Towards Inelastic X-Ray Scattering Spectroscopy with 0.1 meV Resolution

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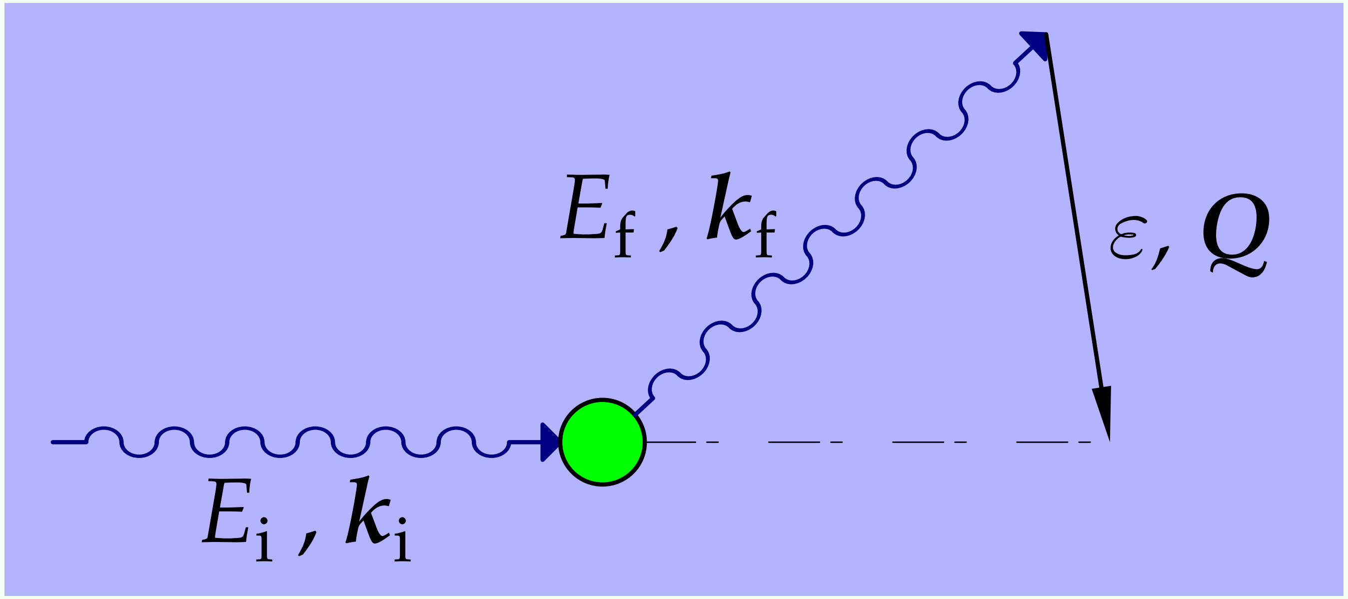
Advanced Photon Source

Content

- IXS spectroscopy: how it works now?
- Angular dispersion - as alternative monochromatization principle.
- Angular-dispersive monochromators, analyzers, IXS spectrometers.
- Results from the prototype 0.7 meV device
- Challenges in achieving 0.1 meV resolution
- Proposed R+D plan

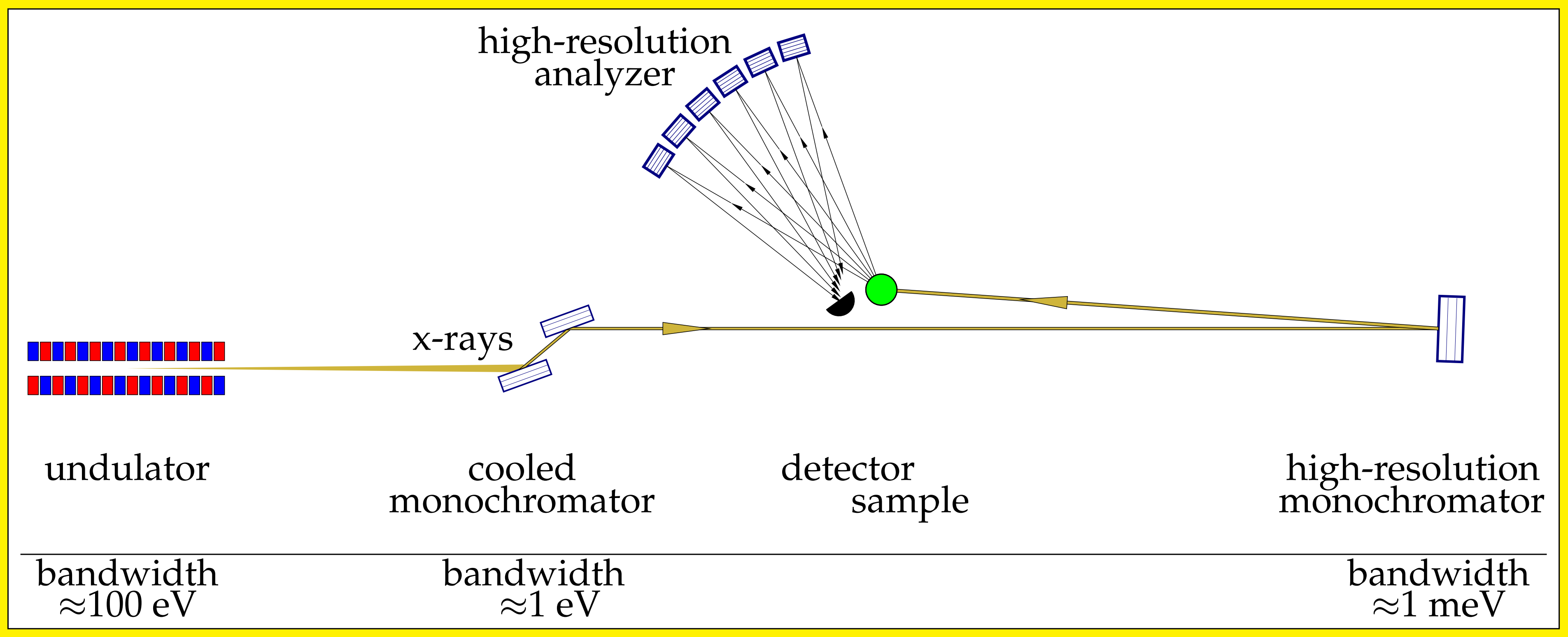
IXS Spectroscopy with Very High Resolution

IXS spectroscopy with very high resolution ($\lesssim 1$ meV) is one of the major techniques for studying vibrational dynamics in condensed matter.



X-ray monochromators and analyzers with meV- and sub-meV-resolution are the main optical components of the spectrometers.

Modern IXS Spectrometer (layout)



Kohra, Matsushita (1972)

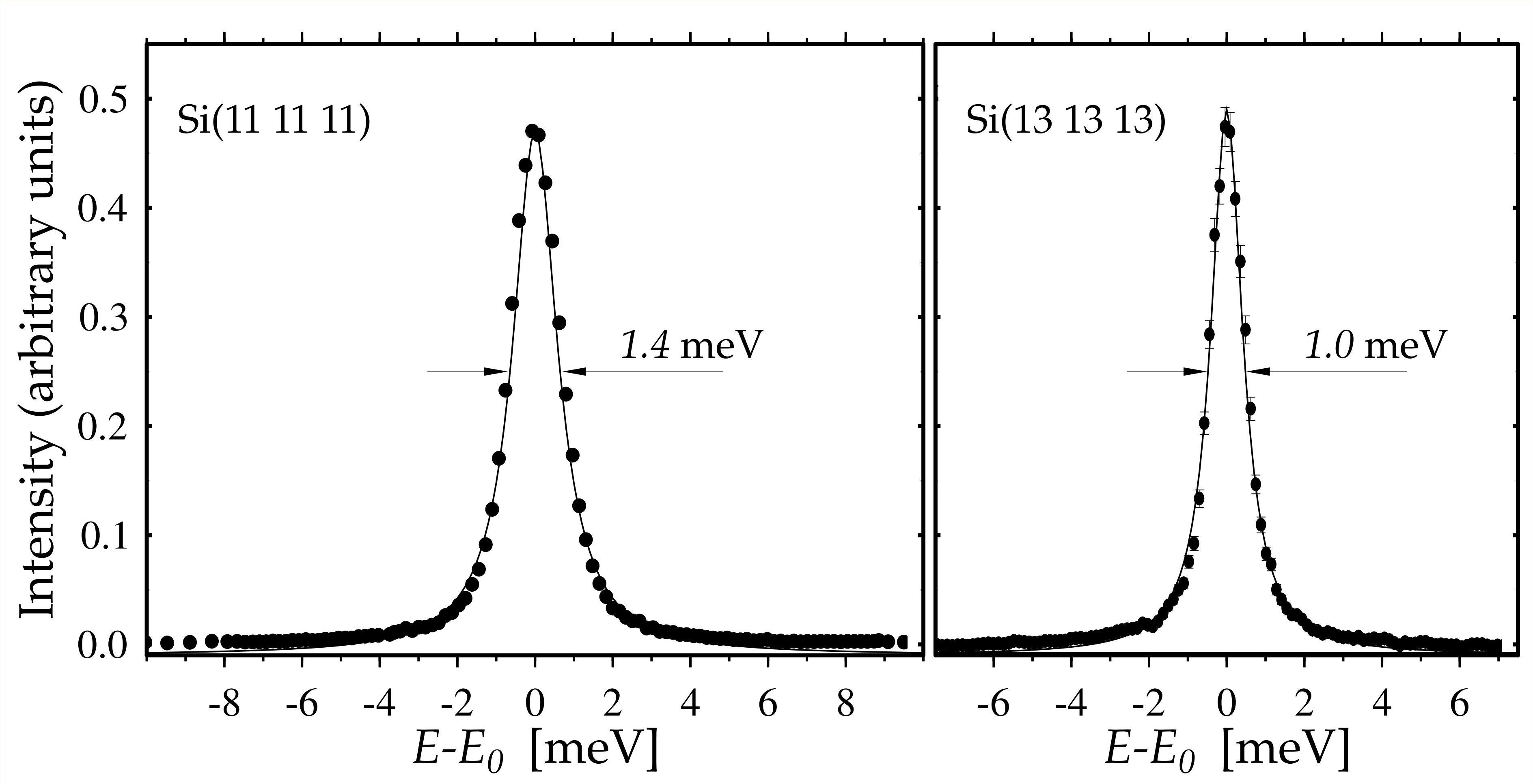
Graef, Materlik (1982)

Burkel, Dorner, Peisl (1987)

Sette, Krisch, et al. @ESRF **Sinn, Alp, et al. @APS** **Baron, et al. @SPring-8**

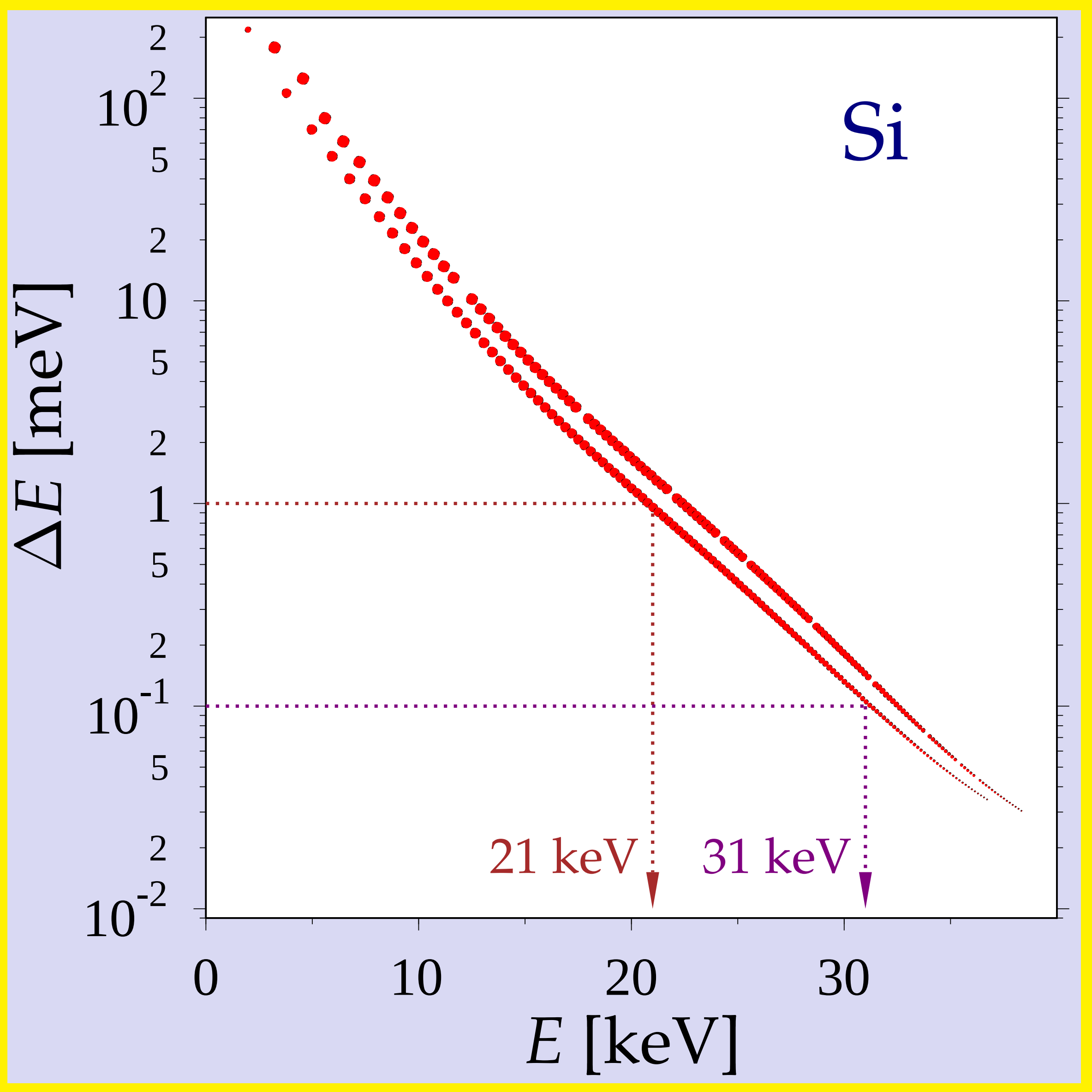
Resolution of the IXS spectrometers & count-rates,

