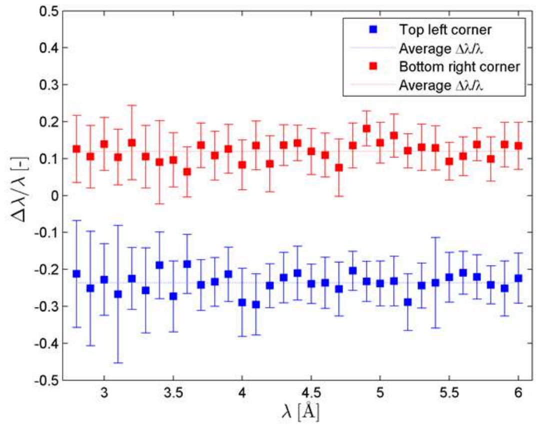
How to deal with it

But know the shiftmap at one wavelength, you know it for all



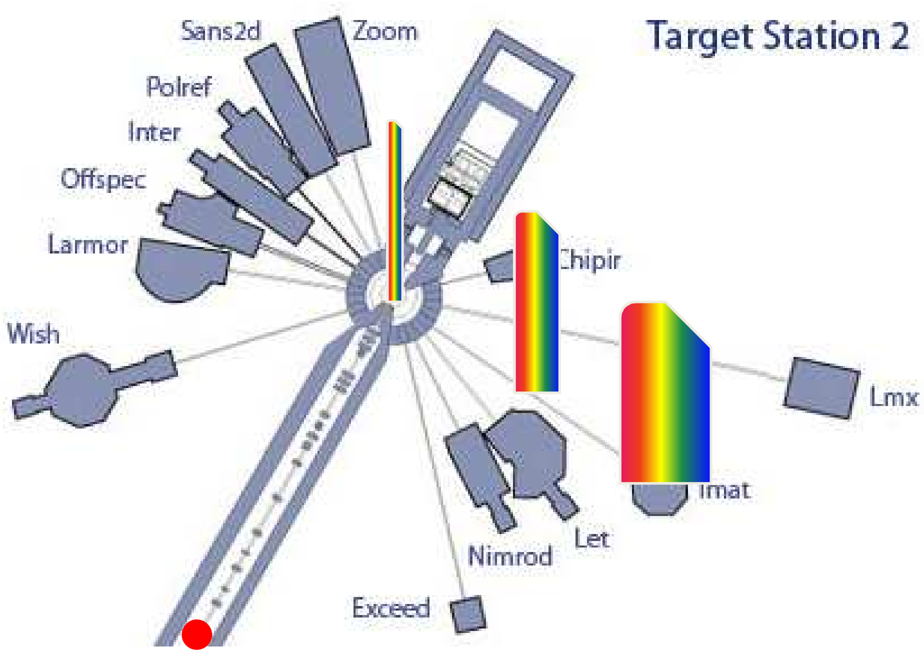
$$\frac{\Delta\lambda_1}{\lambda_1} = \frac{\gamma L}{\alpha R}$$

$$\frac{\Delta\lambda_1}{\lambda_1} = \frac{\Delta\lambda_0}{\lambda_0}$$



Peetermans, S.; Grazzi, F.; Salvemini, F. & Lehmann, E.
Spectral characterization of a velocity selector type monochromator for energy-selective neutron imaging
Physics Procedia, **2013**, *43*, 121-127

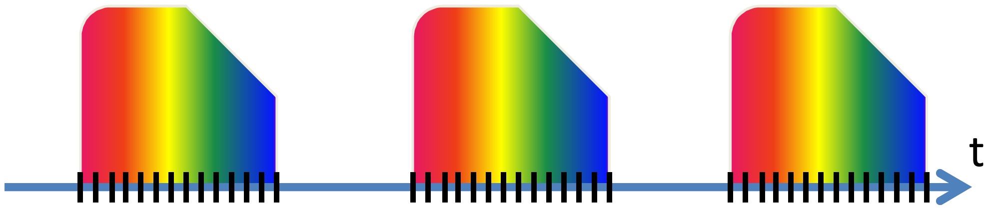
Time of flight



ISIS TS2 lay out (UK)

Pulsed spallation source
e.g. ISIS runs at 50Hz

Record flight time
→ Neutron velocity
→ Neutron energy/wavelength

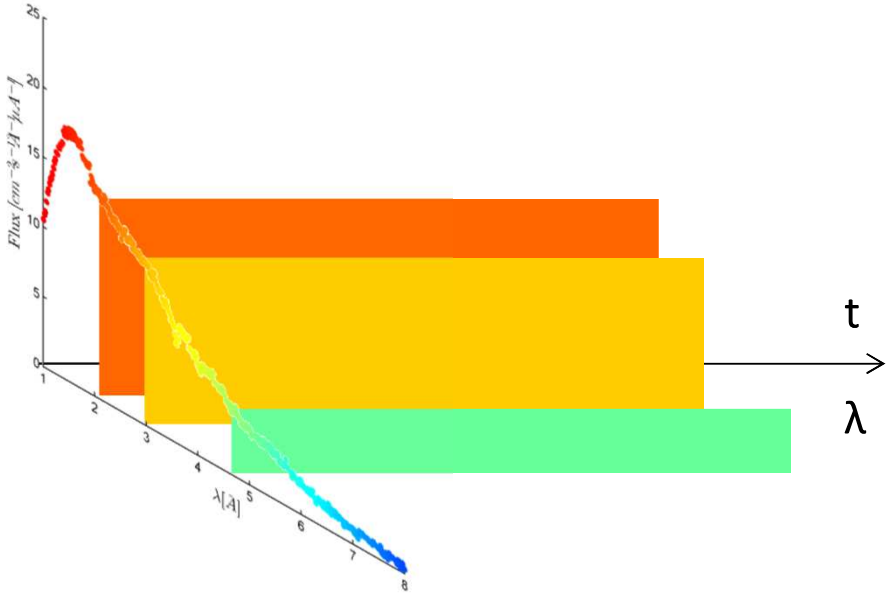


Each short frame (µs) is a monochromatic image
Stack frames (pulses) for sufficient statistics
Whole spectrum recorded at once
Very high energy resolution
Limited by initial pulse width

The right source for the right problem

What if you don't want a full spectrum, just work with a single monochromatic wavelength?

