Progress in the Development of
New Optics for Very High-Resolution Inelastic X-Ray Scattering Spectroscopy

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Advanced Photon Source

Main questions:

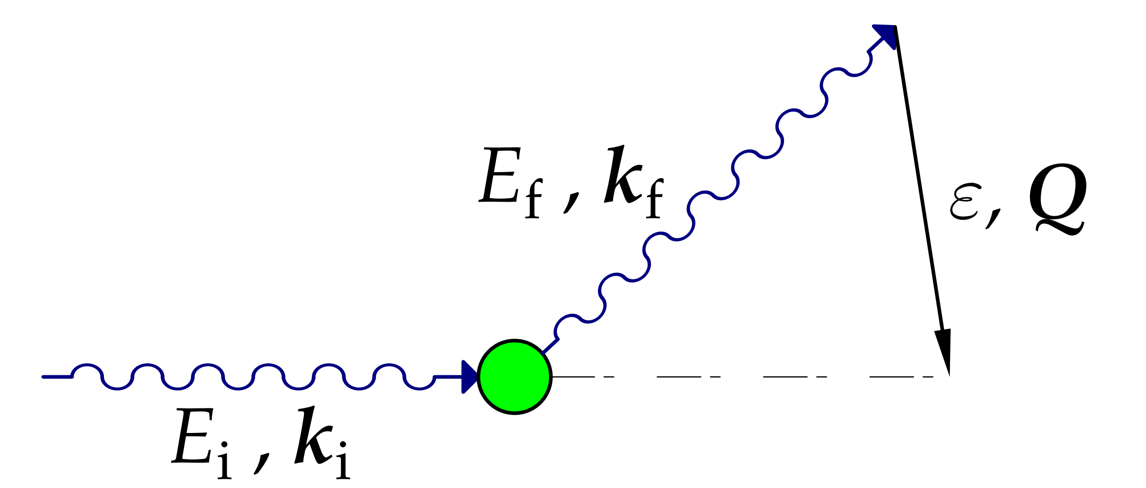
Can we perform the experiments on inelastic x-ray scattering (IXS) better than we are doing now, in terms of:

1. **energy transfer resolution $\Delta\varepsilon$?**

2. **momentum transfer resolution ΔQ?**

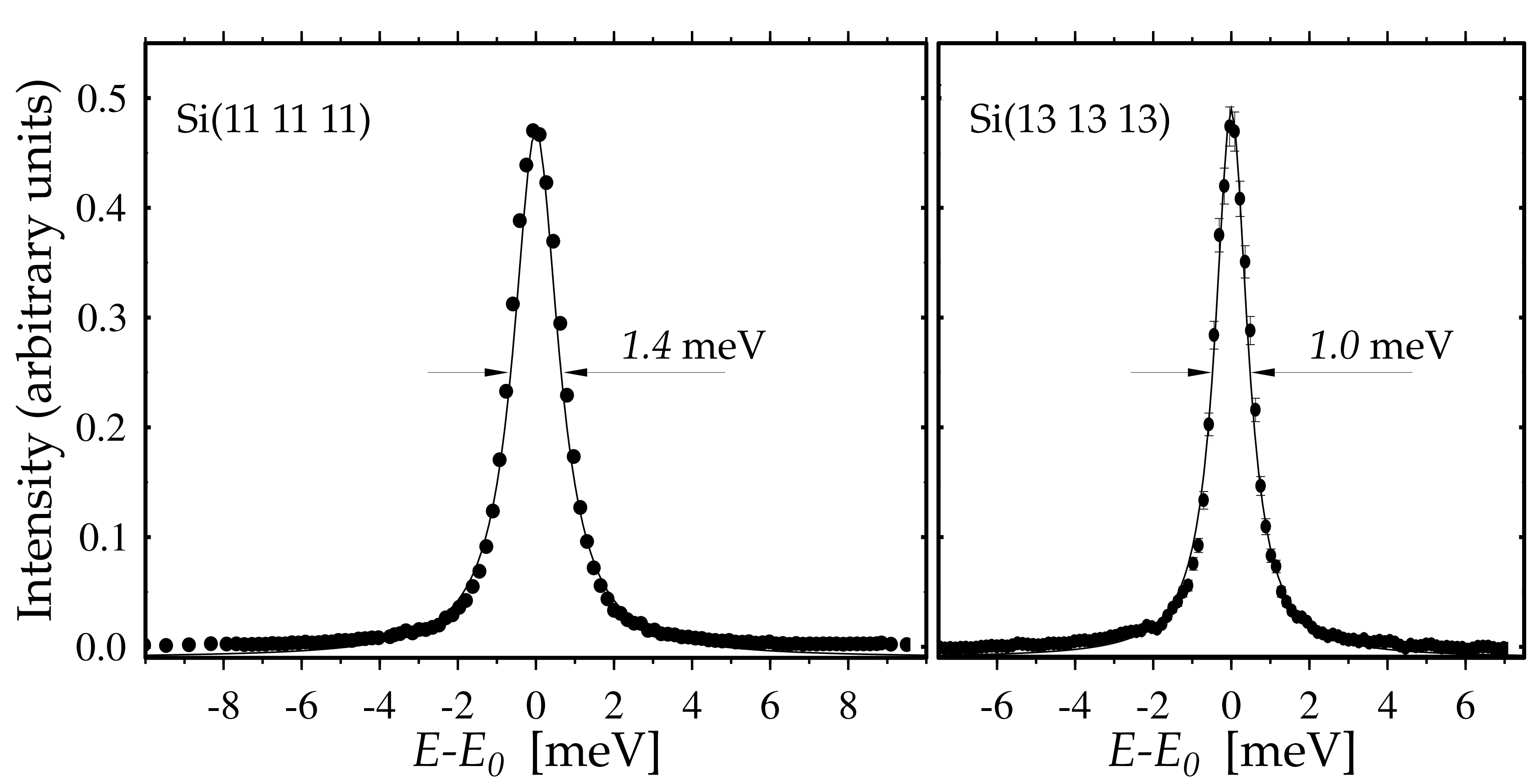
and

3. **count rate?**



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)