$\Delta\varepsilon \geq 1$ meV **$\Delta Q \approx 0.5$ nm^{-1}** **Count-rate $\lesssim 1$ Hz**



Courtesy of M. Krisch (ESRF)

ΔE of Bragg back reflections in Si



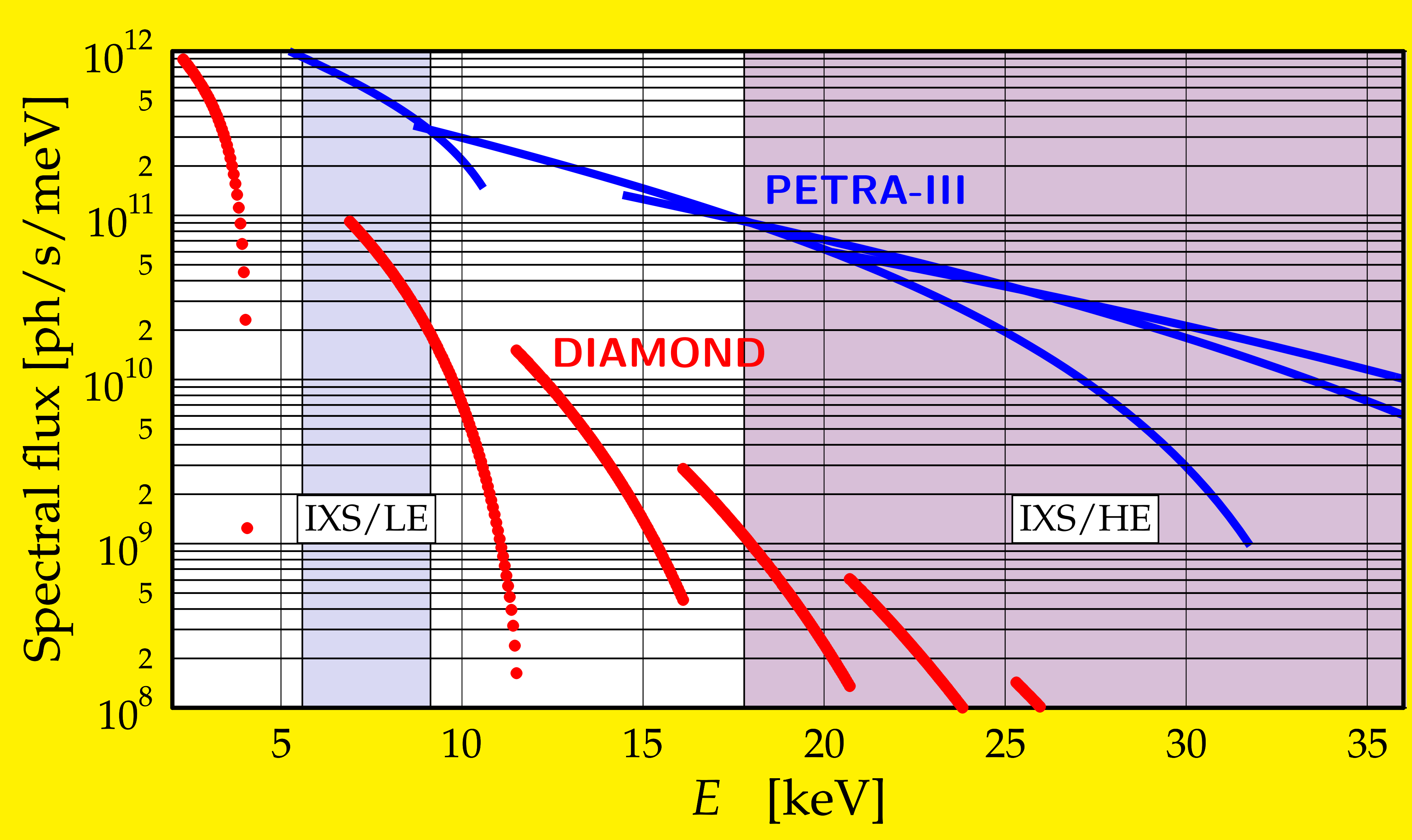
ΔE = spectral width of the Bragg backreflection

E = photon energy in Bragg backreflection

The smaller ΔE is required, the higher indexed Bragg reflection at higher photon energy E has to be used (unfortunately!).

$\Delta E = 0.1$ meV requires $E = 31$ keV

Undulator spectrum



Low-energy photons would be better:

- **Higher count-rates (more photons in the low-energy range).**
- **IXS applicable at low- and intermediate energy SR facilities.**
- **Better momentum resolution ΔQ for the same solid acceptance angle $\Upsilon \times \Upsilon$:**

  $\Delta Q = \Upsilon K$. $\quad K = E/c$.

- **Proximity to K-absorption edges of the important transition metals.**

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- Proximity to K-absorption edges of the important transition metals.

... but ...

Employing low-energy photons is in conflict with the principles underlying single-bounce backscattering monochromators and analyzers.

New concepts, new solutions are required:

Problem:

Spectral width ΔE of the low-indexed Bragg reflections is too large. Typically $\Delta E > 20$ meV.

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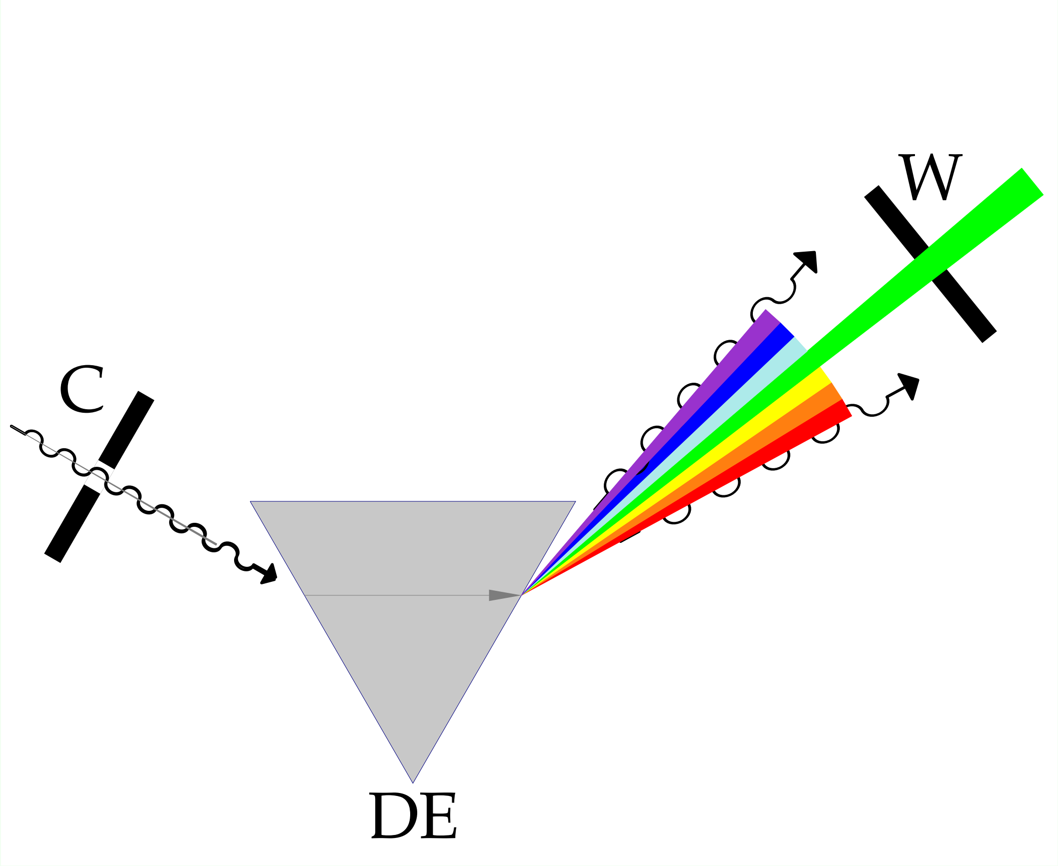
Solution:

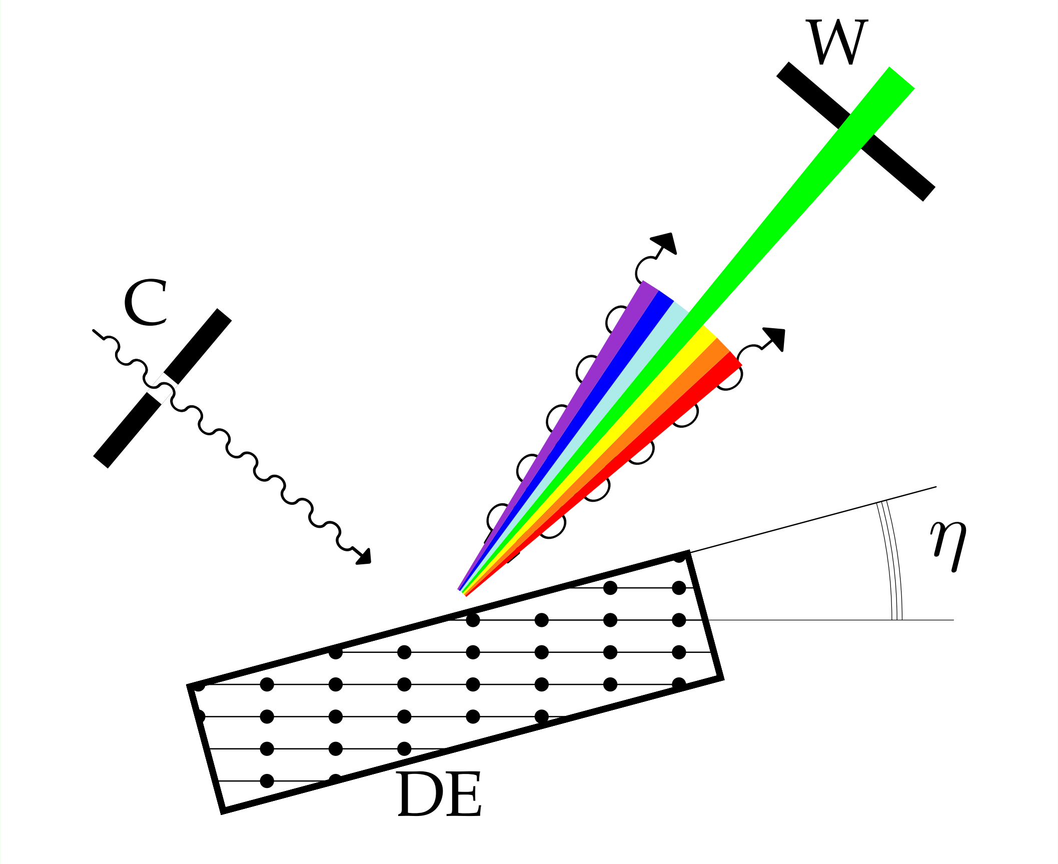
Use a small fraction of it!

New concept illustrated with optical prism



D - dispersing element C - collimator W - wavelength selector

New concept



D - dispersing element C - collimator W - wavelength selector

An asymmetrically cut crystal behaves like the optical prism dispersing the photons with different photon energies: effect of angular dispersion.