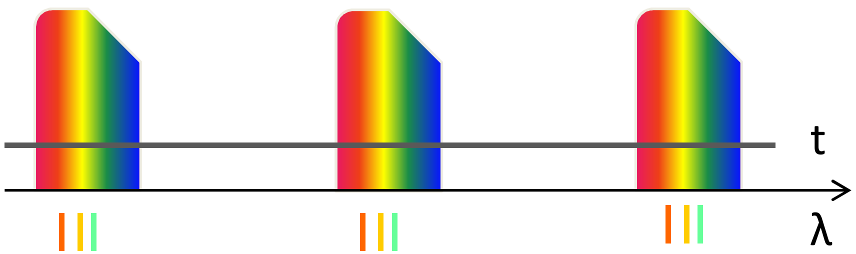
Continuous source



All wavelengths are continuously provided for

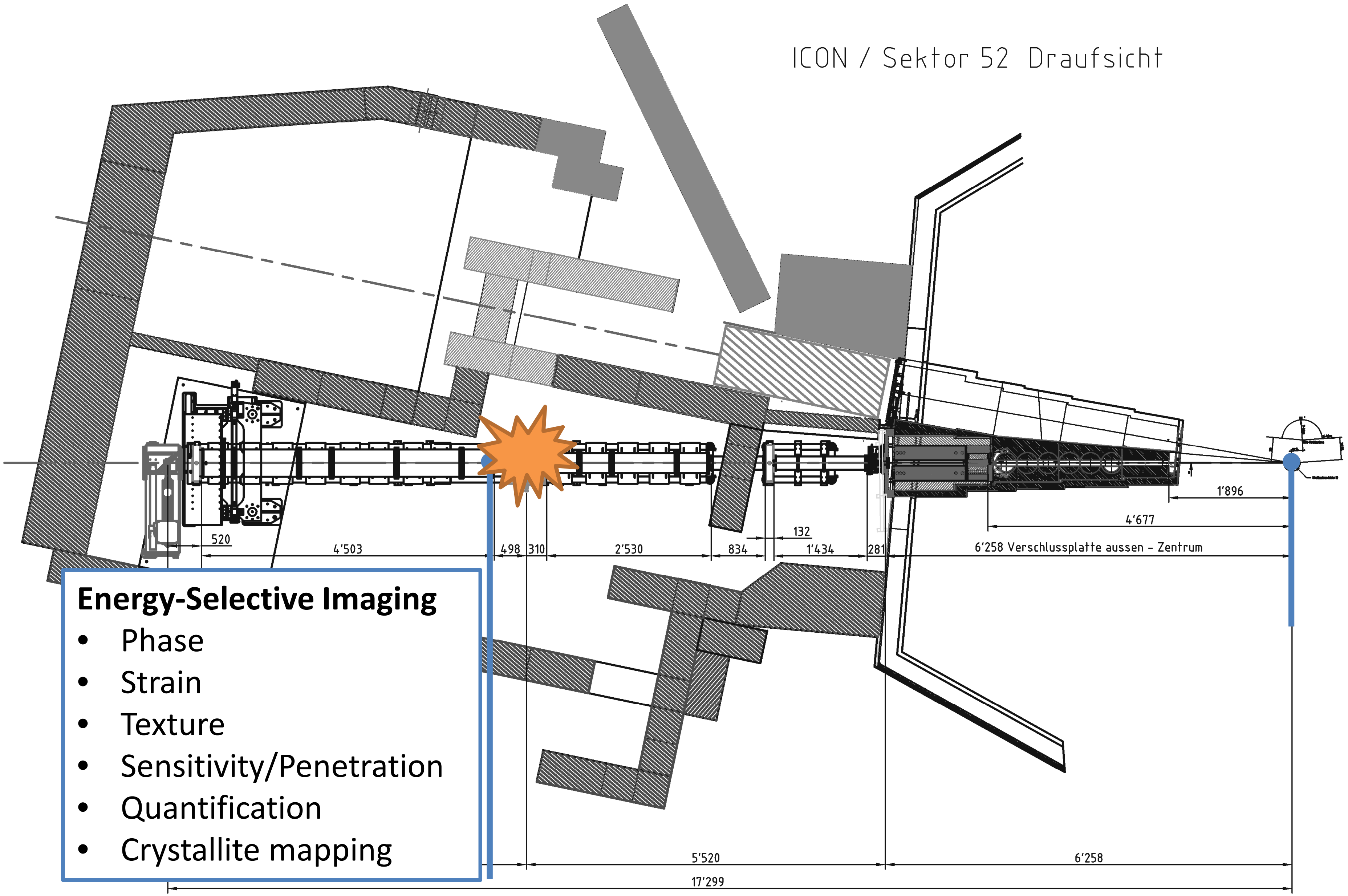
Select one with monochromator

Pulsed source



Record your frame
Wait until next pulse

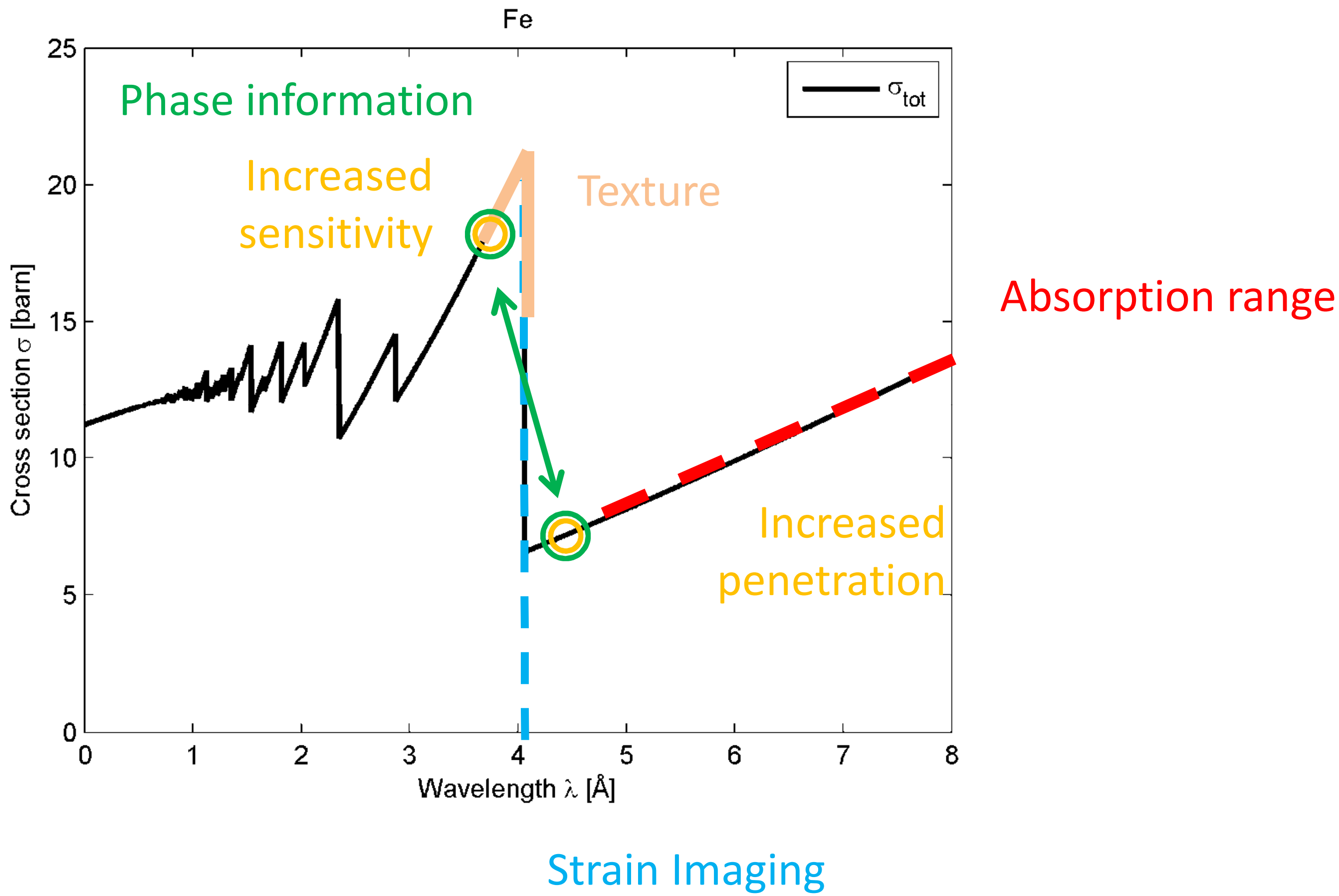
Traditional white beam: time integrated flux important



Energy-Selective Imaging

- Phase
- Strain
- Texture
- Sensitivity/Penetration
- Quantification
- Crystallite mapping

Energy-selective Imaging

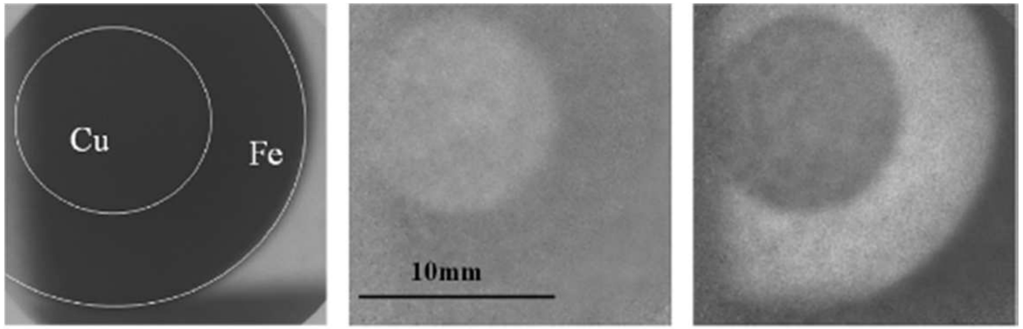


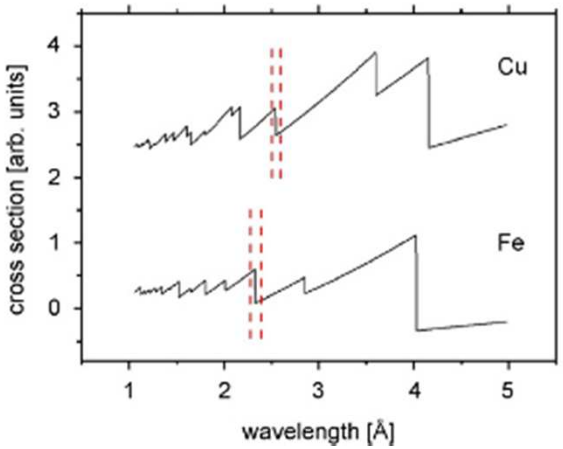
Enhance contrast for one phase

Imaging just before and just after the Bragg edge for Copper

→$\Delta\Sigma_{Cu}$ large, $\Delta\Sigma_{Fe}$ small (its Bragg edge is elsewhere)

Reconstruction of $\Delta\Sigma$ provides phase mapping

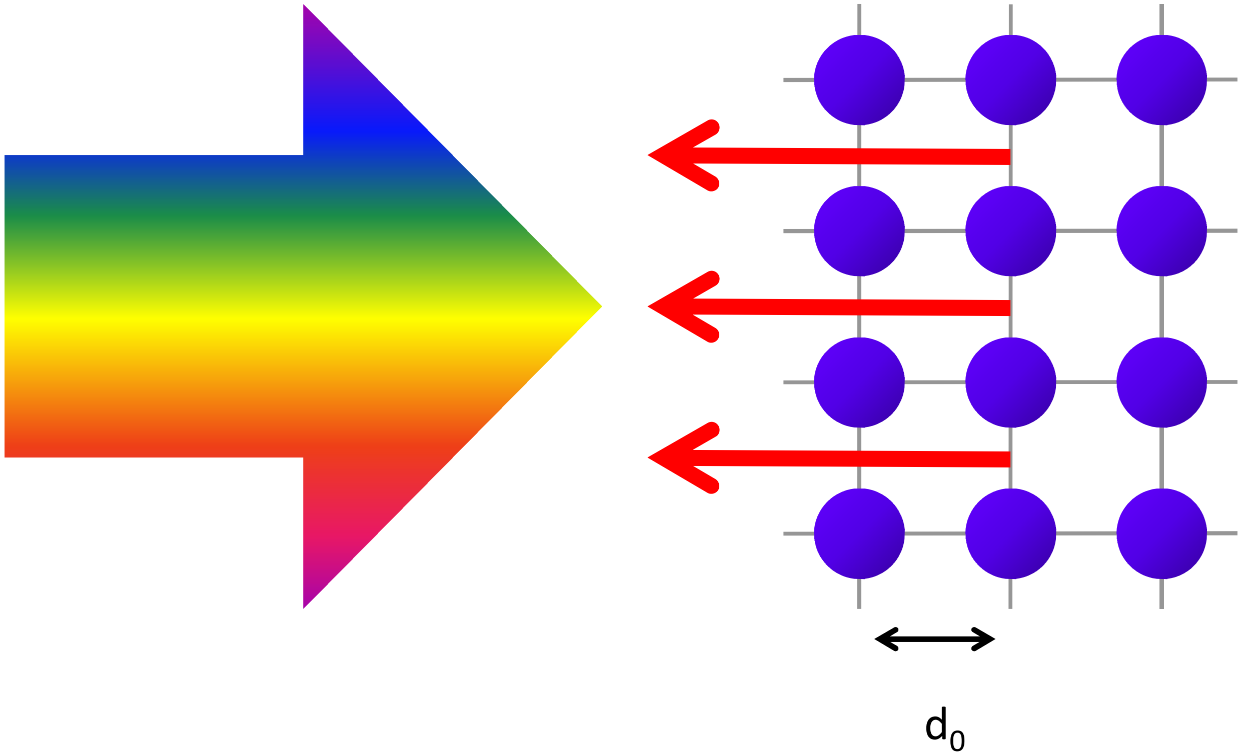




Kockelmann, W.; Frei, G.; Lehmann, E. H.; Vontobel, P. & Santisteban, J.
Energy-selective neutron transmission imaging at a pulsed source
Nuclear instruments and methods in physics research A, **2007**, *578*, 421-434