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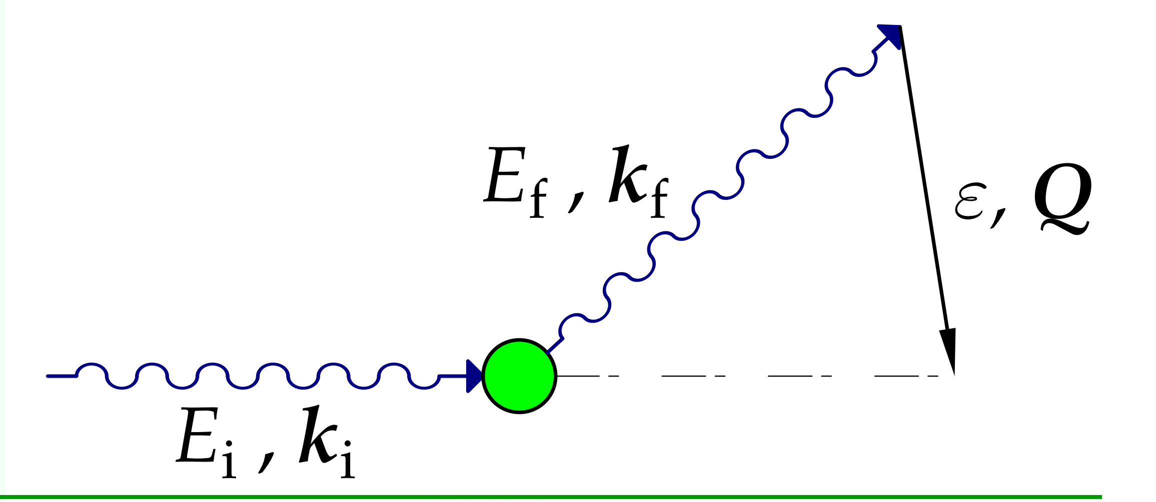
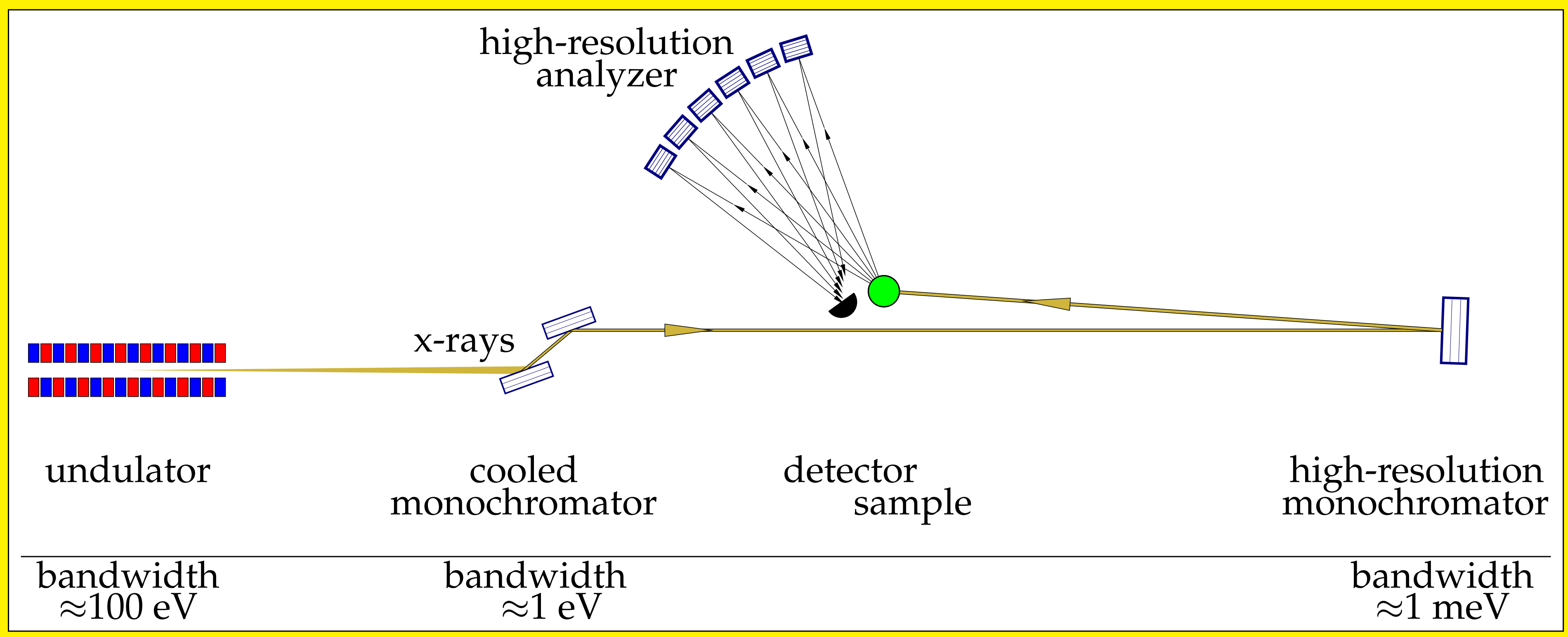


Content

- **IXS spectroscopy with meV resolution: how it works now?**
- **Angular dispersion - as alternative monochromatization principle.**
- $(+, +, \pm)$ **Monochromator.**
- $(+, +, \pm)$ **Analyzer.**
- **Layouts of the novel IXS spectrometer.**
- **Present status of the experimental effort**
- **First experiment: observation of the effect of the angular dispersion**
- **Summary and outlook**

Modern IXS Spectrometer (layout)