Effect of angular dispersion (1)

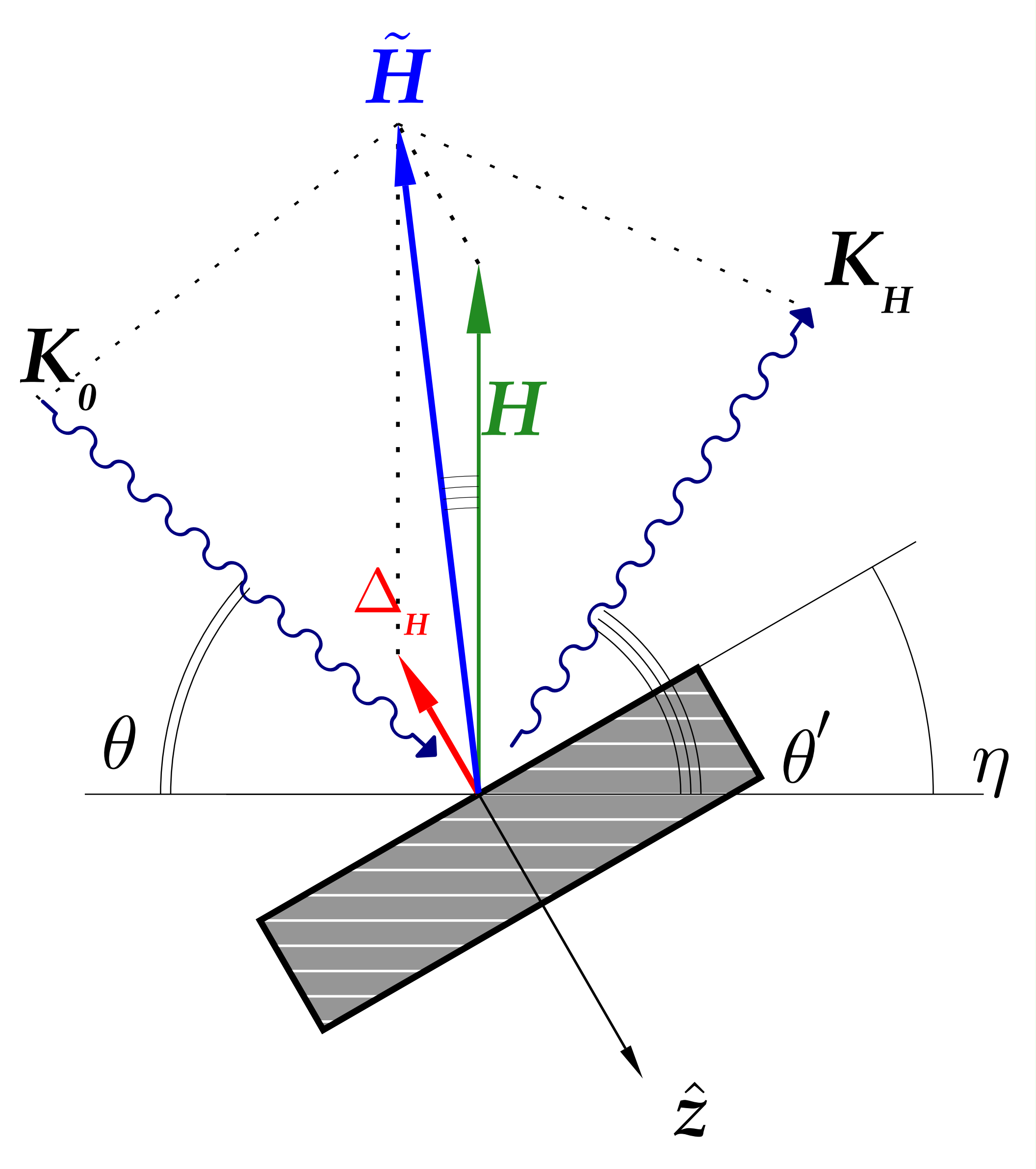
$$\boldsymbol{K}_H = \boldsymbol{K}_0 + \tilde{\boldsymbol{H}}$$

$$\tilde{\boldsymbol{H}} = \boldsymbol{H} + \boldsymbol{\Delta}_H$$

$$\boldsymbol{\Delta}_H = K\,\frac{\alpha}{\sin(\theta-\eta)}\,\hat{\boldsymbol{z}}$$

$$\alpha \propto 1 - n$$

n – refractive index



Effect of angular dispersion (2)

$$\boldsymbol{K}_H = \boldsymbol{K}_0 + \tilde{\boldsymbol{H}}$$

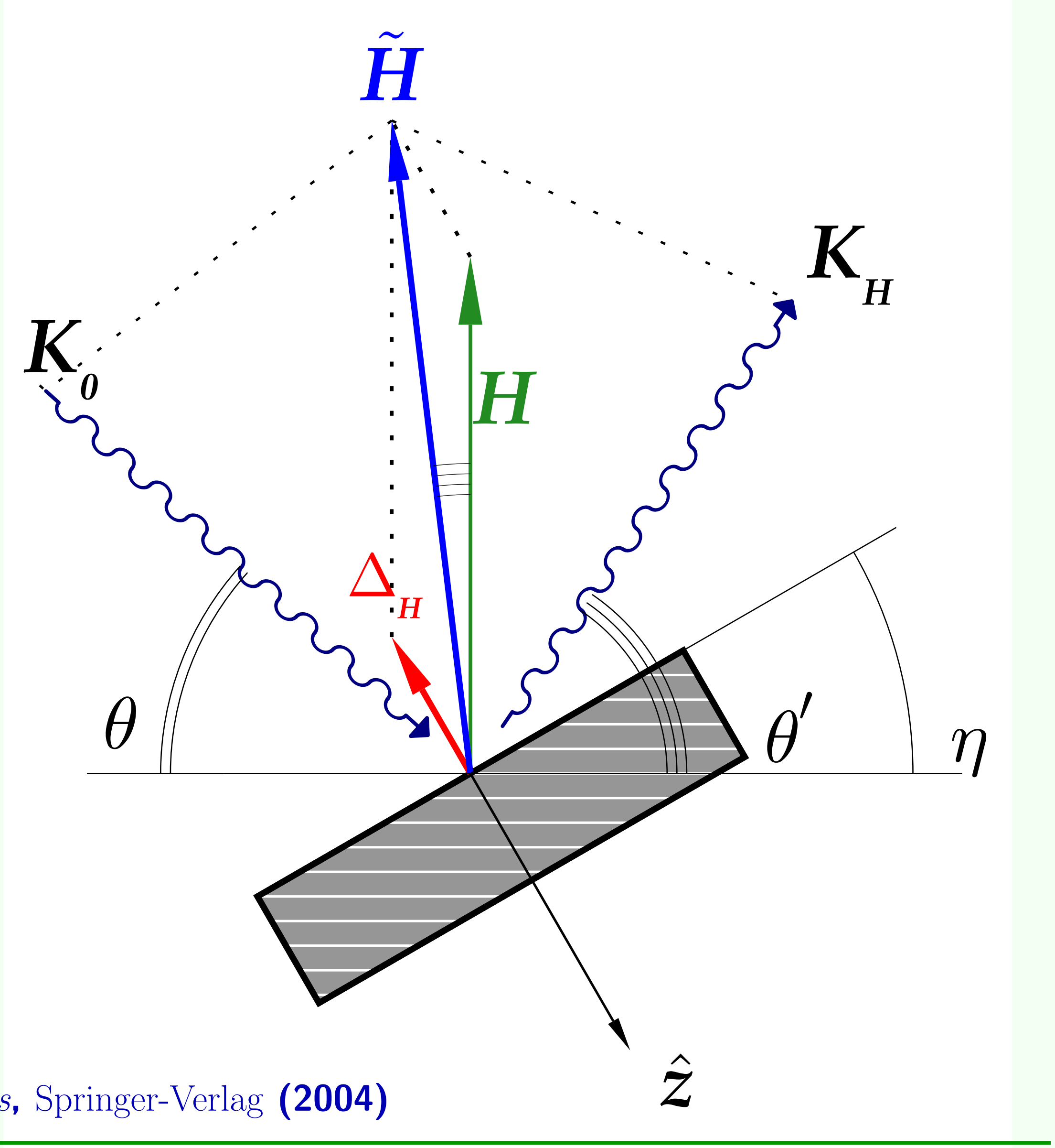
$$\tilde{\boldsymbol{H}} = \boldsymbol{H} + \boldsymbol{\Delta}_H$$

$$\boldsymbol{\Delta}_H = K \frac{\alpha}{\sin(\theta - \eta)} \hat{\boldsymbol{z}}$$

$\theta < \pi/2$

$$\delta\theta' = -\frac{\delta E}{E}(1+b)\tan\theta$$

$$b = -\frac{\sin(\theta-\eta)}{\sin(\theta+\eta)}$$



Yu. Shvyd'ko, *X-Ray Optics*, Springer-Verlag **(2004)**

Effect of angular dispersion (3)

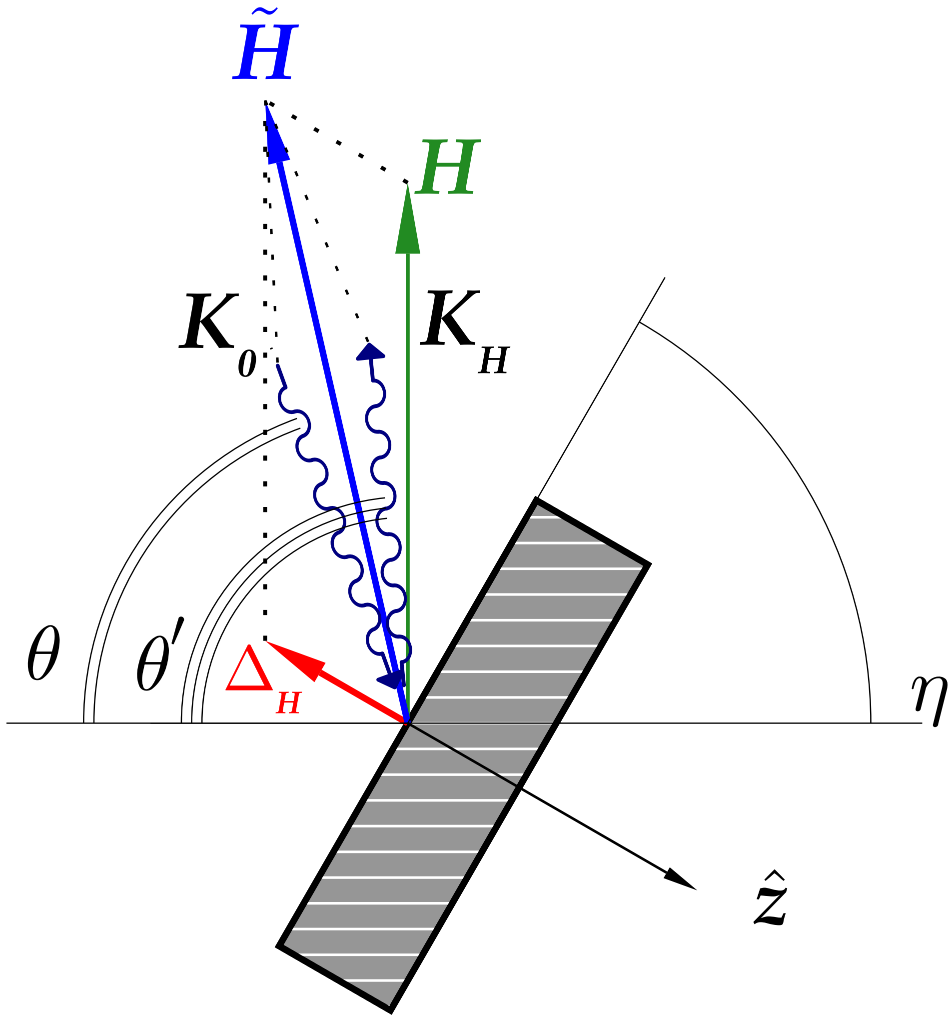
$$\boldsymbol{K}_{H} = \boldsymbol{K}_{0} + \tilde{\boldsymbol{H}}$$

$$\tilde{\boldsymbol{H}} = \boldsymbol{H} + \boldsymbol{\Delta}_{H}$$

$$\boldsymbol{\Delta}_{H} = K \frac{\alpha}{\sin(\theta - \eta)} \hat{\boldsymbol{z}}$$