Bragg edges - unstressed

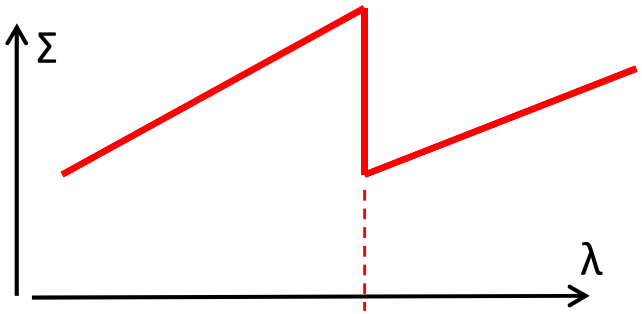
unstressed



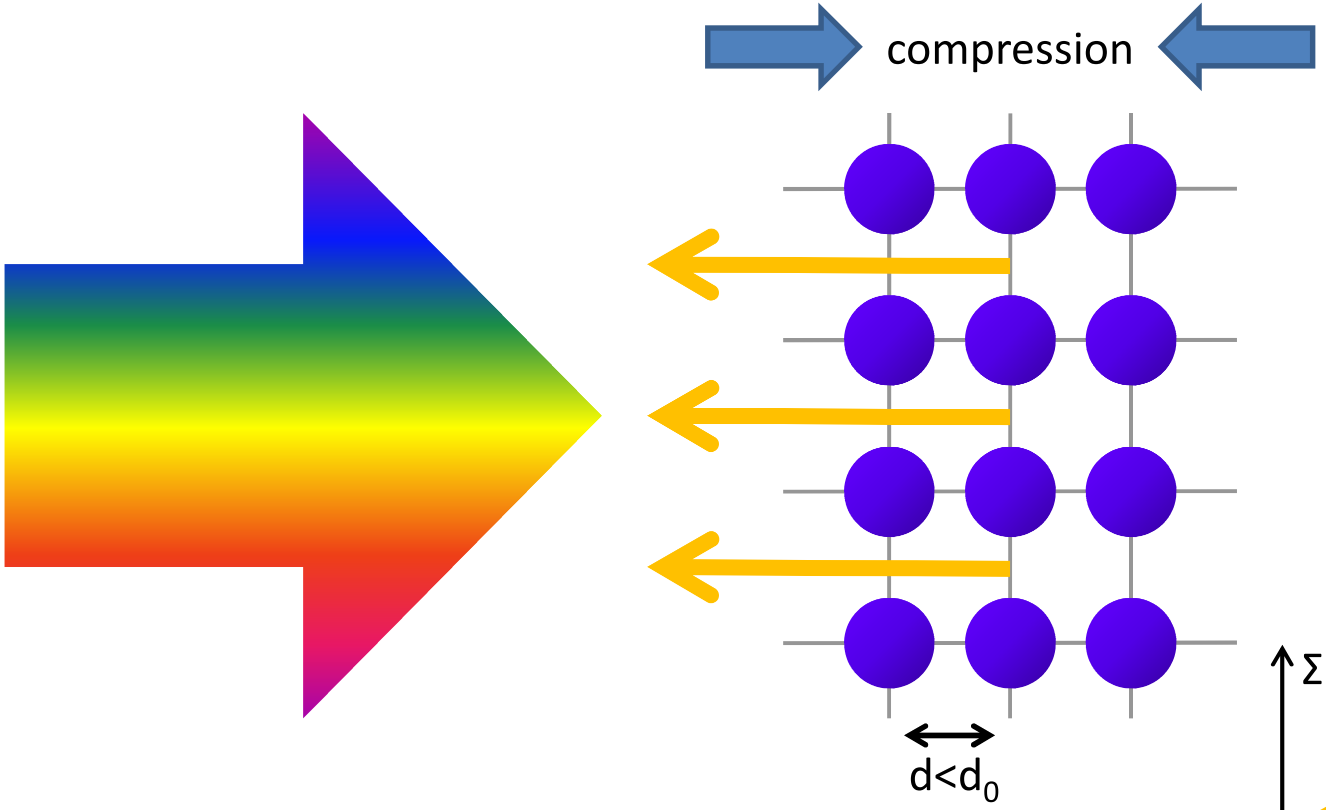
Bragg edge

$$2d_0 \sin \cancelto{90°}{\theta} = \lambda$$

$$2d_0 = \lambda$$



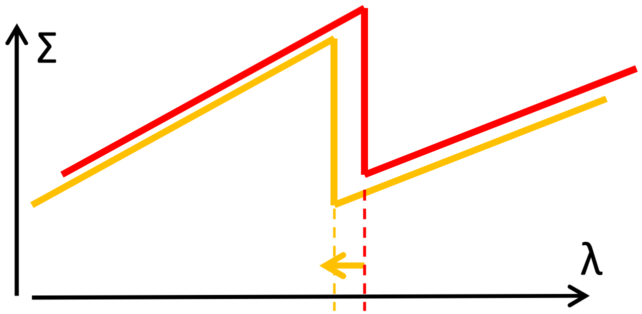
Bragg edges - compression



Bragg edge

$$2d_< \sin \cancel{\theta}^{\,90°} = \lambda$$

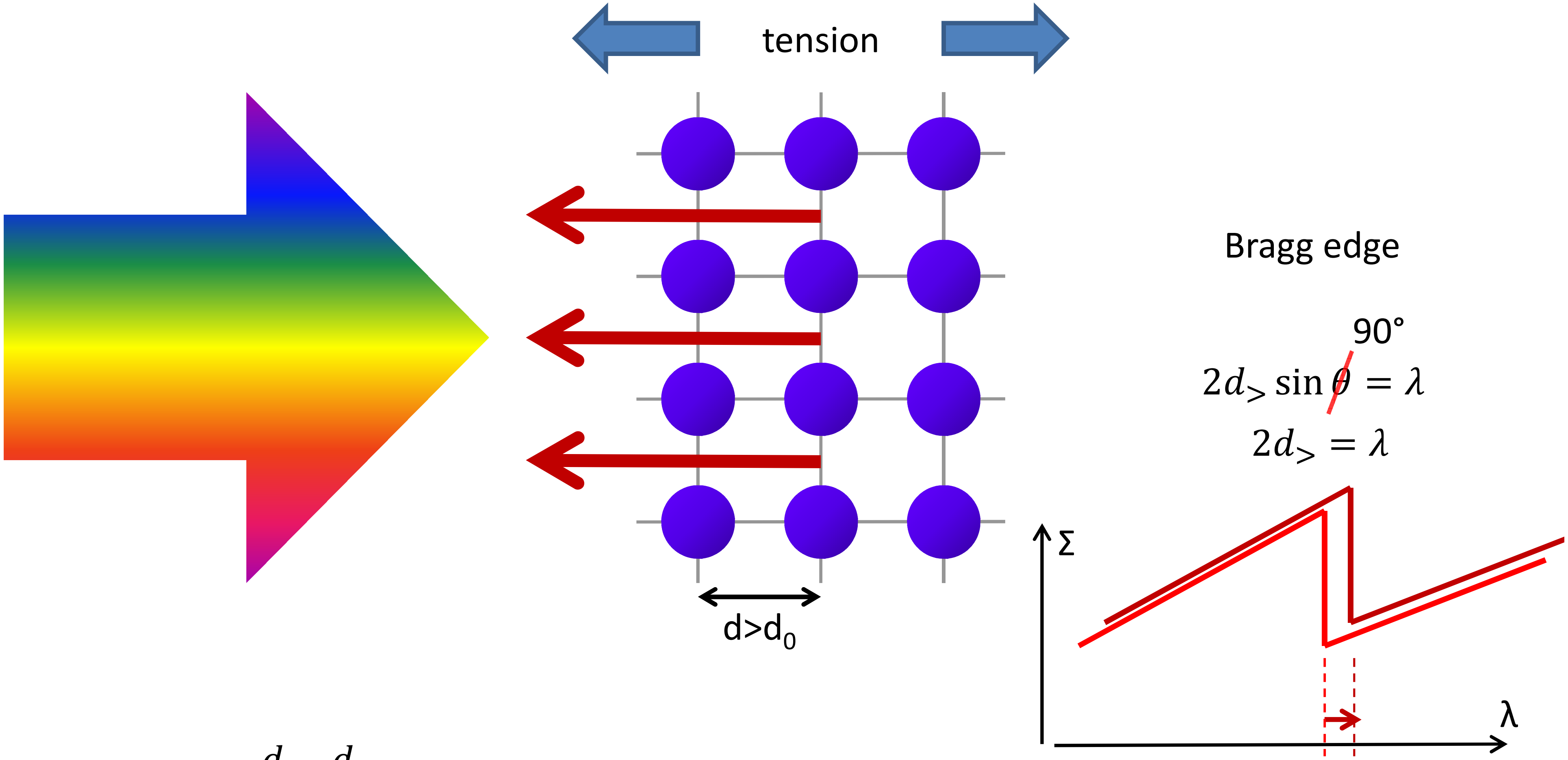
$$2d_< = \lambda$$



Strain $\epsilon = \dfrac{d - d_0}{d_0}$

Stress $\sigma = E\epsilon$

Bragg edges - tension



$$2d_> \sin \cancel{\theta} = \lambda$$

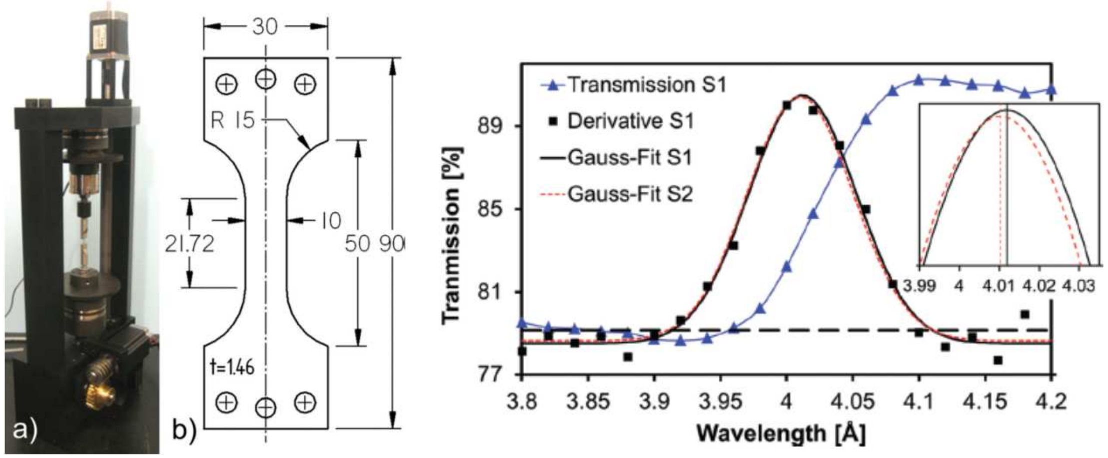
$$2d_> = \lambda$$

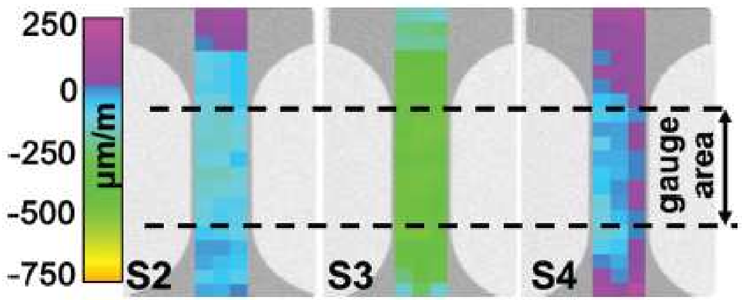
Strain $\epsilon = \frac{d - d_0}{d_0}$

Stress $\sigma = E\epsilon$

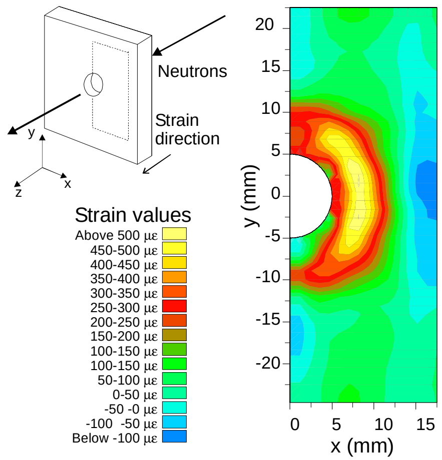
Some examples

Stressing a dogbone sample and perform energy scan
Fit of derivative for increased accuracy in Bragg edge position