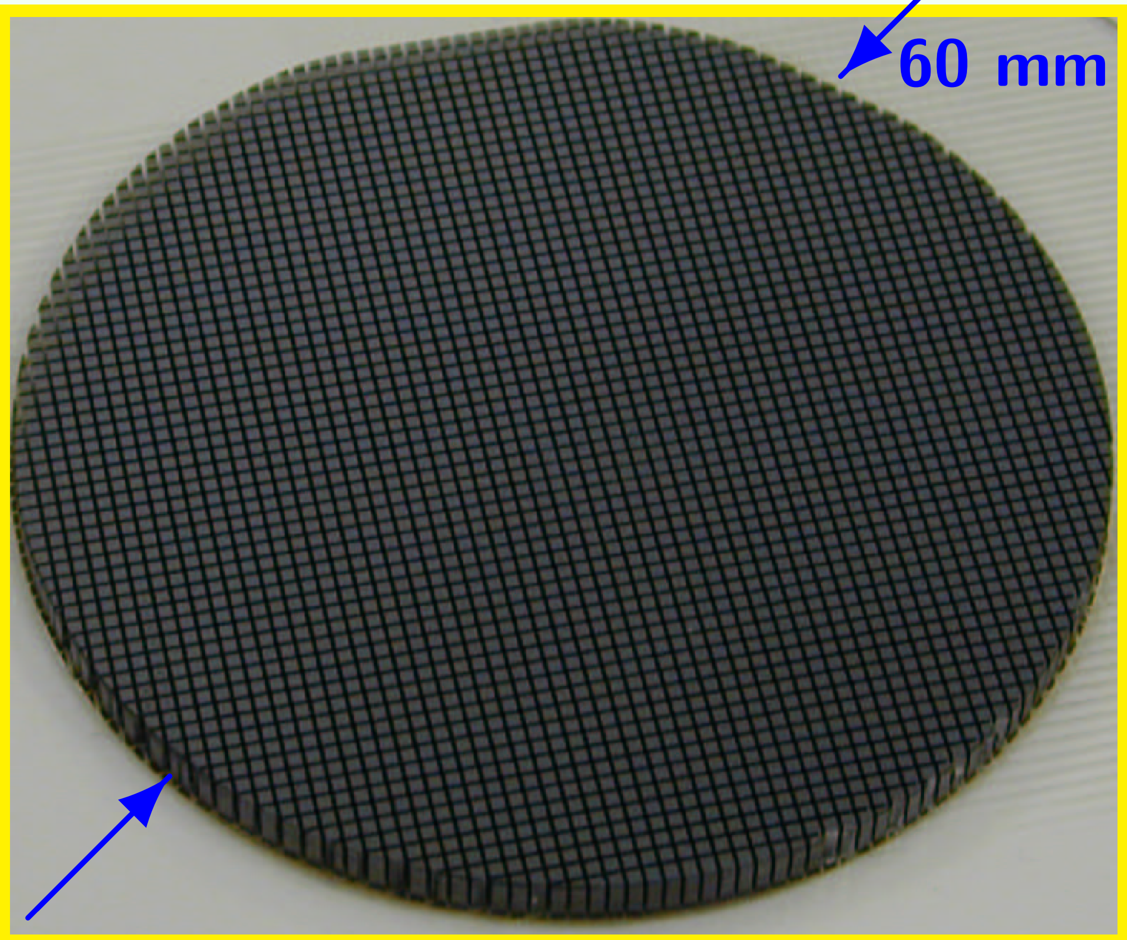


High-resolution analyzer is a two-dimensional array of flat crystals on a sphere of radius $\boldsymbol{R}_{\mathrm{A}}$

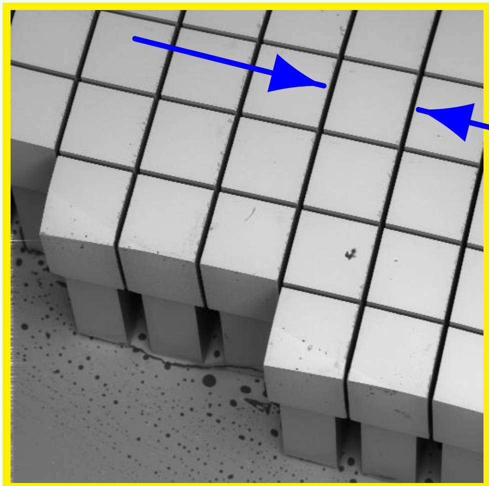
High-Resolution Spherical Crystal Analyzer

Courtesy of H. Sinn (APS)

60 mm

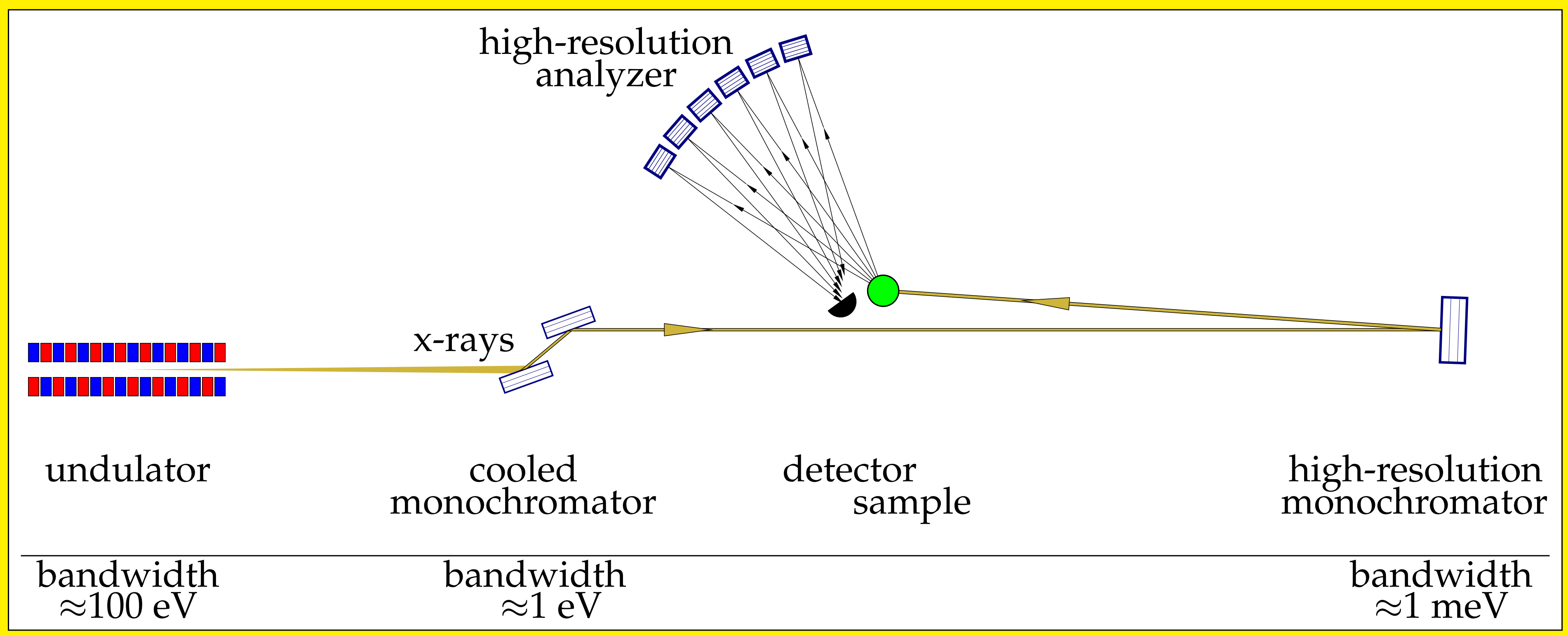


$\simeq$ **3000 pixels**



1 mm

Modern IXS Spectrometer (layout)