$\theta \simeq \pi/2$

$$\delta\theta' = \frac{\delta E}{E} (2 \tan \eta)$$

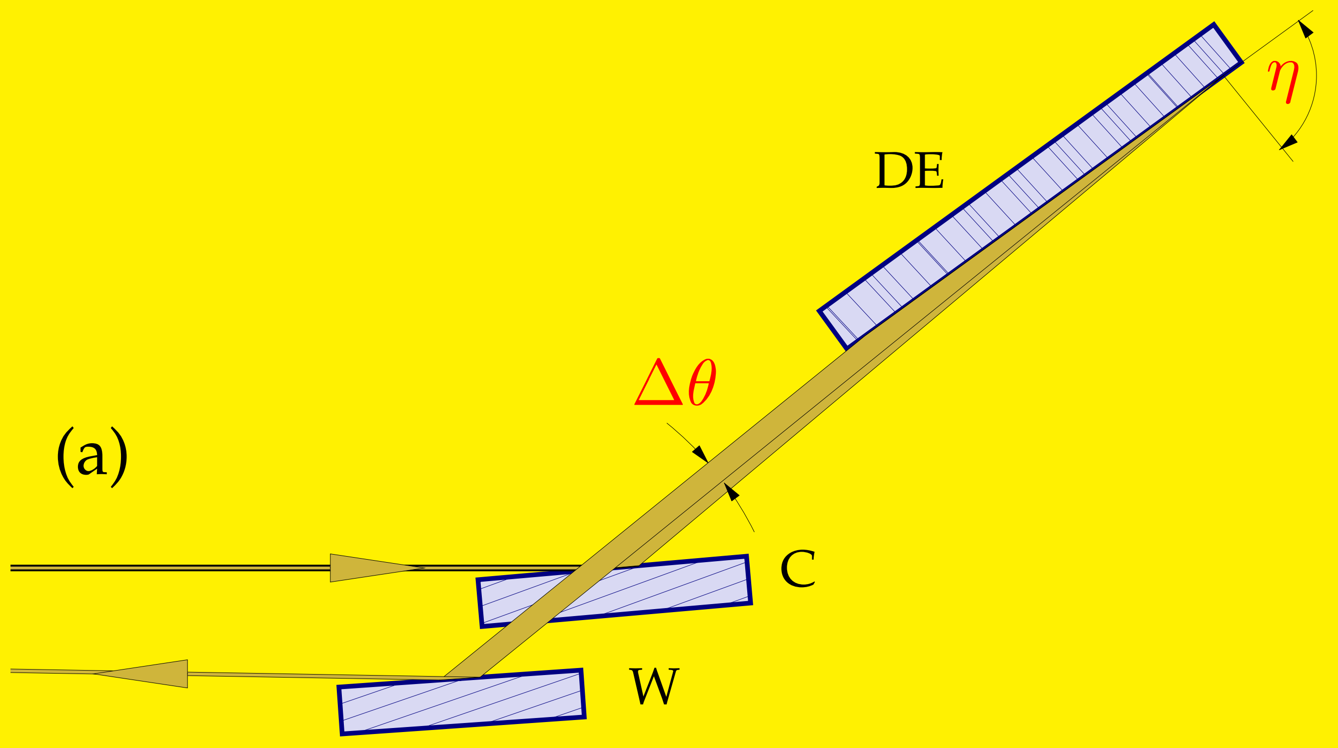


AND: Exact Backscattering is NOT at normal incidence!

CDW-Monochromator and its Spectral Resolution

$$\frac{\Delta E}{E} = \frac{\Delta\theta}{\tan\eta}$$

Yu. Shvyd'ko
X-Ray Optics, Springer-Verlag **(2004)**



The smaller the photon energy E,

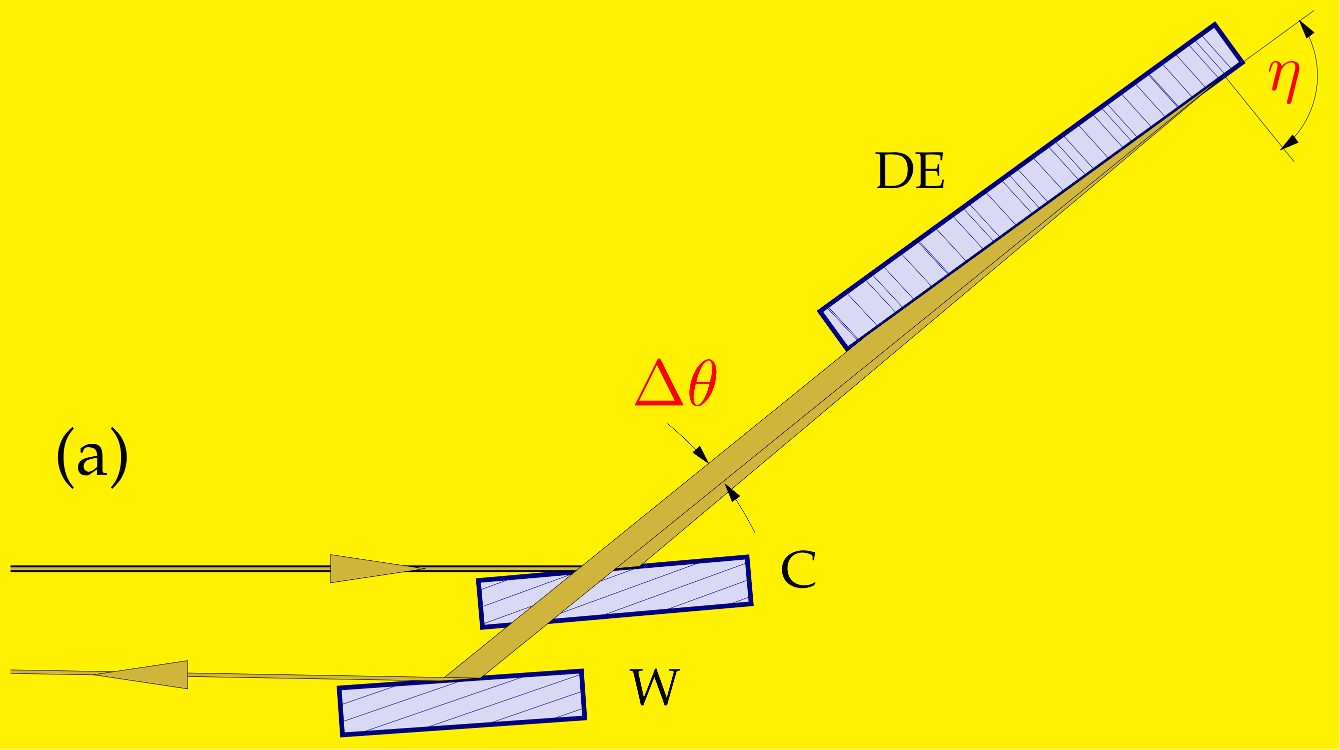
the smaller is the energy bandwidth ΔE (fortunately!).