Woracek, R.; Penumadu, D.; Kardjilov, N.; Hilger, A.; Strobl, M.; Wimpory, R. C.; Manke, I. & Banhart, J.
Neutron Bragg-edge imaging for strain mapping under in situ tensile loading
Journal of Applied Physics, **2011**, *109*, 093506-1 - 093506-4

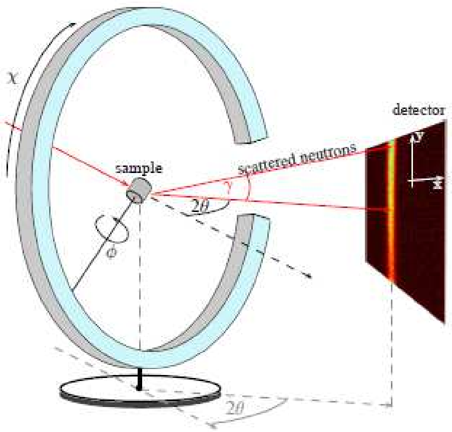


Santisteban, J, *et al. Strain imaging by Bragg edge neutron transmission*
Nuclear instruments and methods in physics research A,
2002, *481*, 765-768

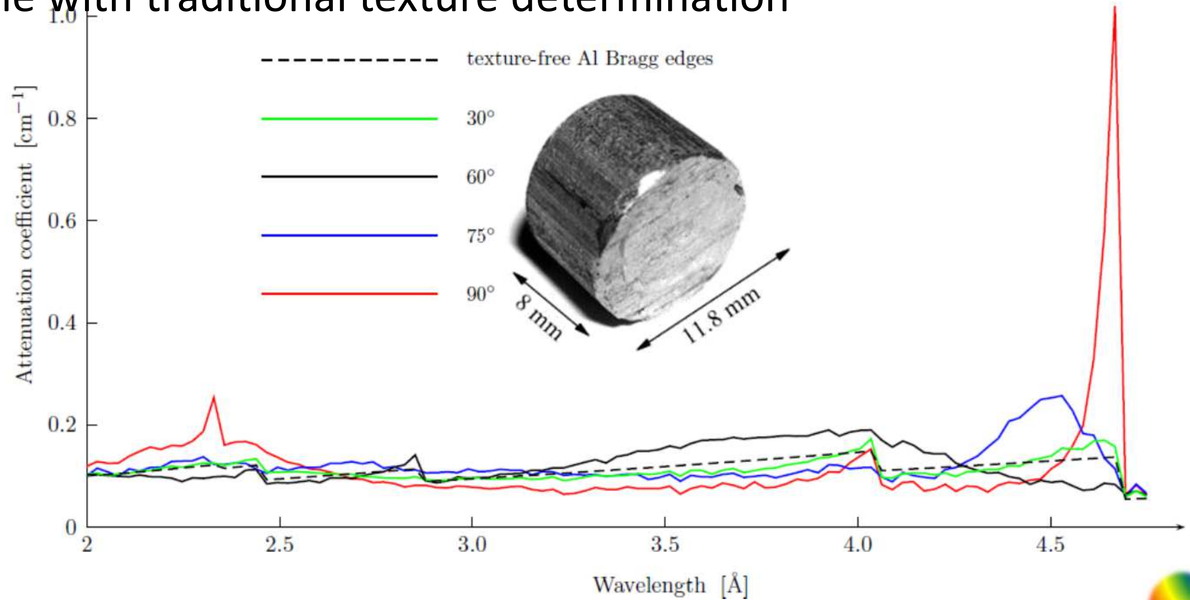
Texture

- No longer all random orientations
- Some orientations occur more than others (preferred orientation / texture)
- Corresponding wavelengths will be scattered out more, others less
- Deformed Bragg edge pattern

- Rotation dependent – be carefull in tomography
- Rather qualitative (only March-Dollase texture can be treated mathematically)
- But spatial resolved – combine with traditional texture determination



Experimentally determining diffraction strength for each sample orientation in eulerian cradle



Bragg edge pattern for a textured aluminium cylinder

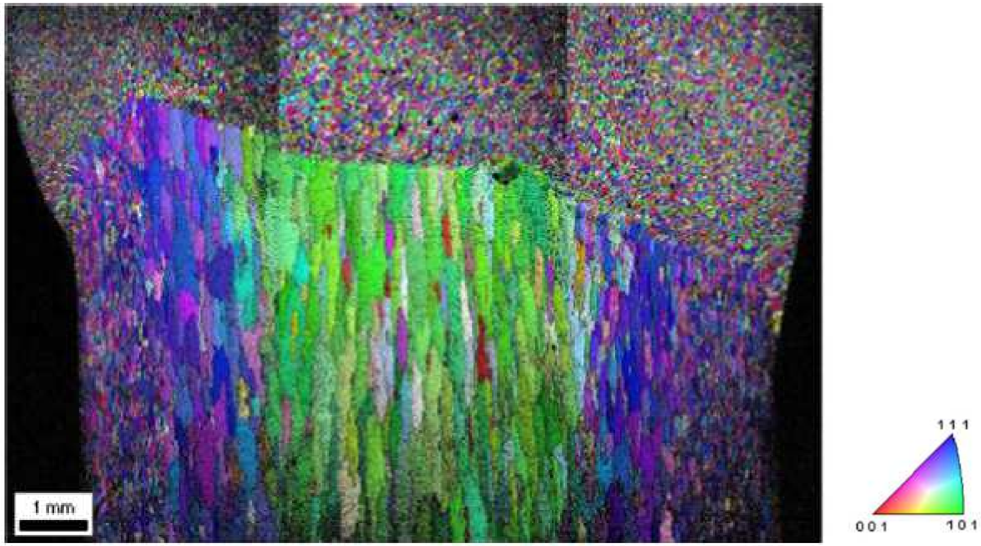
Boin, M. Developments towards the tomographic imaging of crystallographic structures. *The Open University*, 2011

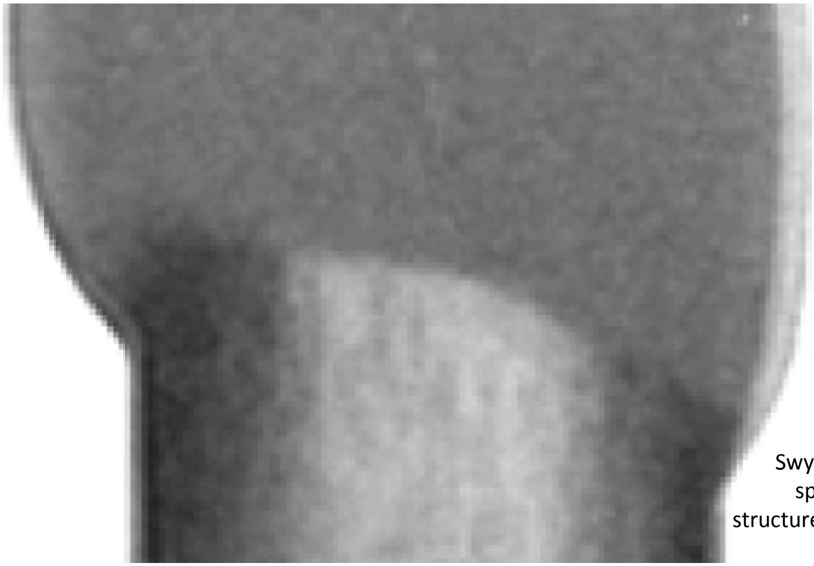
Texture example

Weld on
rolled aluminium

Orientation map from
electron microscopy

Surface mapping
~ 1day

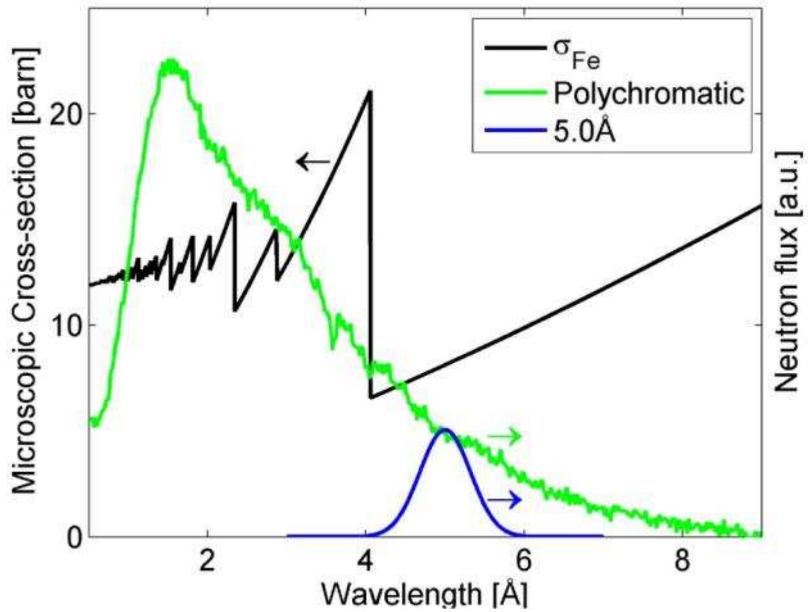


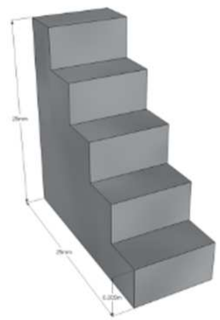


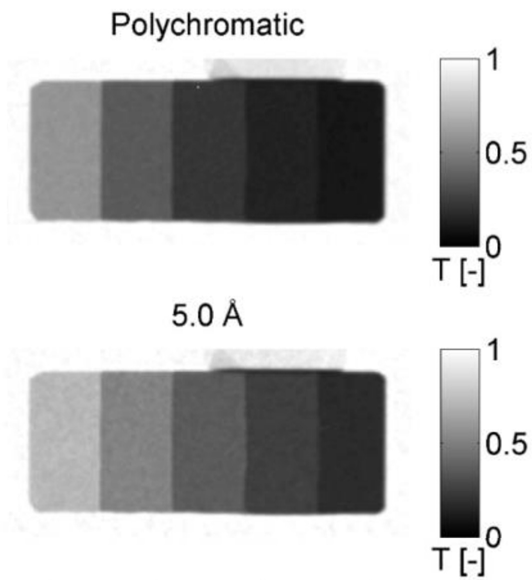
Radiograph at 4.6Å
Bulk, ~1min

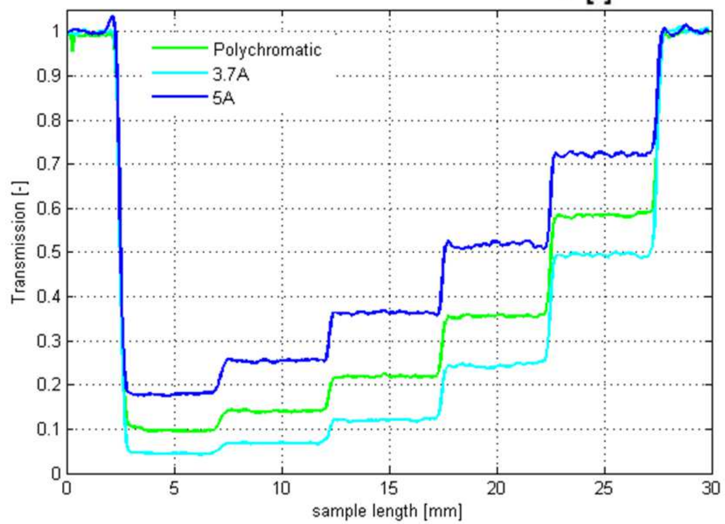
Lehmann, E.; Peetermans, S.; Josic, L.; Leber, H. & van Swygenhoven, H. Energy-selective neutron imaging with high spatial resolution and its impact on the study of crystalline-structured materials. *Nuclear Instruments and Methods in Ph… Research, Section A*, **2014**, *735*, 10…