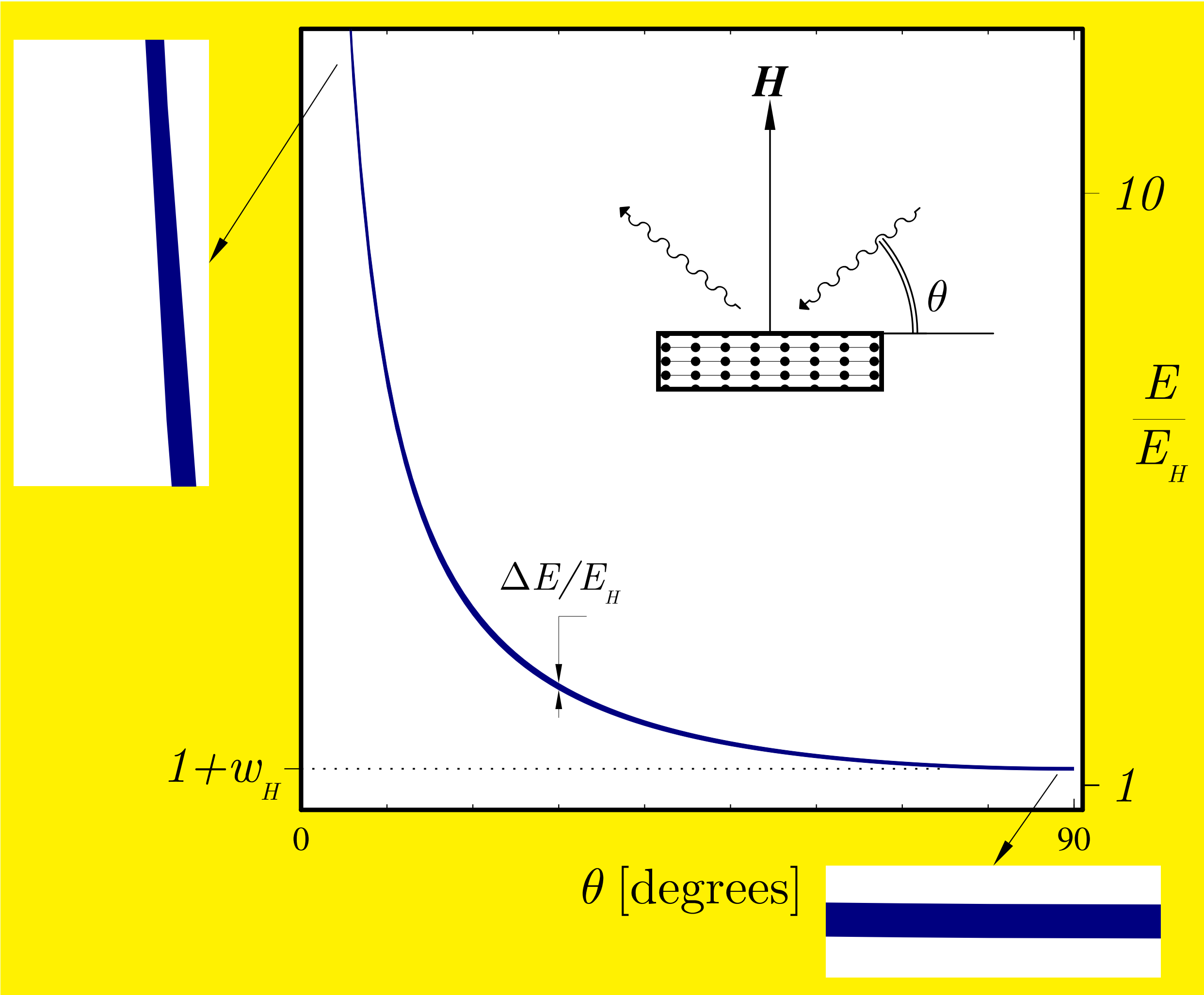
Kohra, Matsushita (1972)

Graef, Materlik (1982)

Burkel, Dorner, Peisl (1987)

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

X-ray Bragg diffraction and crystal reflectivity



Bragg's law:

$$E\,\sin\theta = E_H\,(1 + w_H)$$

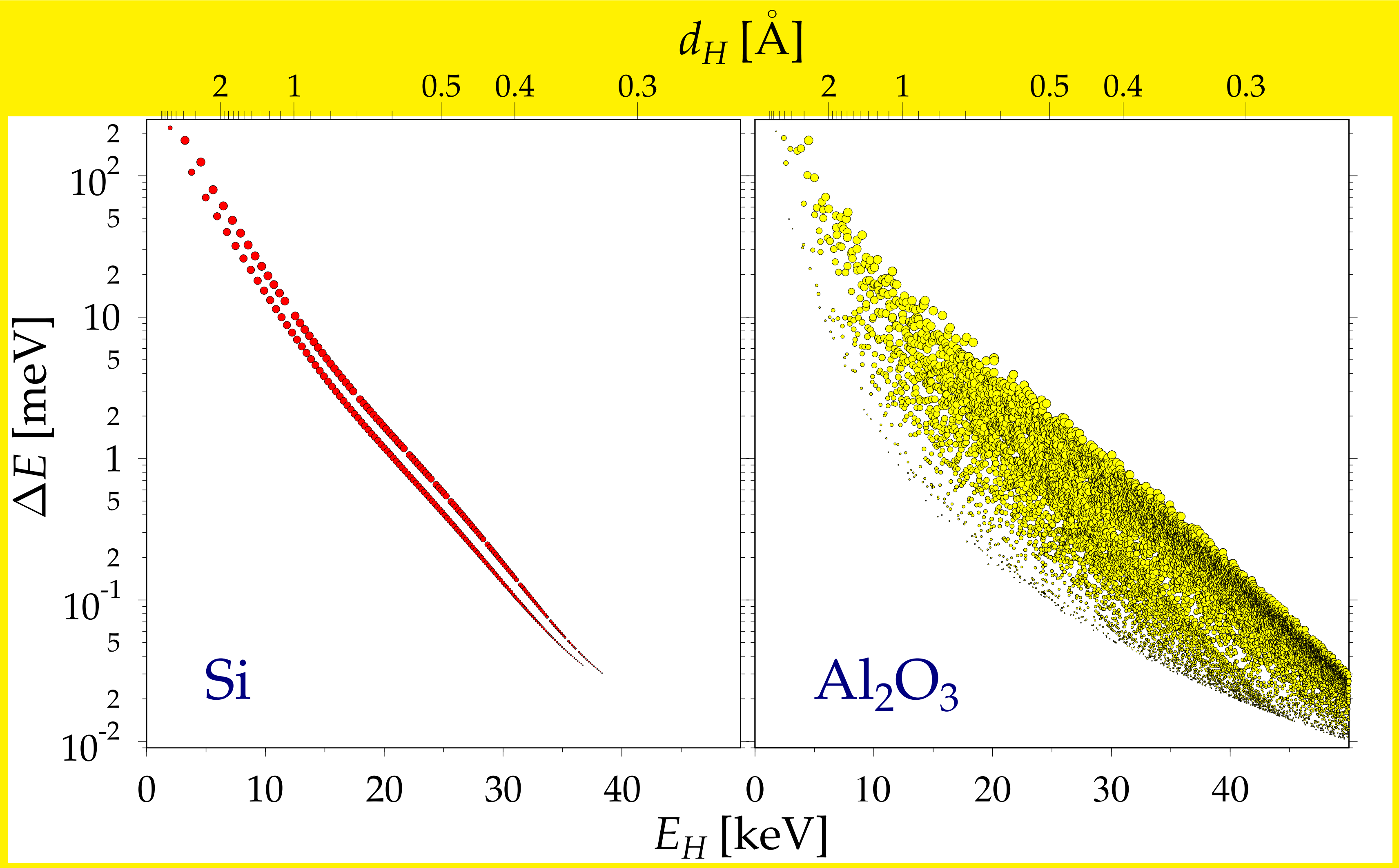
$$E_H = \tfrac{1}{2}\,\hbar\,c\,H$$

$$H = 2\pi/d_H$$

$$w_H \lesssim 10^{-4}$$

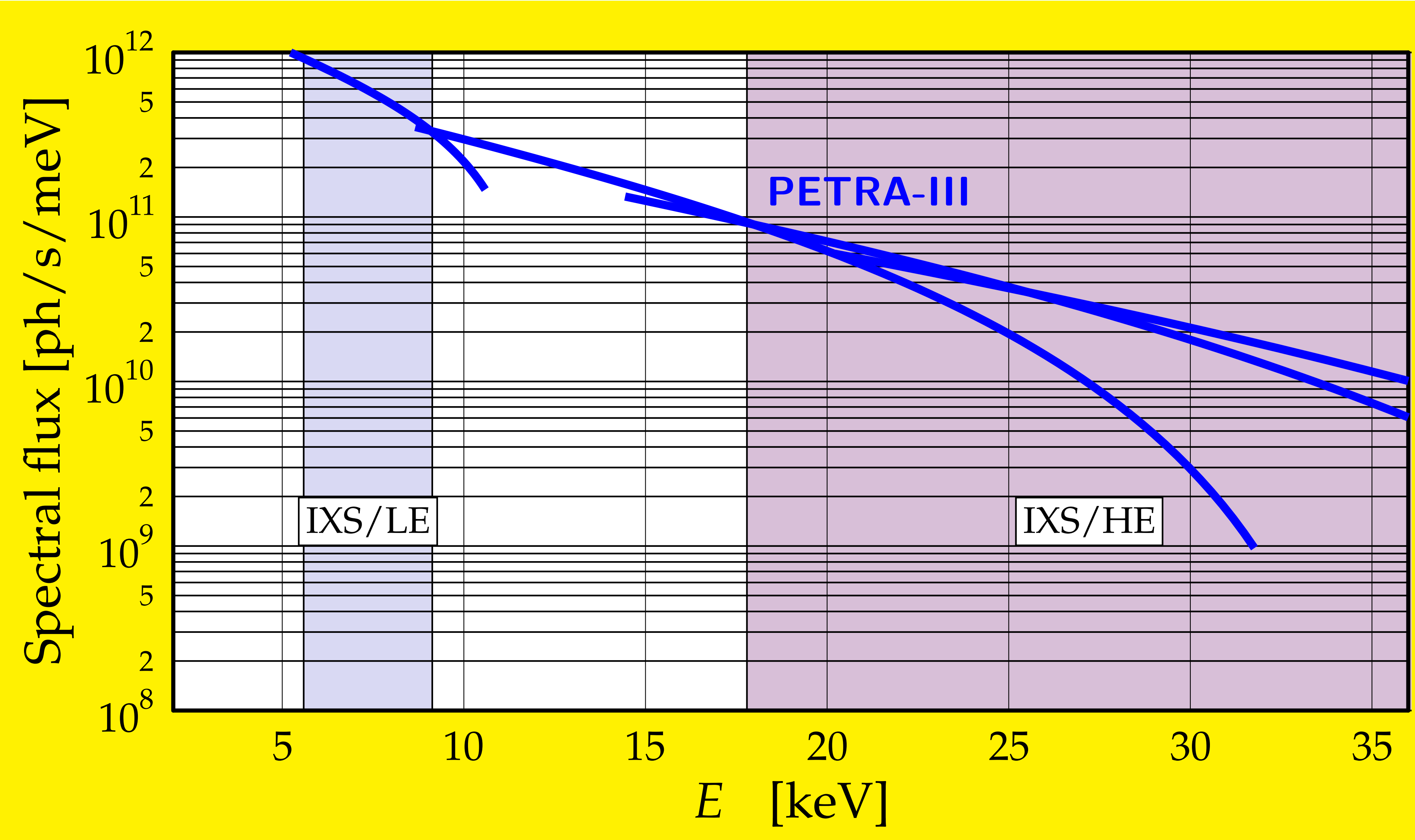
$$\frac{\Delta E}{E} = f(H) = \text{const}$$

ΔE of Bragg reflections in Si, and in Al_2O_3 (sapphire)