Spectral resolution of the CDW-monochromator

$$\frac{\Delta E}{E} = \frac{\Delta\theta}{\tan\eta}$$

$$\frac{\Delta E}{E} = 10^{-6} - 10^{-8}$$

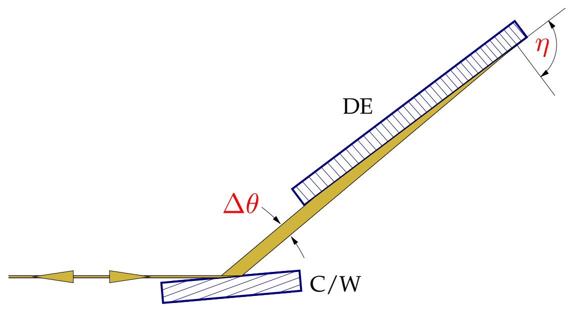
is feasible

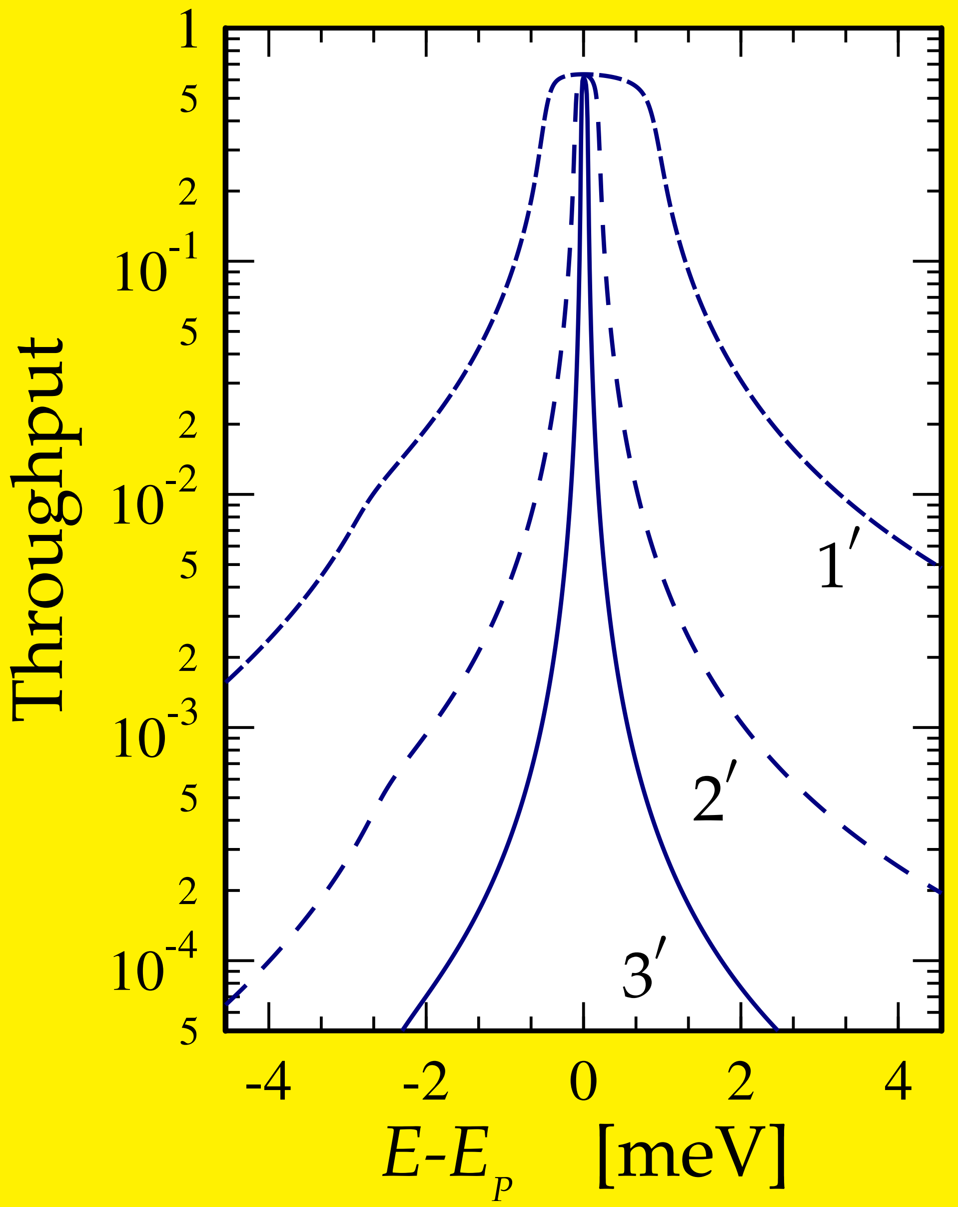


$$E = 10 \text{ keV} \Rightarrow \quad \Delta E = 10 - 0.1 \text{ meV}$$

$$E = 5 \text{ keV} \Rightarrow \quad \Delta E = 5 - 0.05 \text{ meV}$$

Throughput of the CDW-monochromator



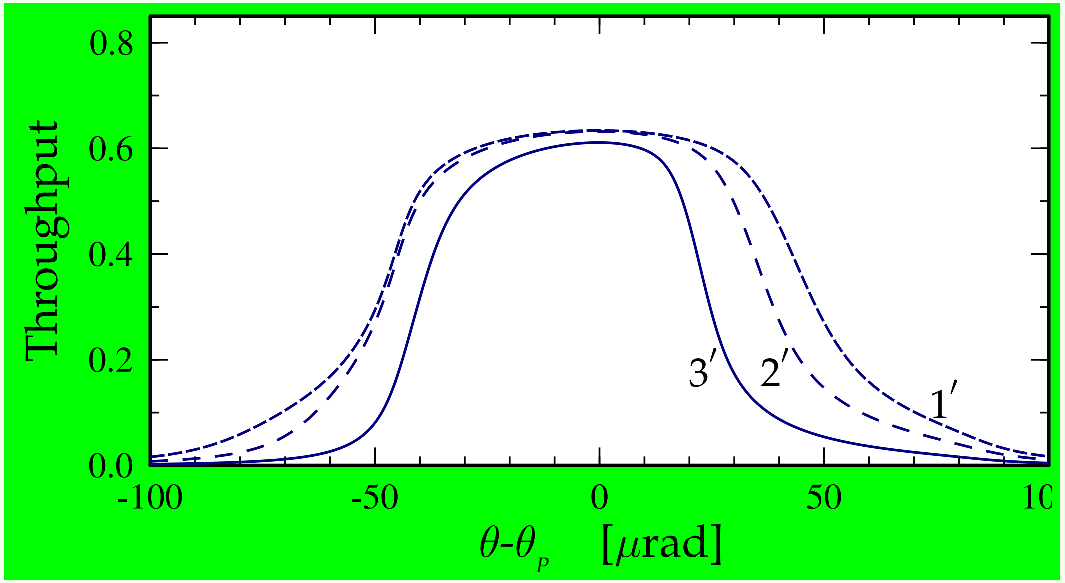


$E = 9.1$ **keV**

$1' : \Delta E = 1.5$ **meV** $(\eta = 85°)$

$2' : \Delta E = 0.3$ **meV** $(\eta = 89°)$

$3' : \Delta E = 0.09$ **meV** $(\eta = 89.6°)$

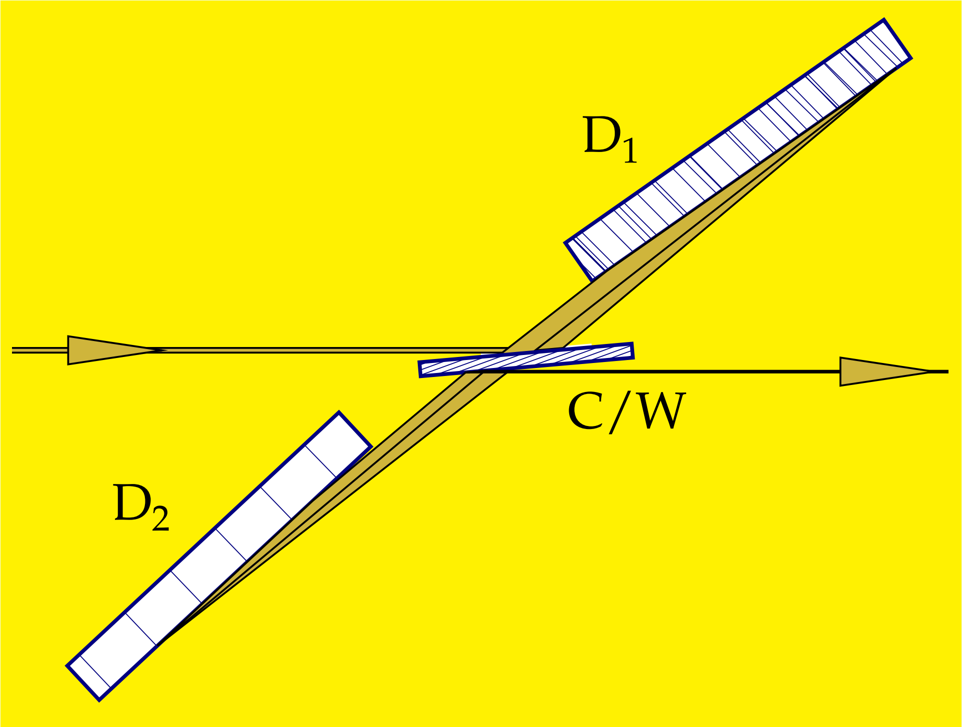


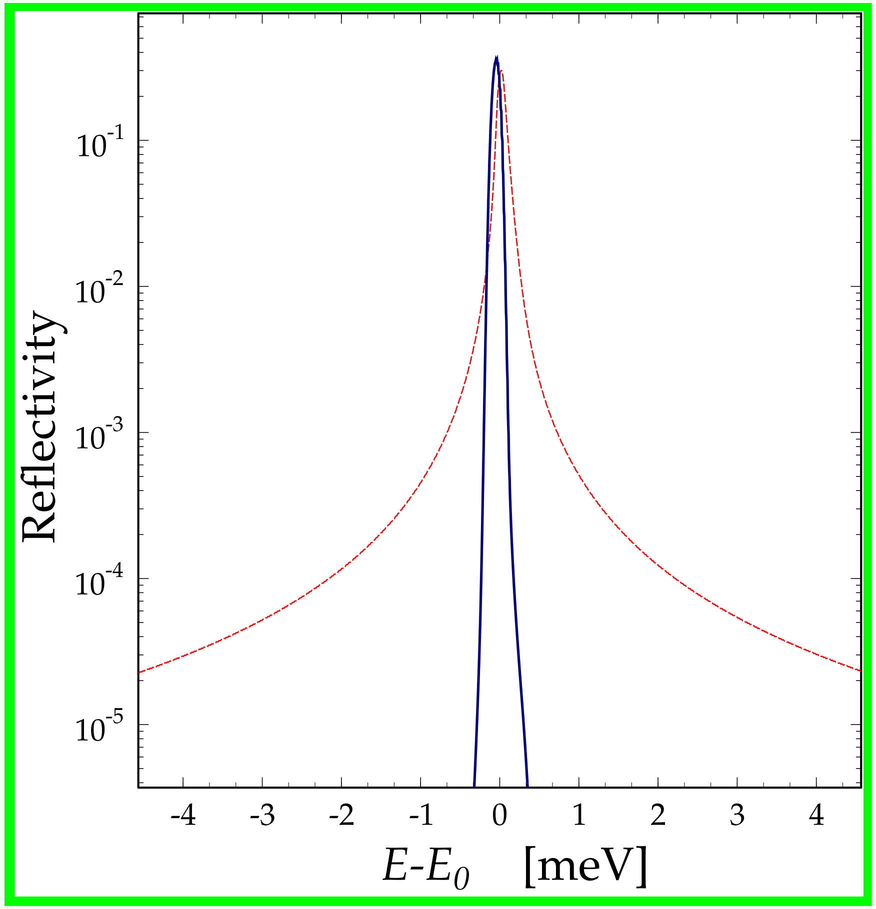
Angular-Dispersive In-line CDDW-monochromator

The angular dispersion is enhenced by a factor of 2:

Smaller asymmetry angle!

Shorter dispersing elements D_1, D_2!

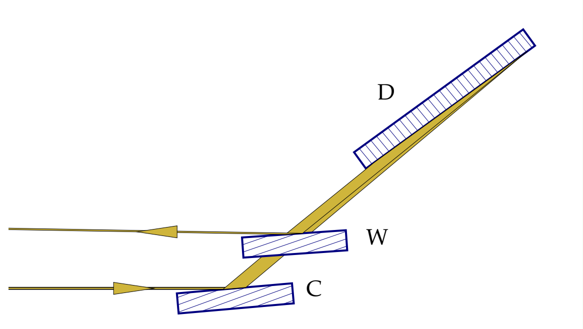




Angular-dispersive CDDW monochromator:
$E = 9.1315$ **keV,** $\Delta E = 0.1$ **meV, D=Si(008)**

Single-bounce backscattering monochromator:
$E = 31.02$ **keV,** $\Delta E = 0.1$ **meV, Si(1 3 27).**

Features of the CDW-monochromators



1. $\Delta E/E$ is independent of E or of Bragg reflection.

2. The smaller the photon energy E the smaller is the bandpass ΔE.

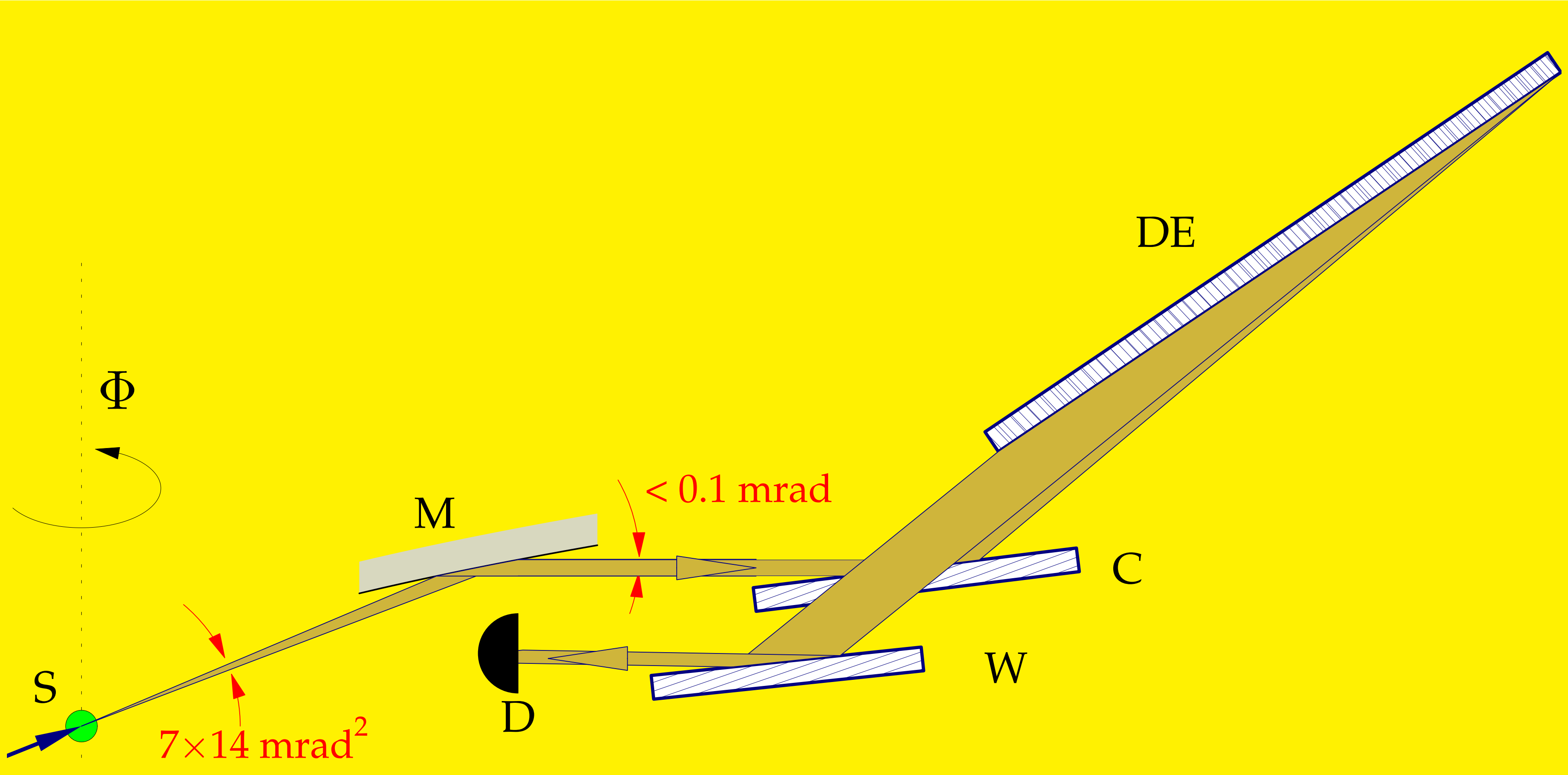
3. ΔE can be varied by changing η (E is fixed).

4. The peak throughput T and the angular acceptance $\Delta\theta$ are almost constant (while changing η).

5. Steep wings in the spectral function.

6. The temperature control and energy tuning is technically not demanding (for x-ray photons in the low-energy region $5 - 10$ keV).