Increased sensitivity / transmission







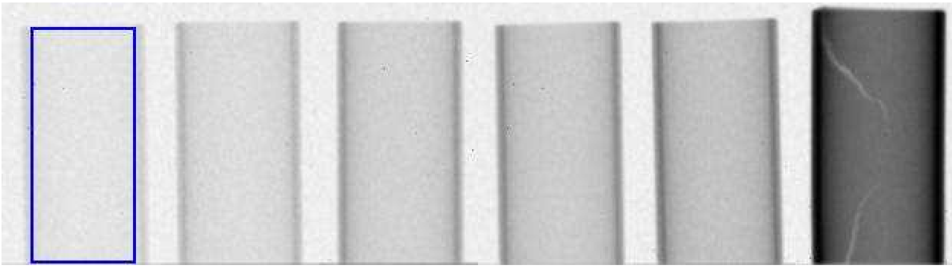


- Increased transmission just past the Bragg cut-off
- Maximum attenuation just before the Bragg cut-off: highest sensitivity to small amounts

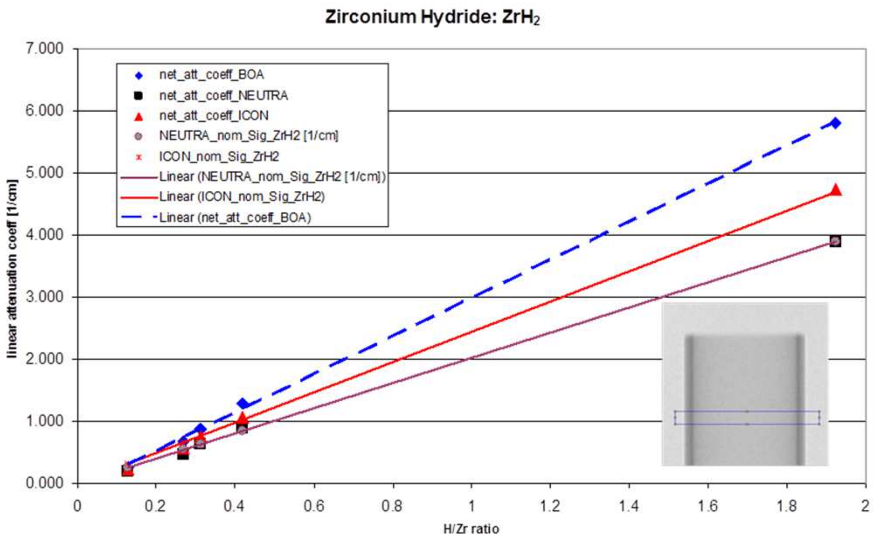
Sensitivity example

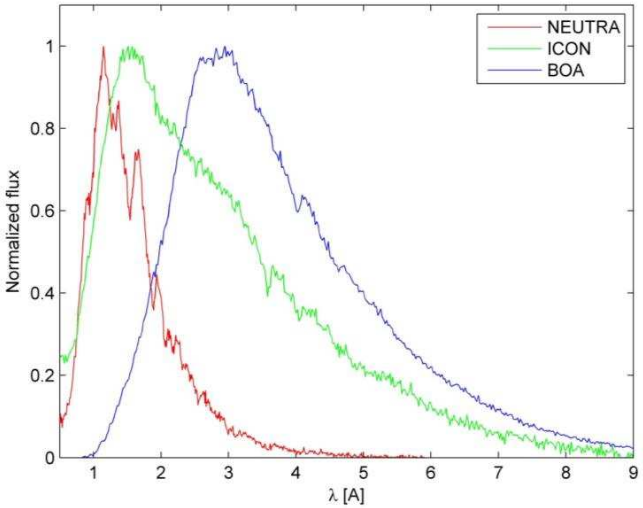
Sample	H/Zr ratio
8	0
9	0.127
10	0.268
11	0.312
12	0.418
13	1.924

- Hydrogen content in Zirconium (LOCA accidents NPP, e.g. Fukushima)
- The colder the spectrum, the more sensitive to small amounts of H: NEUTRA – ICON – BOA



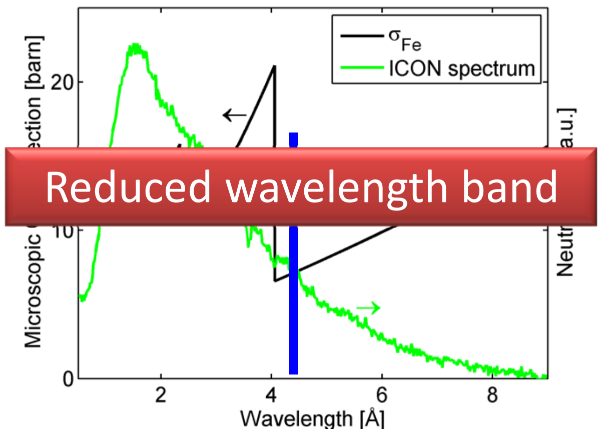
Samples: M. Grosse, KIT
Experiment: P. Vontobel, A. Kaestner, S. Peetermans, T. Panzner, PSI
Analysis: P. Vontobel, PSI



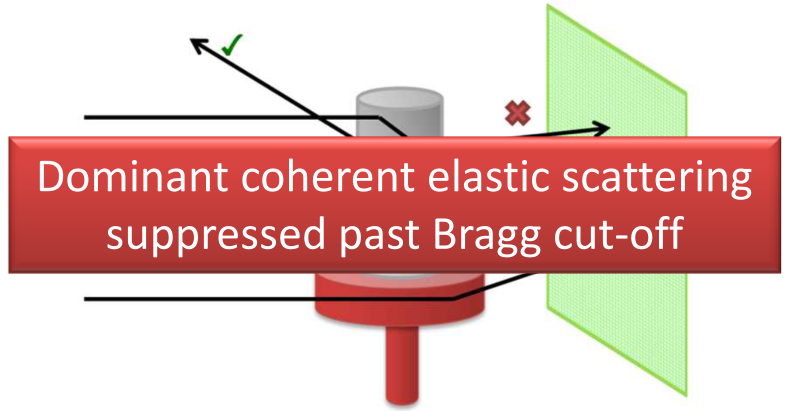


White beam problems…

Beam hardening



Scattering contributions



Short wavelengths attenuated more

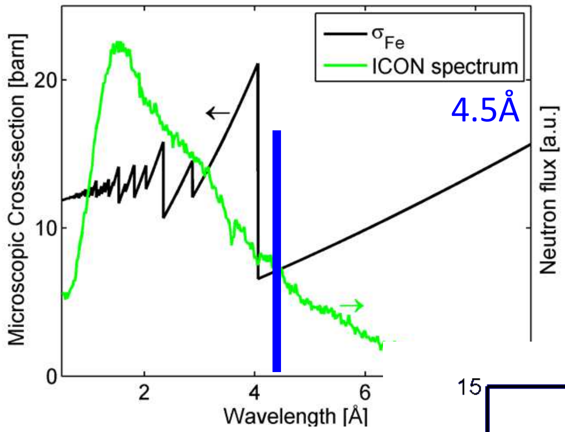
Thickness increases
→ colder beam
→ lower effective cross-section

Scattered neutrons still hit the detector behind the sample

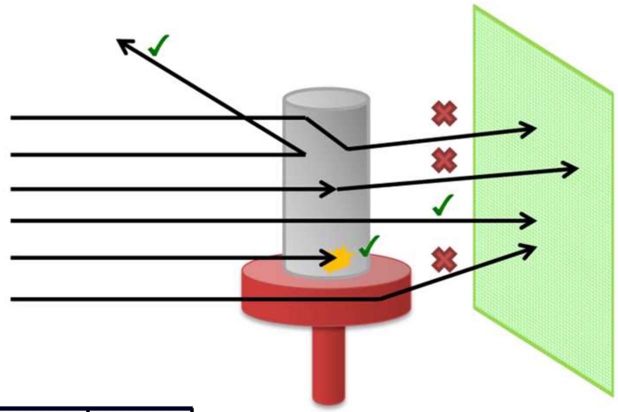
Transmission through sample overestimated
Cross-section underestimated