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

Undulator spectrum



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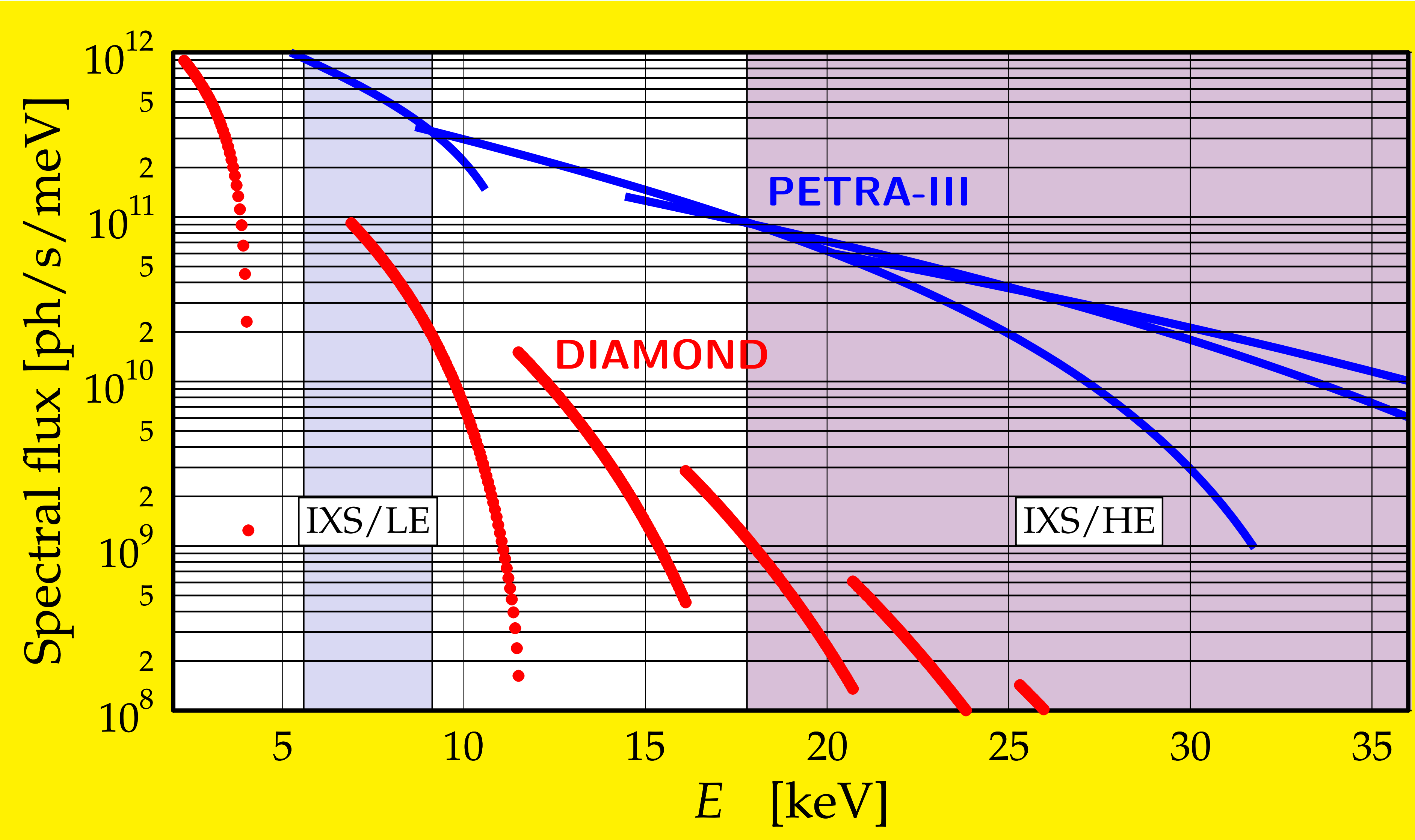

Low-energy photons would be better:

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

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

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

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... but ...

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

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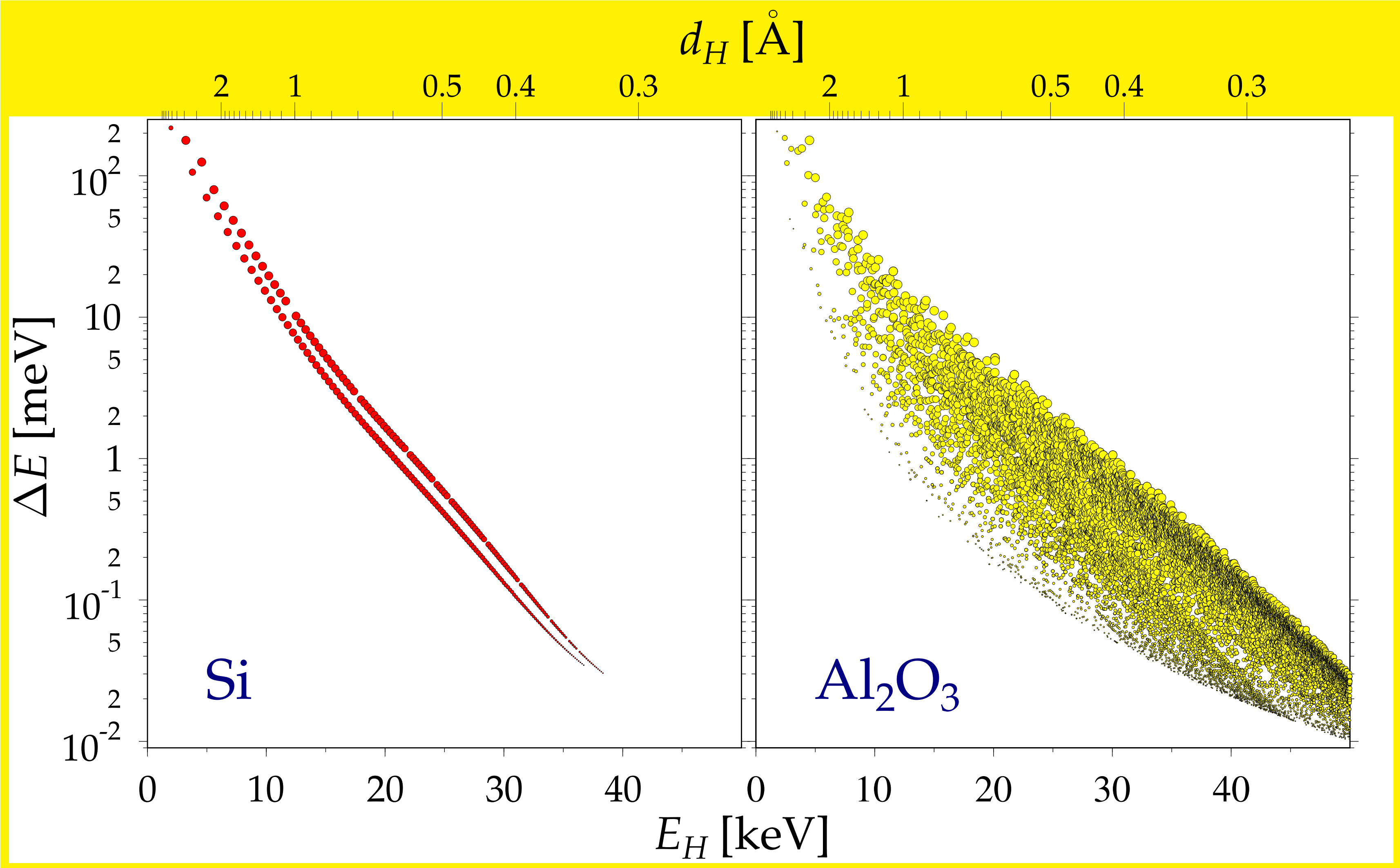

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New concepts, new solutions are required:

Problem:

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

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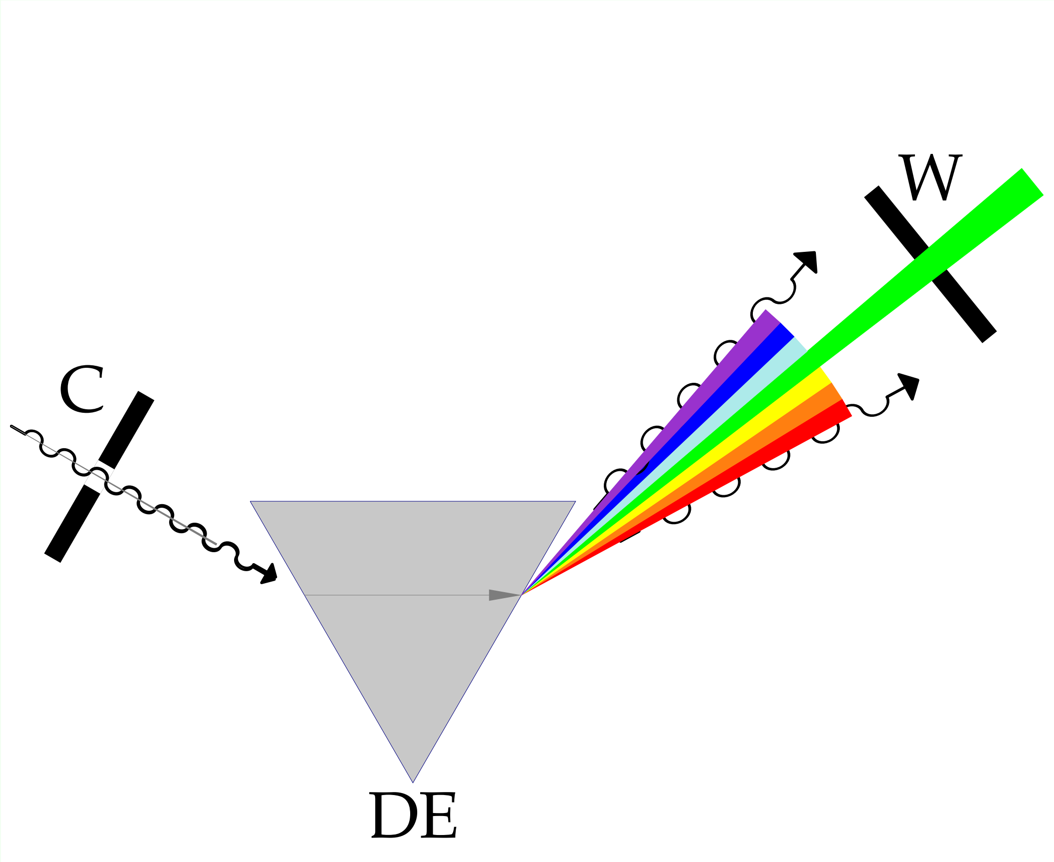
Solution:

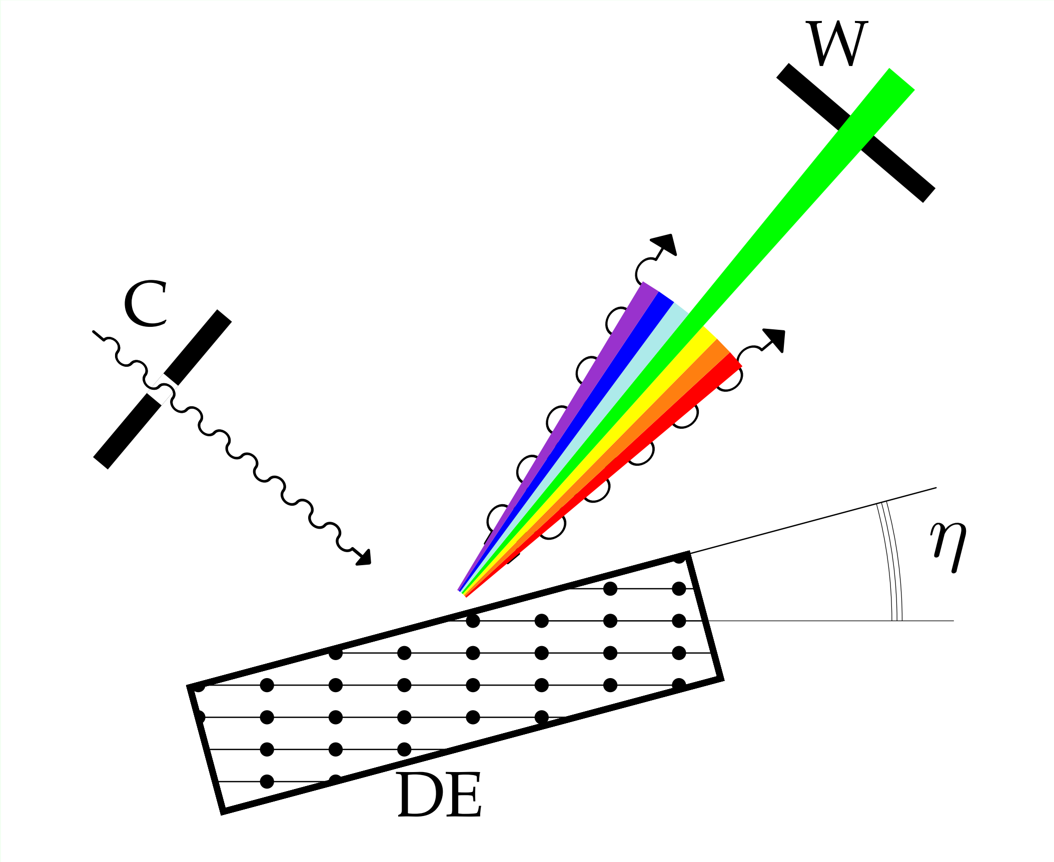
Use a small fraction of it!