CDW-Analyzer



M - collimating KB graded-multilayer mirrors

D - dispersing element

D - detector

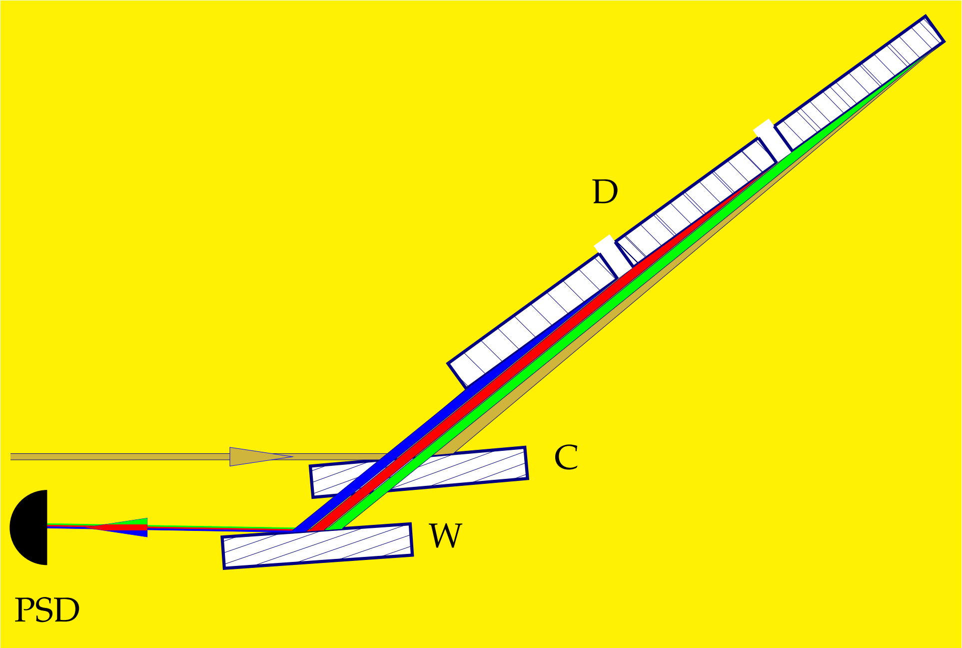
S - sample

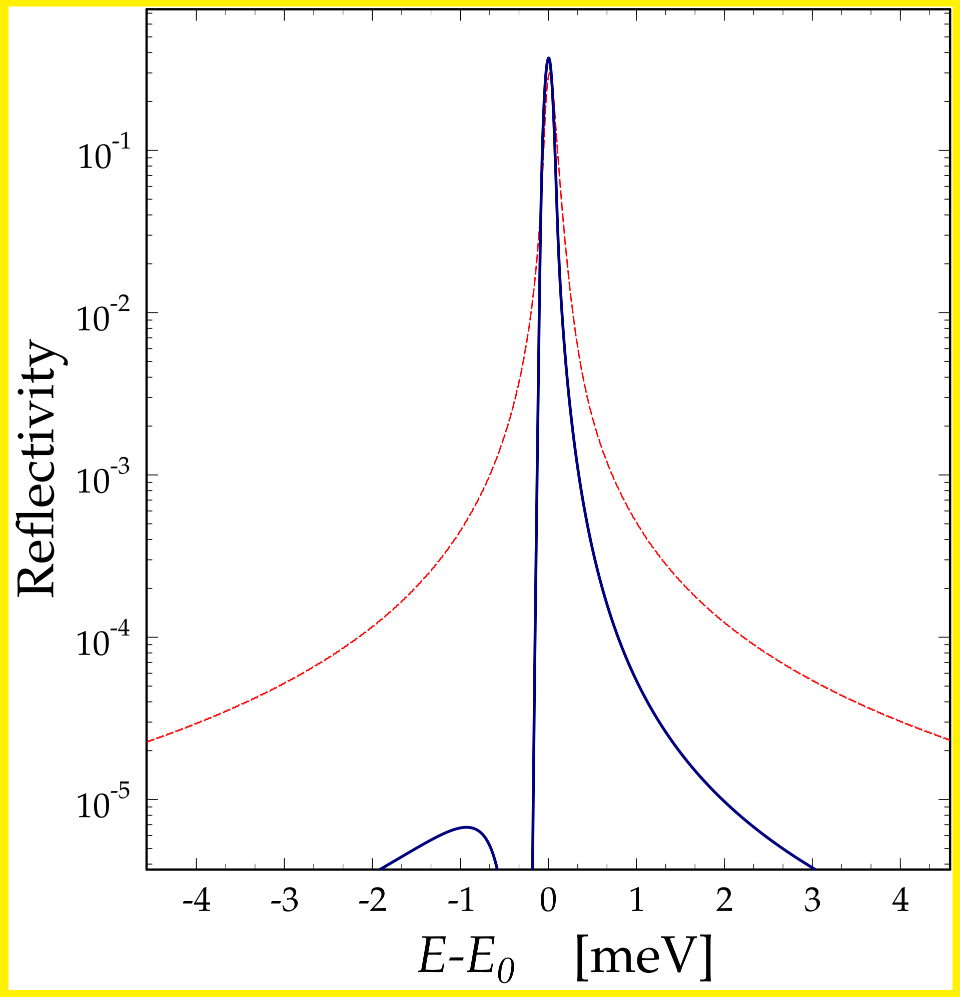
W - wavelength selector

C - collimator

CDW-Segmented Analyzer

To overcome technical problems, associated with the big length (1-2 m), the dispersing element can be built of several independent segments which need not be perfectly aligned or have precisely the same temperature.





PSD - position sensitive detector
D - dispersing elements
W - wavelength selector
C - collimator

Angular-dispersive CDW analyzer:
$E = 9.1315$ **keV,** $\Delta E = 0.1$ **meV, D=Si(008)**

Single-bounce backscattering analyzer:
$E = 31.02$ **keV,** $\Delta E = 0.1$ **meV, Si(1 3 27).**

Initial Experimental Results

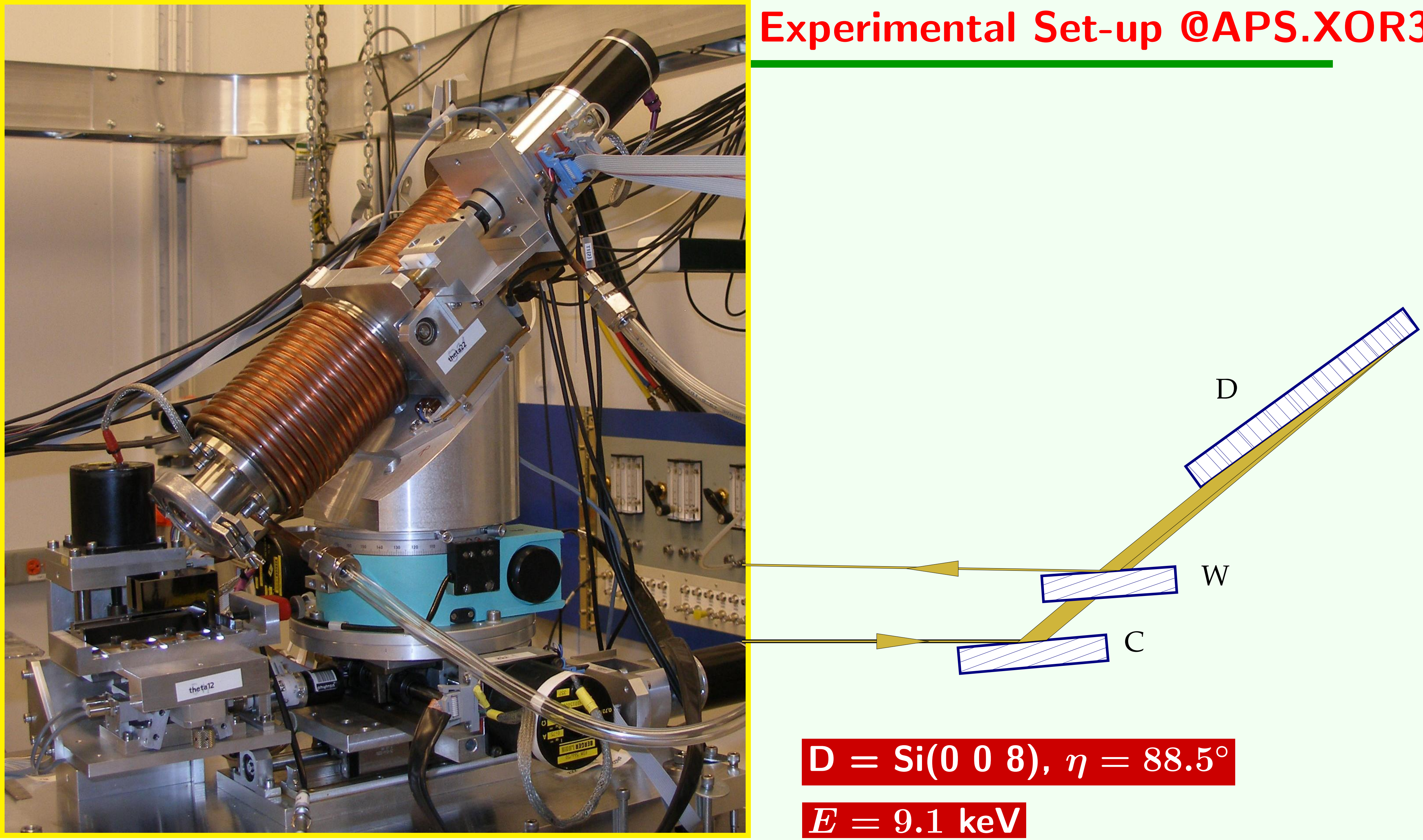
The main questions to be addressed:

can we observe the effect of angular dispersion?

and

can we demonstrate a monochromator based on this principle?

Experimental Set-up @APS.XOR3



D = Si(0 0 8), $\eta = 88.5°$

$E = 9.1$ keV

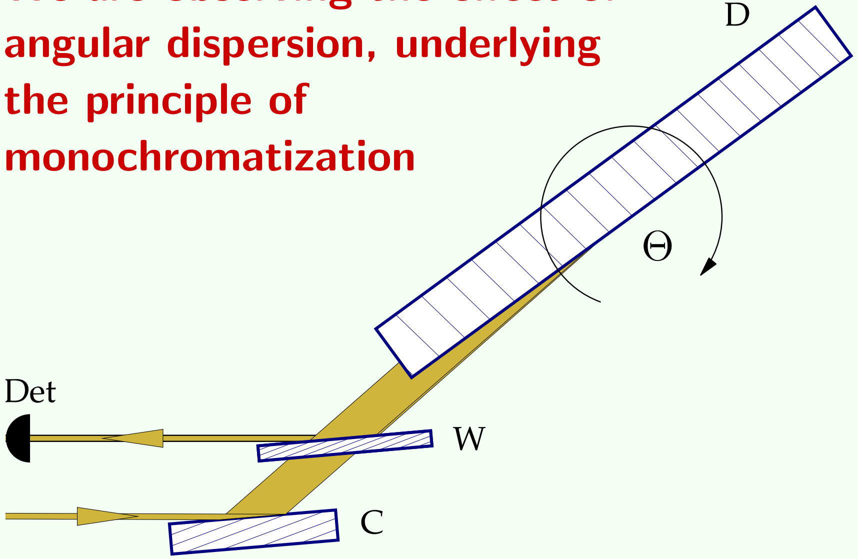
Bragg Backscattering from Asymmetrically Cut Crystal

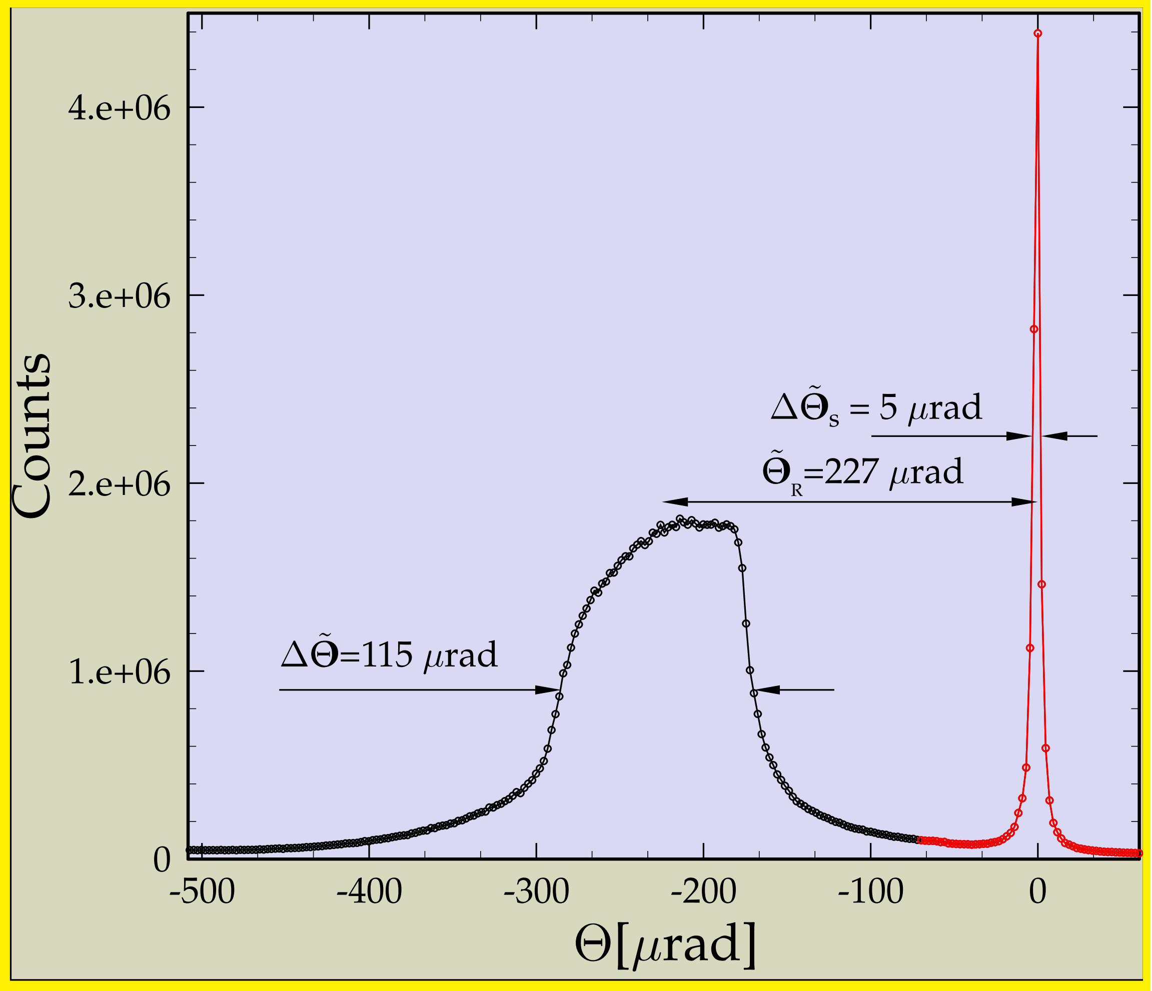
What do we observe?