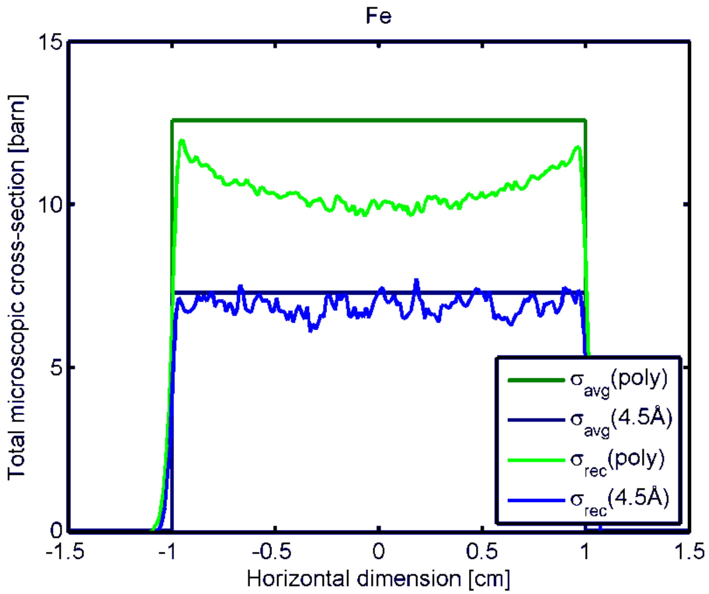
White beam problems

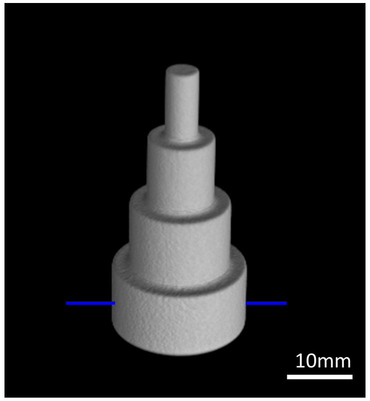
Beam hardening



Scattering contributions



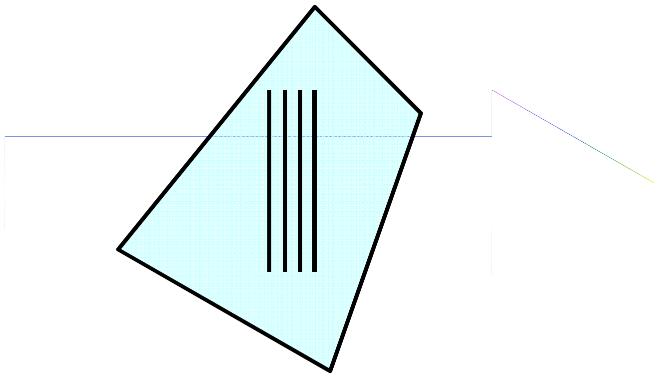


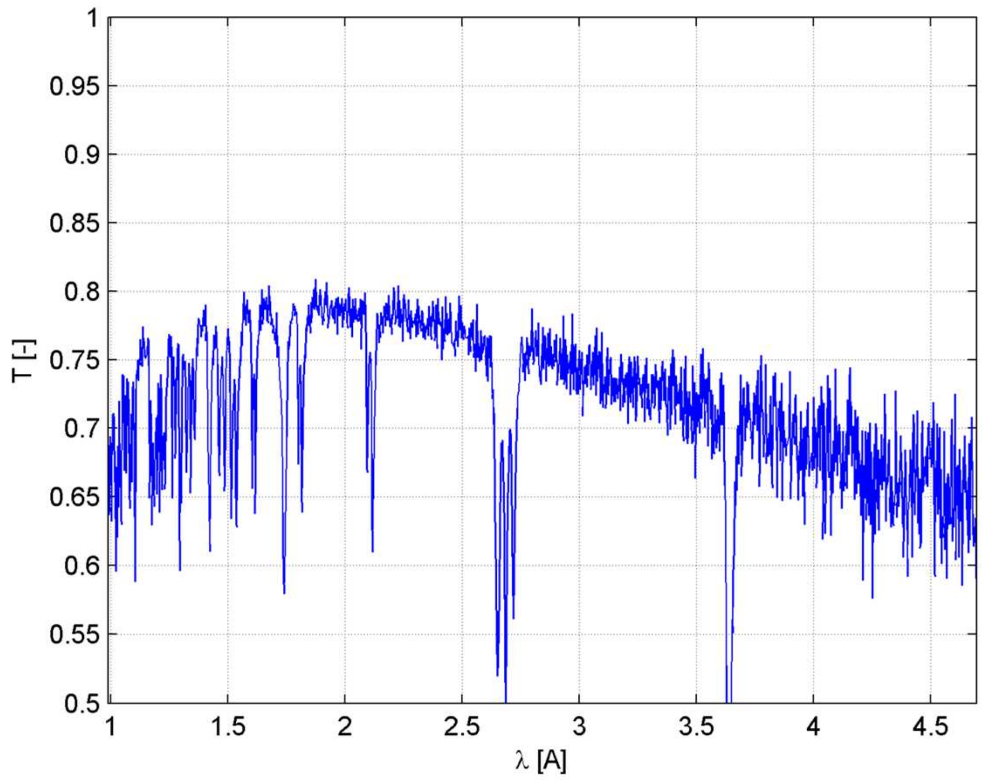


Iron step cylinder
Tomography
(polychromatic/4.5Å)
ø 20mm σ-profile

Single Crystal (SX): Bragg dips

$$2d_{hkl} \sin\theta = \lambda_{hkl}$$





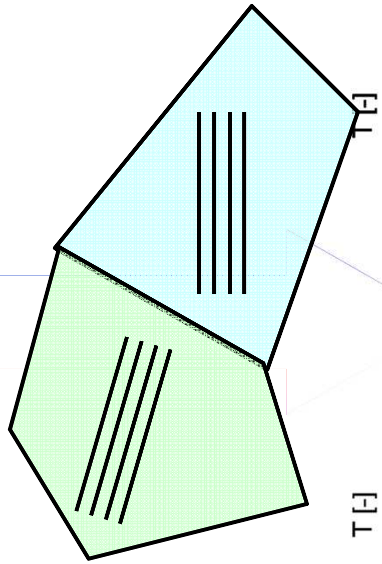
Single crystal: Bragg dips

Position: crystallographic phase,
crystal orientation

Width: crystal quality (mosaicity)

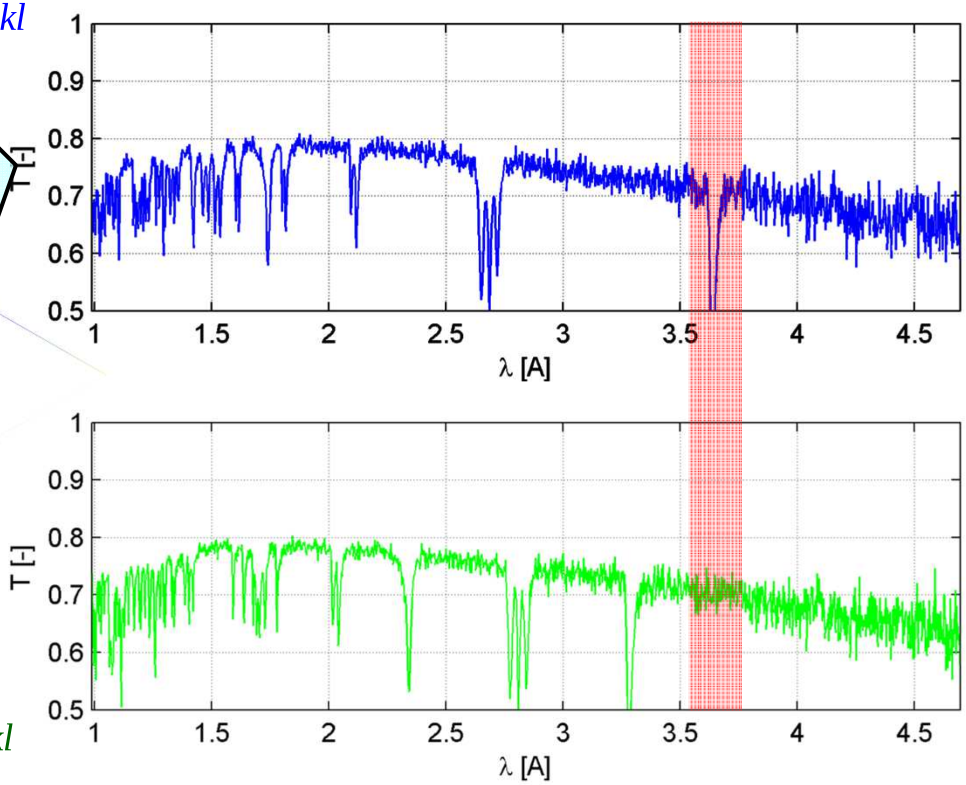
Single Crystal(lite)

$$2d_{hkl}\sin\theta_1 = \lambda_{hkl}$$



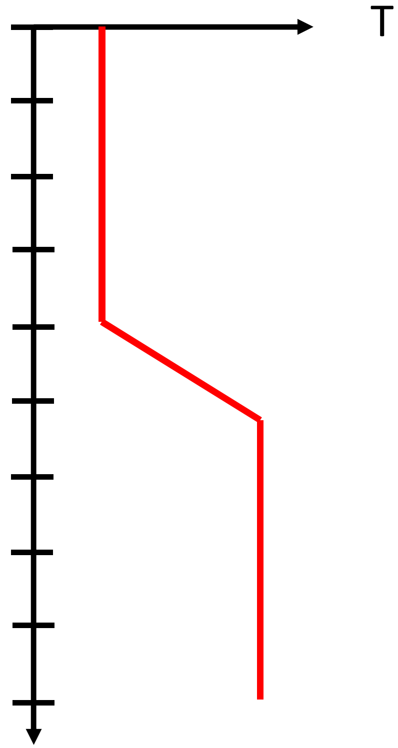
$$2d_{hkl}\sin\theta_2 = \lambda_{hkl}$$