1. **Exact Backscattering takes place NOT at normal incidence to atomic planes: angular shift $\Delta\Theta_R = 227\ \mu$rad**

2. **Angular dispersion: $2 \times \Delta\Theta_a = 230\ \mu$rad**

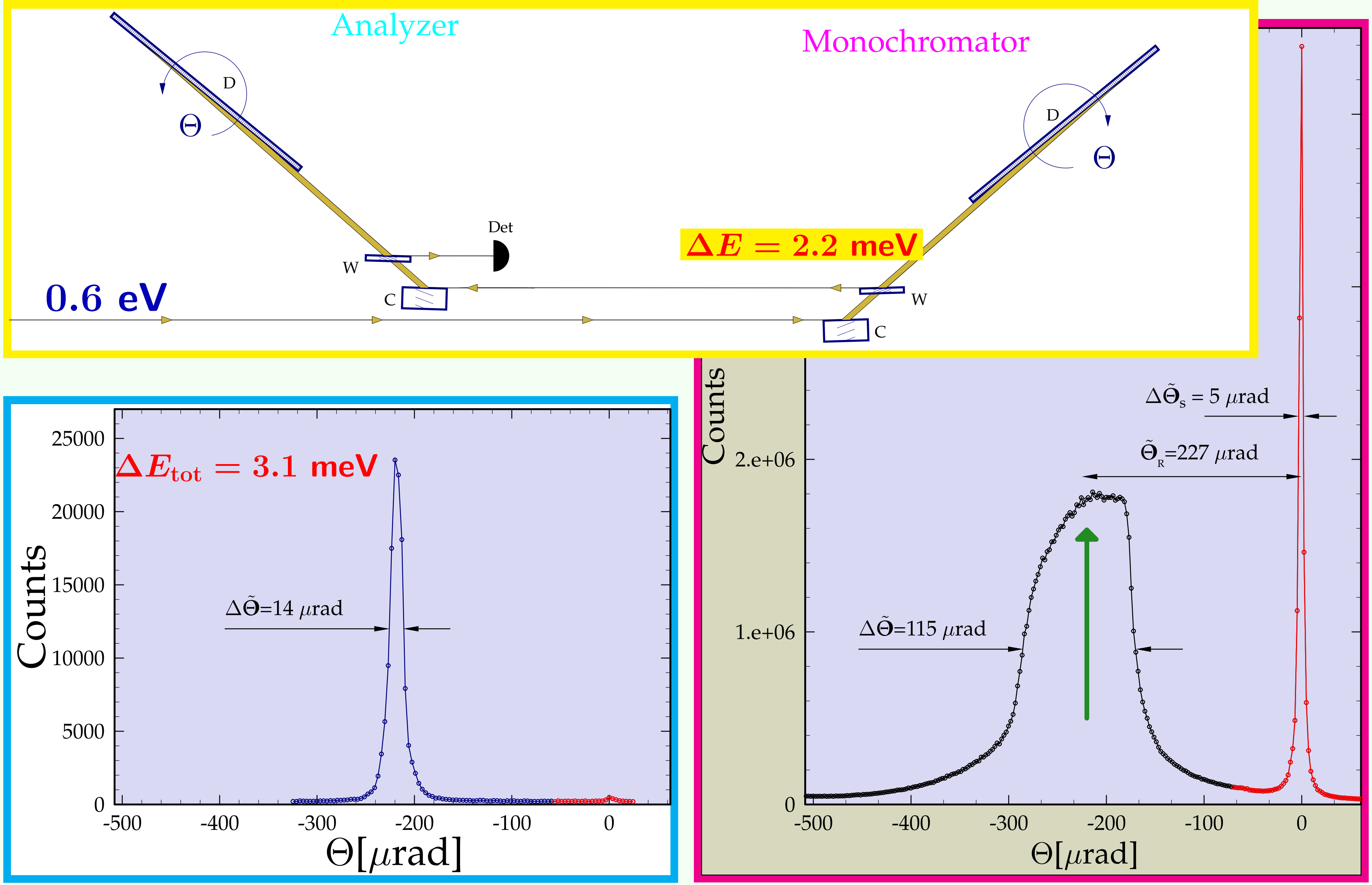
We are observing the effect of angular dispersion, underlying the principle of monochromatization





Shvyd'ko, Lerche, Kütgens, Rüter, Alatas, Zhao, *PRL* 97 (2006)

Energy Resolution Measurements



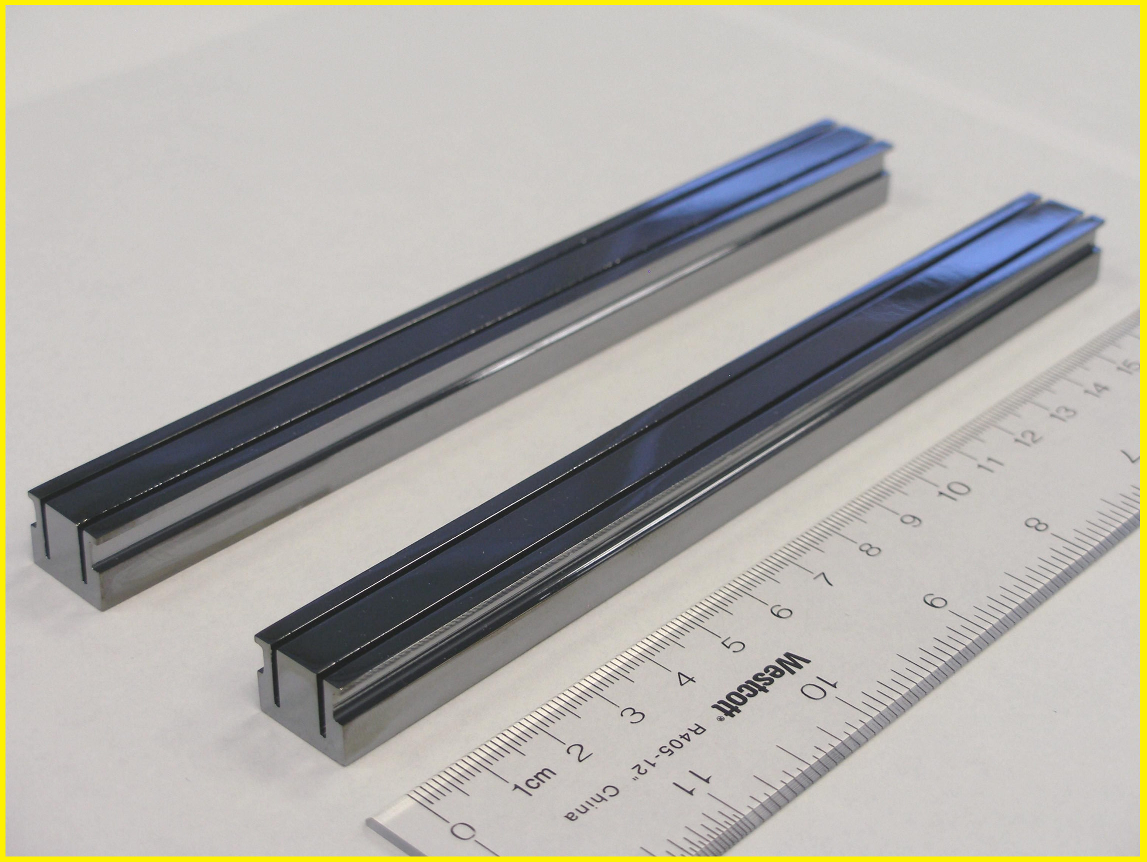
Why is it Broadened?

Good news: angular dispersion is working both as a physical effect, and the principle of monochromatization.

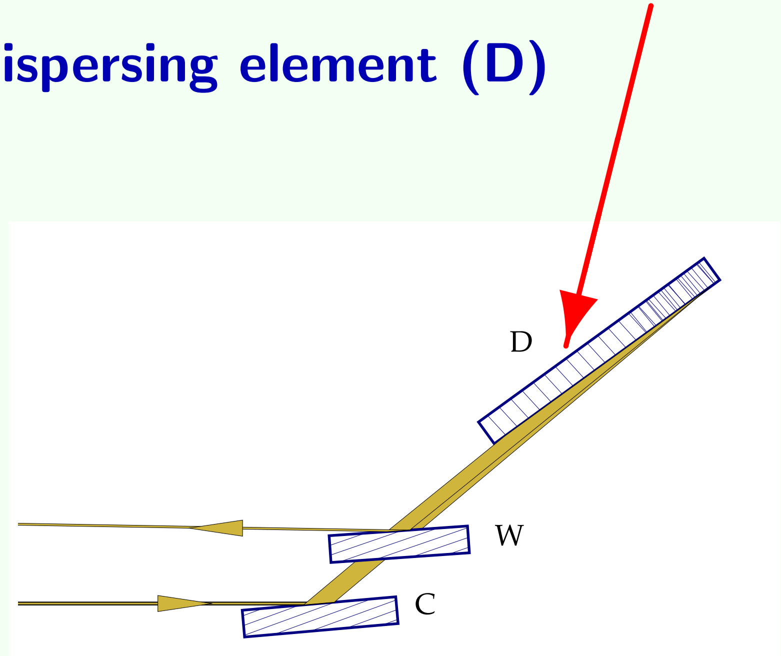
However: we are measuring $\Delta E = 2.1$ meV instead of a design value of 0.7 meV. Why?

- Crystal imperfection?
- Strain due to mounting?
- Strain due unfavorable crystal shape?
- Insufficient surface flatness?
- Temperature variations along the dispersing element?

Dispersing Silicon Crystal Elements = Long Crystals



dispersing element (D)

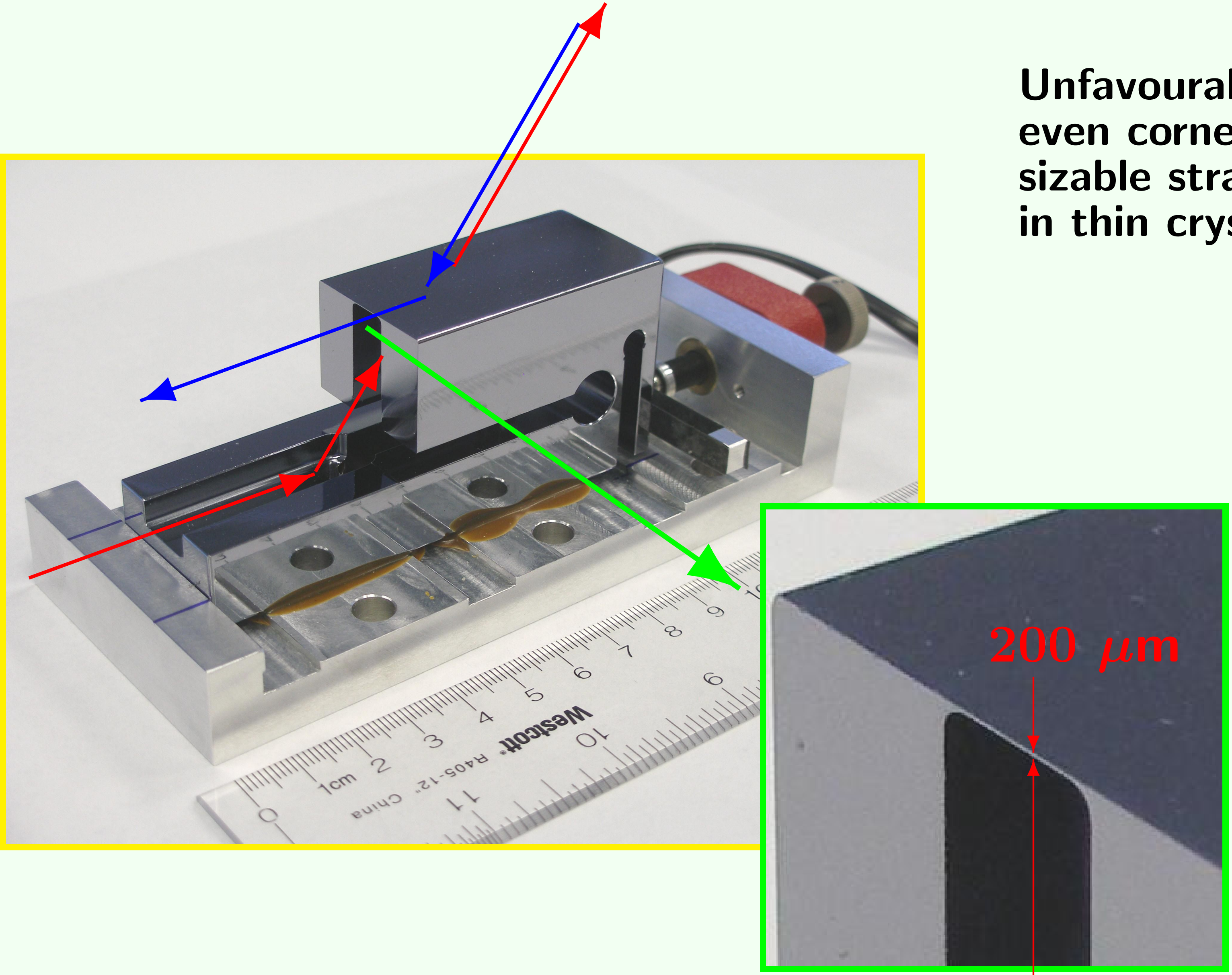


$\Delta E/E = 7 \times 10^{-8}$!