Add 2nd crystal



Different orientation,
Different Bragg dip location

Monochromatic imaging

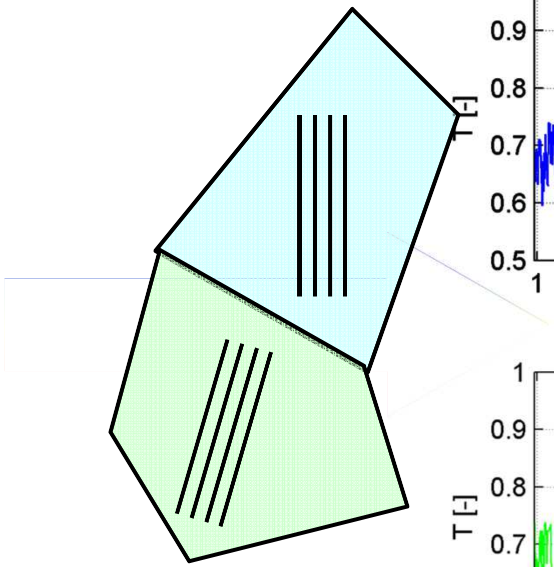


Crystallite contrast
if above spatial
resolution

PAUL SCHERRER INSTITUT

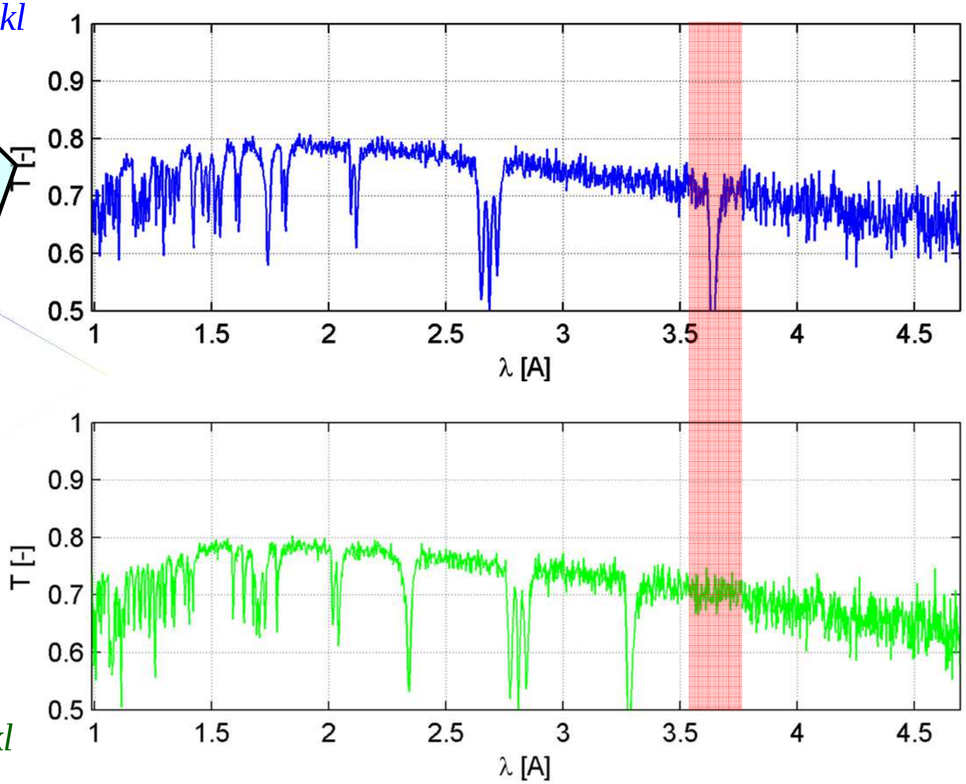
Single Crystal: Bragg dips

$$2d_{hkl} \sin \theta_1 = \lambda_{hkl}$$



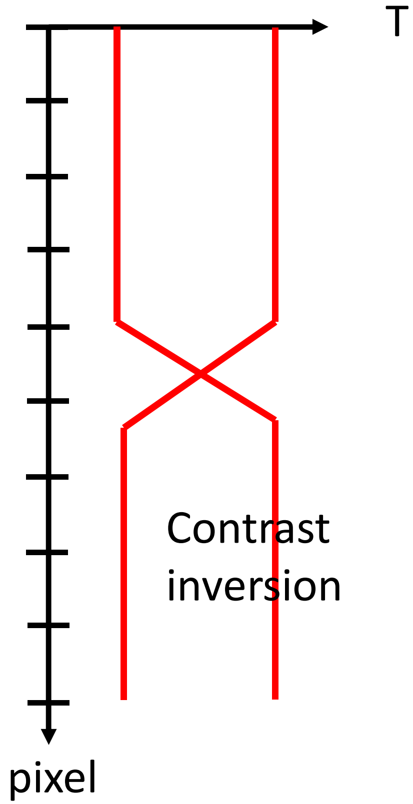
$$2d_{hkl} \sin \theta_2 = \lambda_{hkl}$$

Add 2nd crystal



Different orientation,
Different Bragg dip location

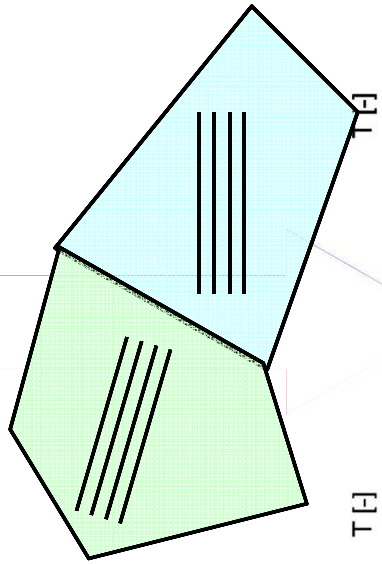
Monochromatic imaging



Crystallite contrast
if above spatial
resolution

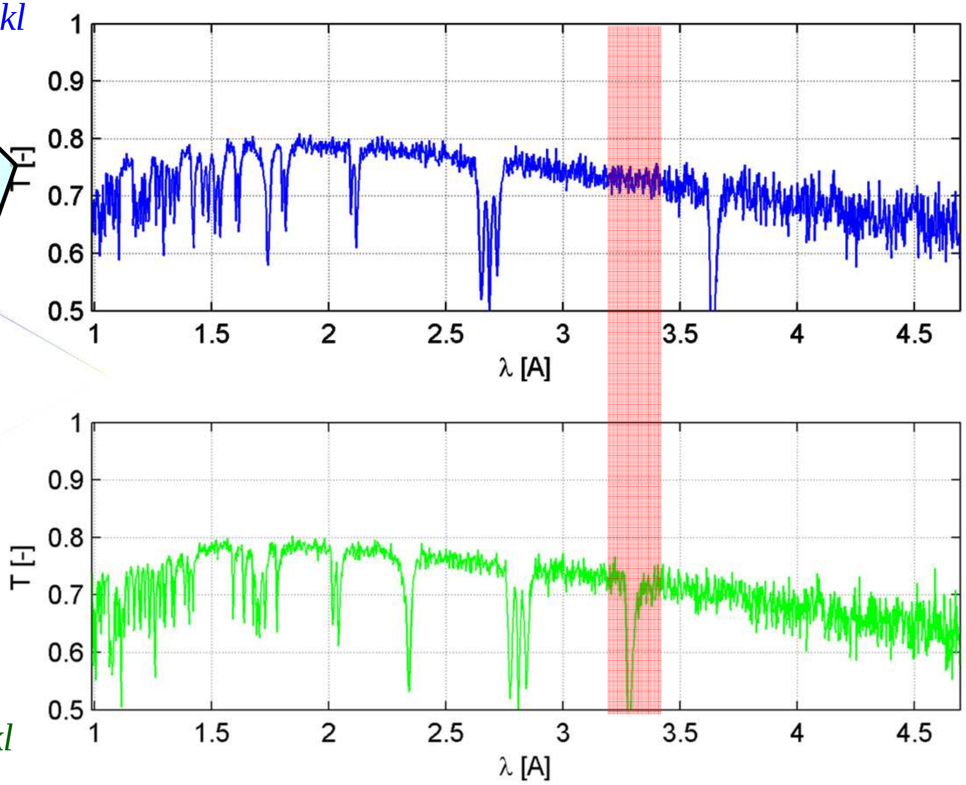
Single Crystal: Bragg dips

$$2d_{hkl}\sin\theta_1 = \lambda_{hkl}$$



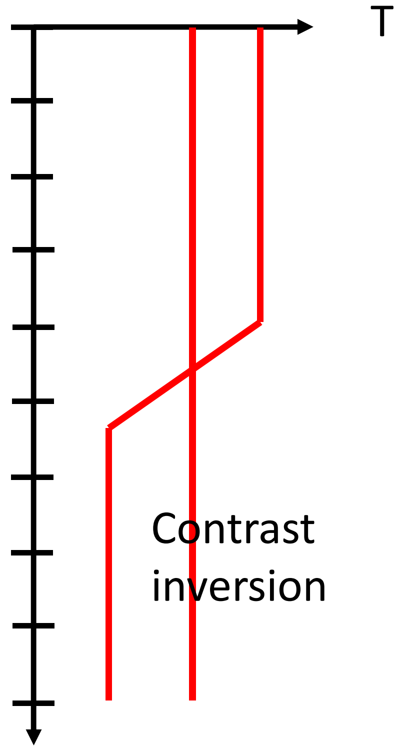
$$2d_{hkl}\sin\theta_2 = \lambda_{hkl}$$

Add 2nd crystal



Different orientation,
Different Bragg dip location

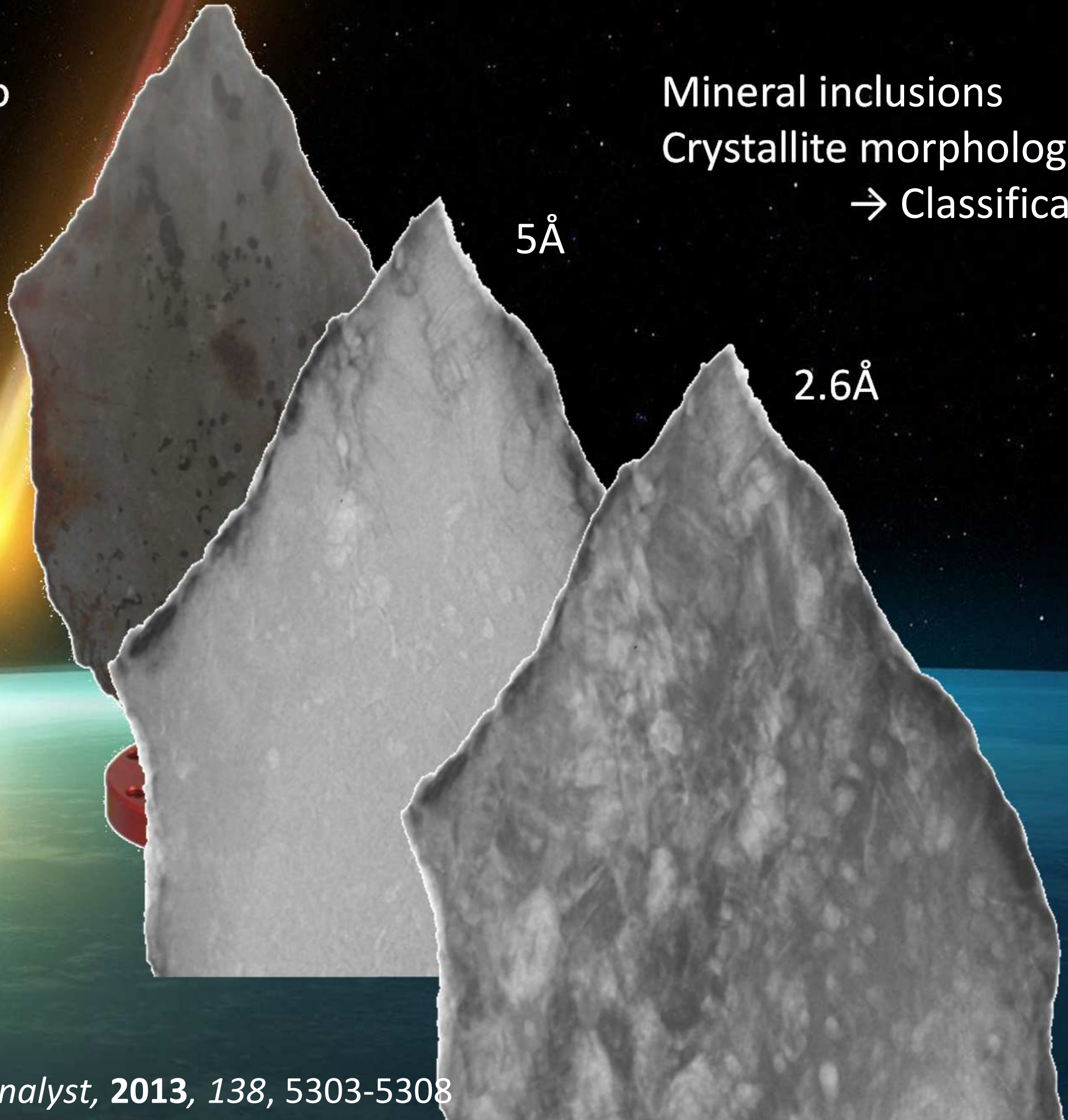
Monochromatic imaging



Crystal size contrast
$\lambda > 2d_{max}$
No diffraction contrast,
pure elemental contrast
if above spatial resolution