1. **Si of highest quality and purity is required, $\rho \geq 50$ kΩ·cm**
   Used: $\rho \simeq 1$ kΩ·cm

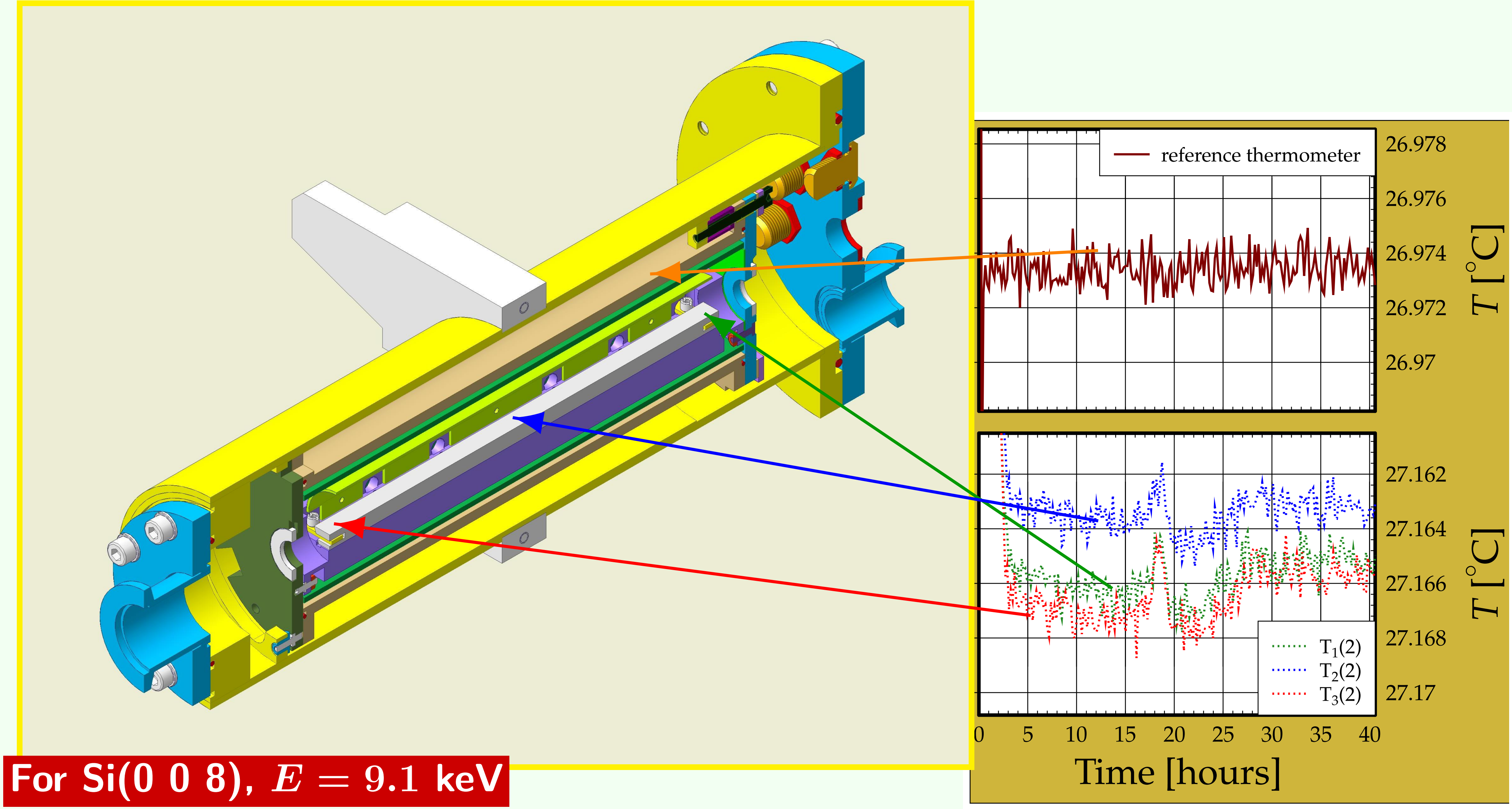
2. **Strain-free crystals and strain-free mounting are required.**

Collimator and Wavelength-Selector



Unfavourable crystal shape, even corners, can induce sizable strain especially in thin crystal parts.

Temperature variations along the dispersing element



For Si(0 0 8), $E = 9.1$ keV

0.1 meV $\Rightarrow$ 4.3 mK

Total variation: $\Delta T \lesssim 4$ mK $\Rightarrow$ $\Delta E \lesssim 0.1$ meV

Why is it Broadened?

- Crystal imperfection? (plausible)
- Strain due to mounting? (plausible)
- Strain due unfavorable crystal form? (plausible)
- Insufficient surface flatness? (plausible)
- Temperature variations along the dispersing element? (no)

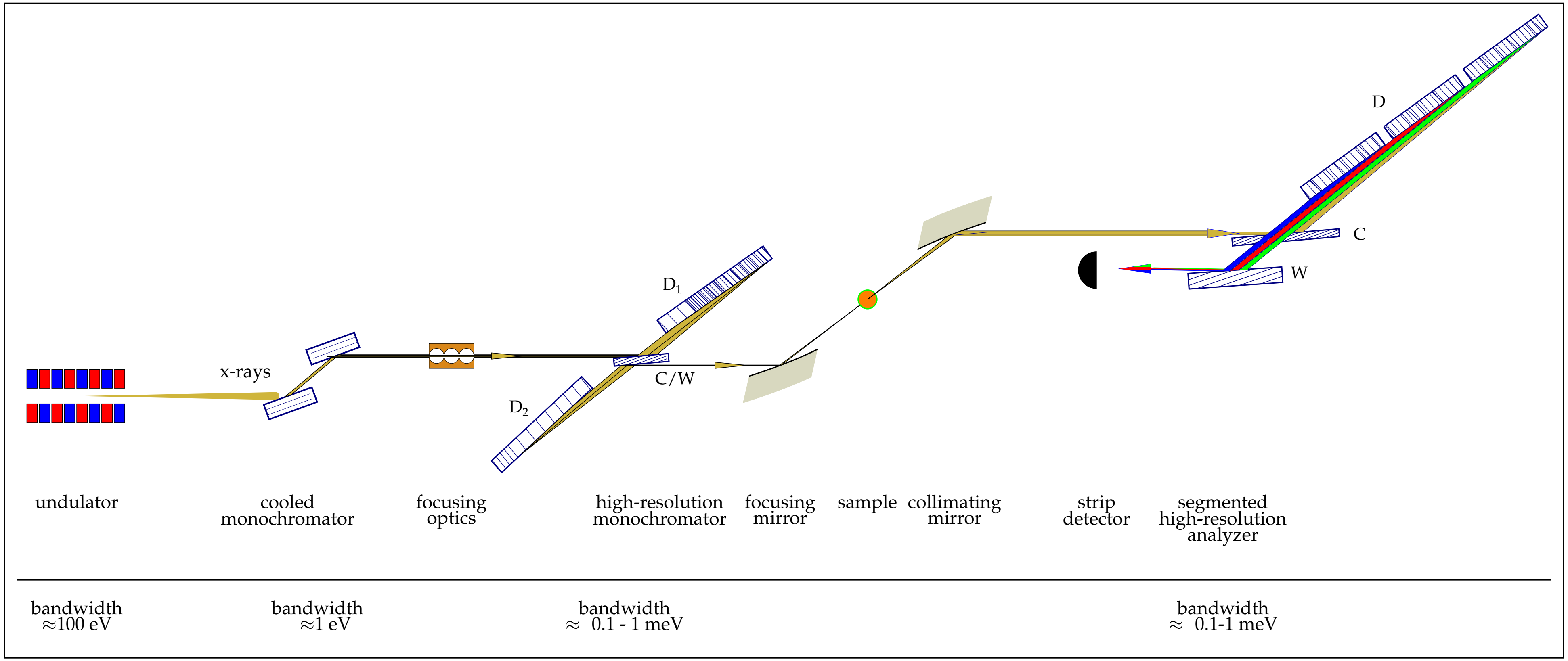
The plausible reasons have to be studied experimentally as part of the R+D program.

Summary of Initial Results

- The effect of angular dispersion in x-ray Bragg diffraction from asymmetrically cut crystals was observed experimentally.
- The effect of angular dispersion offers a new means for monochromatization of medium energy x-rays (5-10 keV), to meV and sub-meV bandwidths, not limited by the intrinsic widths of Bragg reflections.
- The CDW monochromator for 9.1 keV x-rays was demonstrated. The measured energy bandwidth of the monochromator is 2.2 meV (design value = 0.7 meV).
- Worse than theoretical resolution is attributed to (i) crystal imperfections, (ii) strain due to mounting, etc.
- Next goals: achieve 0.7 meV resolution, build 0.35 meV prototype IXS spectrometer, build 0.1 meV IXS spectrometer for NSLS-II.

Prerequisites for Building a Working Instrument

An IXS spectrometer with 0.1 meV resolution, operating at medium photon energies is feasible. There are no limitations in principal.



Prerequisites:

Identification of the technical problems

Resources

Technical Challenges in Building a Working Instrument

Crystal fabrication:

highest quality Si,
precisely machined 200-300 mm long crystals (1 mrad precise cuts),
precisely machined 60 mm long and 0.2 mm thin crystals (1 mrad precise cuts),
strain-free mounting,
polishing 20-30 cm long crystals with slope error $\lesssim 0.2$ mrad

Temperature control:

temperature homogeneity and stability 0.5 mK (=0.01 meV)

Multi-crystal alignment:

CDW analyzer with up to 10 crystal segments, parallel to $50\ \mu$rad

Collimating optics:

divergence $\lesssim 50\ \mu$rad

Timeline for Developing a 0.1 meV IXS spectrometer

Scope: To develop an IXS spectrometer with 0.1 meV resolution, operating at medium photon energies at NSLS-II

FY07-08: Develop prototype spectrometer for 9 keV x-rays with a resolution of 0.35 meV at APS.

FY09: Install and test prototype spectrometer at APS.

FY10: Engineering design for 0.1 meV spectrometer based on lessons learned.

FY11-12: Fabricate 0.1 meV spectrometer.

FY13: Commission 0.1 meV spectrometer at NSLS-II.

0.35 meV Prototype IXS Spectrometer: R+D plan

FY07: (selected items)

1. Redo crystals to achieve design resolution (0.7 meV) of the existing CDW test monochromators.

2. Dynamical theory simulations of the 0.35 meV CDDW in-line monochromator and CDW segmented analyzer.

3. 0.35 meV segmented CDW analyzer - design and production of elements:
   - 3.1. Production of the long crystals, and quality test.
   - 3.2. Multi-crystal alignment using micro- and nano-positioning systems.
   - 3.3. Construct and built multi-crystal pre-alignment arrangement.
   - 3.4. Thermally stable and homogeneous enclosure for the analyzer.
4. 0.35 meV in-line CDDW monochromator - design and production of elements:
   - 4.1. Production of the long crystals, and quality test.
   - 4.2. Collimator and wavelength selector production, and quality test.
   - 4.3. Thermally stable and homogeneous enclosure for the monochromator.
   - 4.4. Crystal alignment using weak link mechanisms.
5. Development of the computer based multi-sensor temperature control system.

6. Design and procurement of the focusing ($5\times 20\mu$m^2) and collimating mirrors (acceptance 7×14 mrad^2, divergence 0.08×0.3 mrad^2).

0.35 meV Prototype IXS Spectrometer: R+D plan

FY08: (selected items)

1. Place order for production of the in-line CDDW monochromator.
2. Place order for production of the multi-segmented CDW analyzer.
4. Fabricate long dispersive crystals elements for the CDDW monochromator.
5. Fabricate long dispersive crystals elements for the CDW analyzer.
6. Fabricate crystals for the C/W elements, and weak link alignment system.
7. Procure and commission strip detector and appropriate software for acquisition and handling the data.
7. Install and test focusing and collimating mirrors pairs on APS undulator beamline (3ID or 9ID)
8. Install and test the CDDW monochromator (3ID or 9ID).
9. Install and test the CDW spectrometer (3ID or 9ID).

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