Example: monochromatic imaging of the Mont-Dieu meteorite

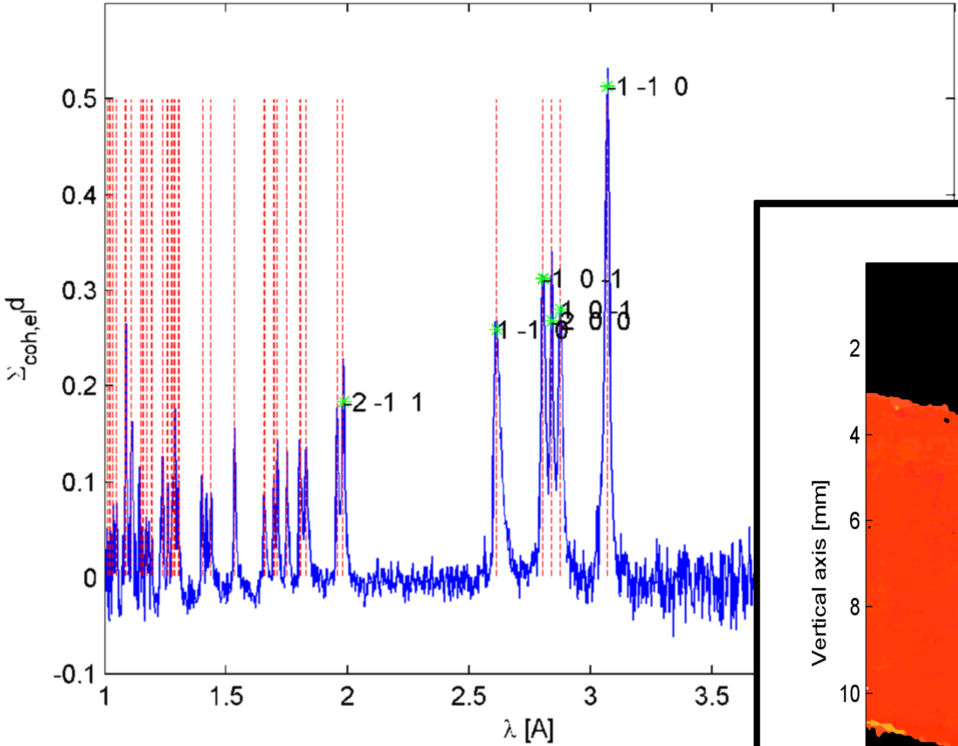
Mineral inclusions
Crystallite morphology
→ Classification of meteorites



www.voicechronicle.com

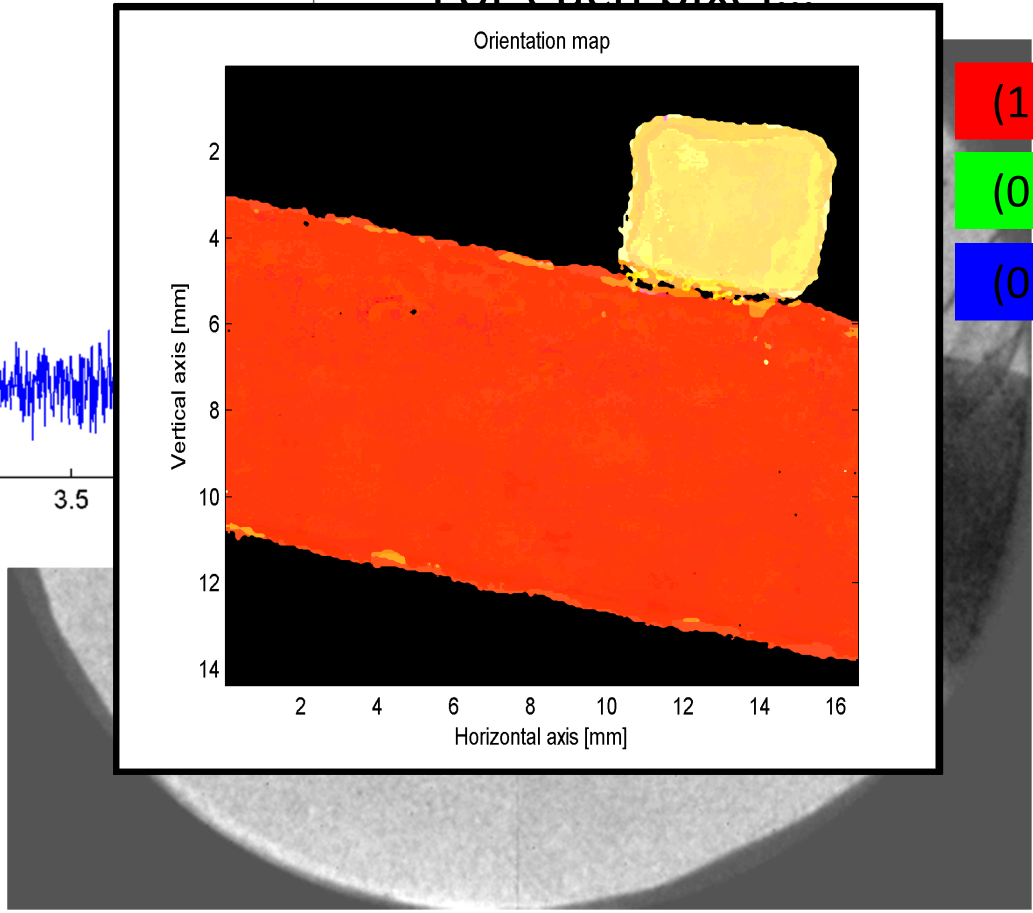
Peetermans, S. *et al.*, *Analyst,* **2013**, *138*, 5303-5308

Energy scan of single crystal



Mapping of crystal properties (orientation, phase, mosaicity) at 100µm spatial resolution

For each pixel…



(1 0 0)

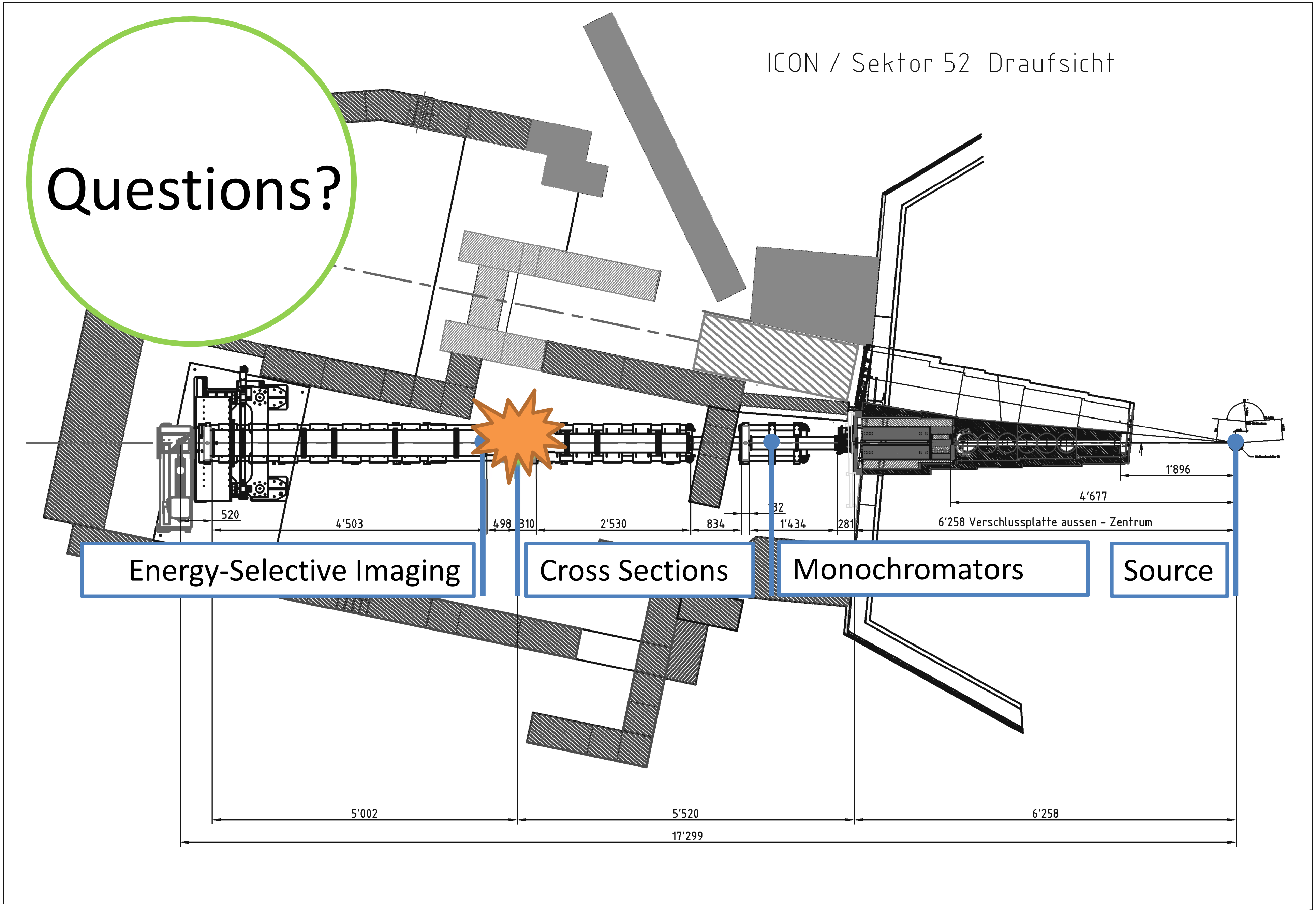
(0 1 0)

(0 0 1)

X3

(1 0 0) closely aligned to the beam

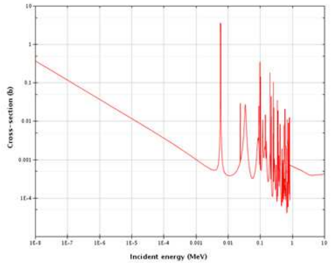
15° difference between both


Questions?
ICON / Sektor 52 Draufsicht
Energy-Selective Imaging
Cross Sections
Monochromators
Source
520
4'503
498
310
2'530
834
32
1'434
281
6'258 Verschlussplatte aussen – Zentrum
1'896
4'677
5'002
5'520
6'258
17'299

I have some questions

The absorption cross-section is 3 barn at 2.5Å, what's it at 5Å?

A. 6 barn



C. 9 barn

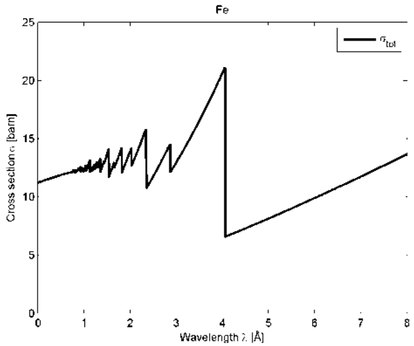
D. I'll have to look it up

$\sigma_{abs} \sim \lambda$

If I want exploit the Bragg edges to see a phase transition, I will choose a spectrum that is

A. Thermal

B. Cold



I expect the Bragg edge position to shift by 0.01Å due to strain. The best option is to use:

A. Velocity selector

B. Double Crystal Monochromator

C. Time of flight

Velocity selector	Double crystal monochromator	Time of flight
$\Delta\lambda/\lambda \approx 15\%$	$\Delta\lambda/\lambda \approx 2\text{-}5\%$	$\Delta\lambda/\lambda \approx 0.5\%$