New concept illustrated with optical prism



DE - dispersing element C - collimator W - wavelength selector

New concept



DE - dispersing element C - collimator W - wavelength selector

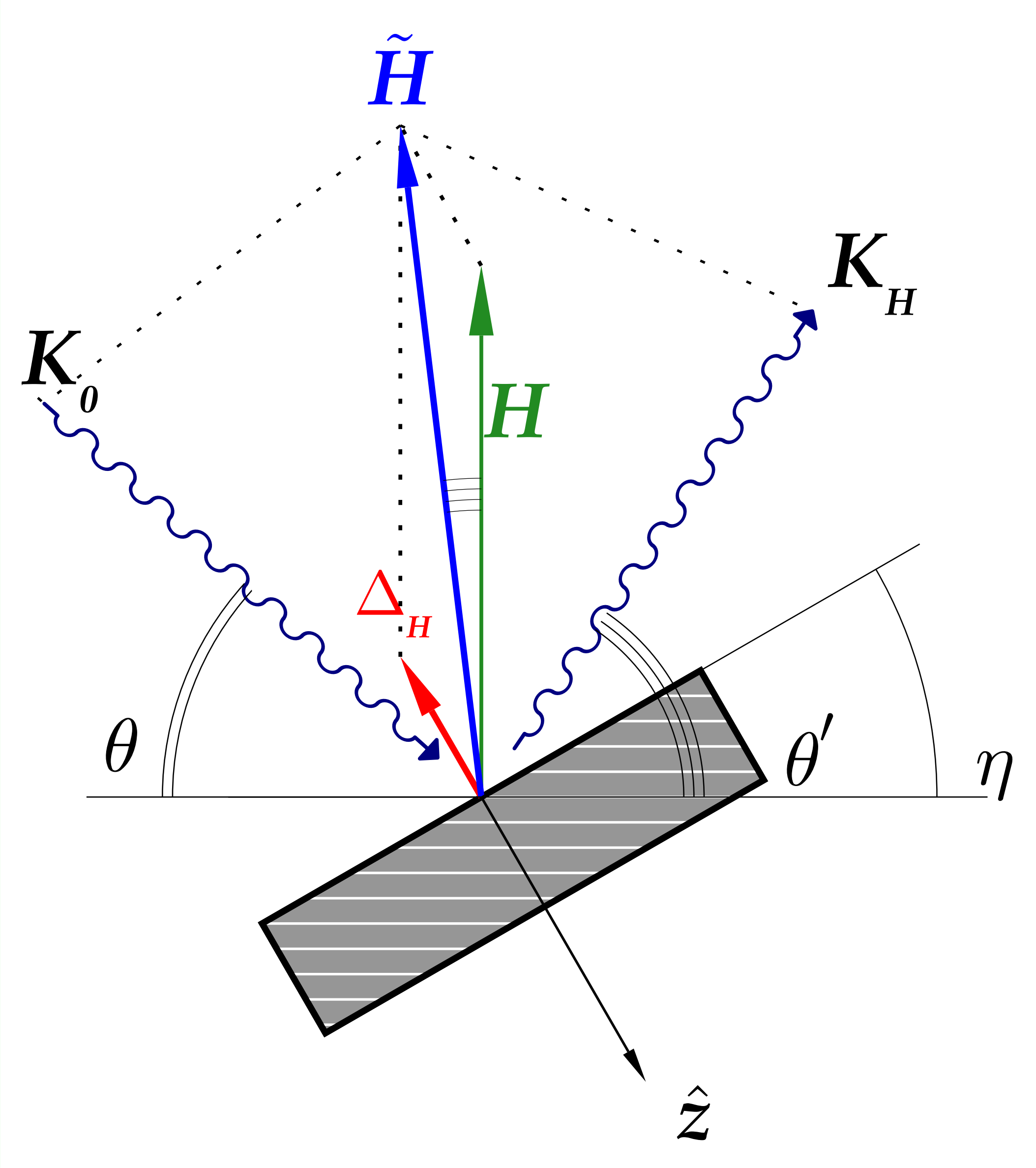
An asymmetrically cut crystal behaves like the optical prism dispersing the photons with different photon energies: effect of angular dispersion.

Effect of angular dispersion (1)

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

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

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



$$\alpha \propto 1 - n$$

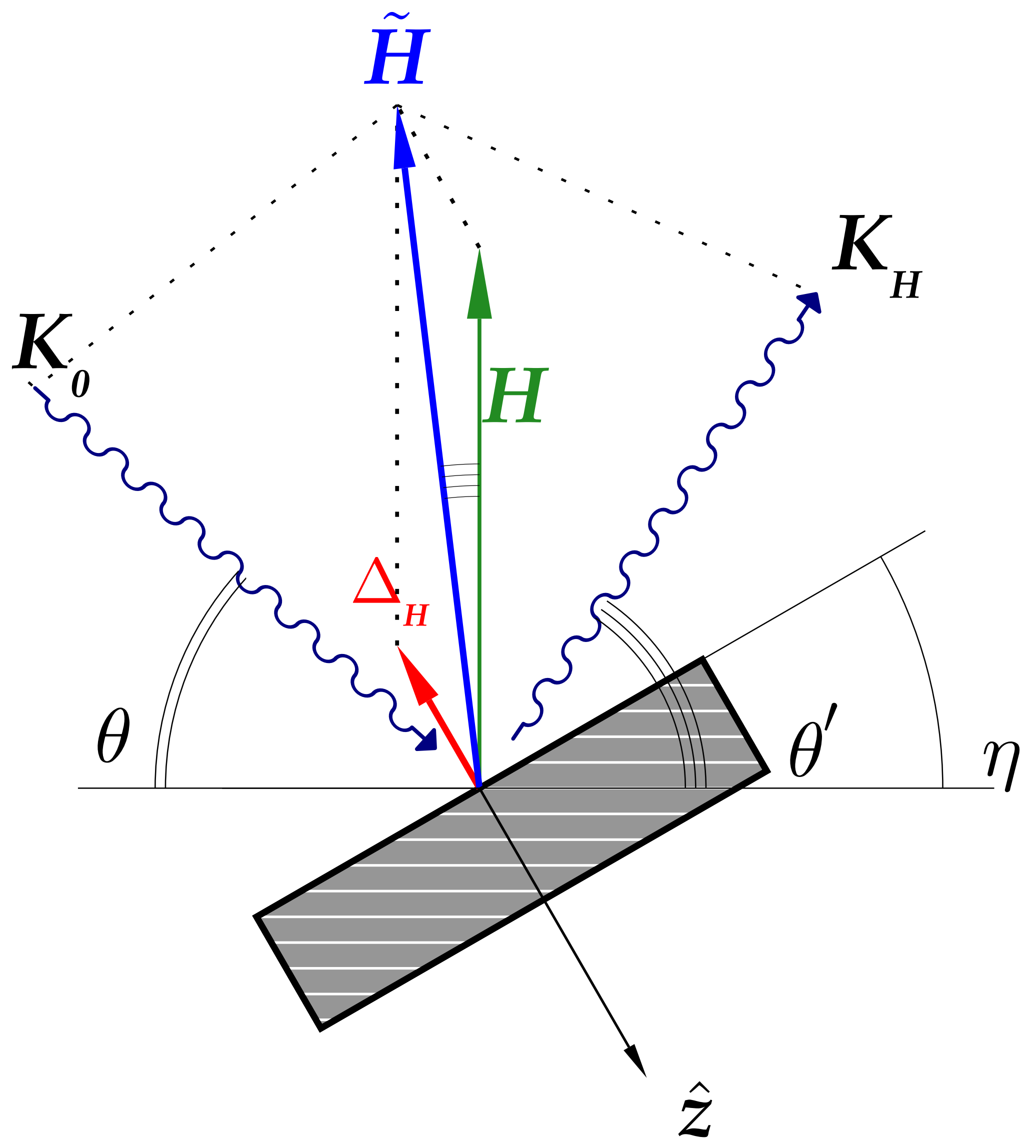
n – refractive index

Effect of angular dispersion (2)

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

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

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



$\theta < \pi/2$

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

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

Effect of angular dispersion (3)

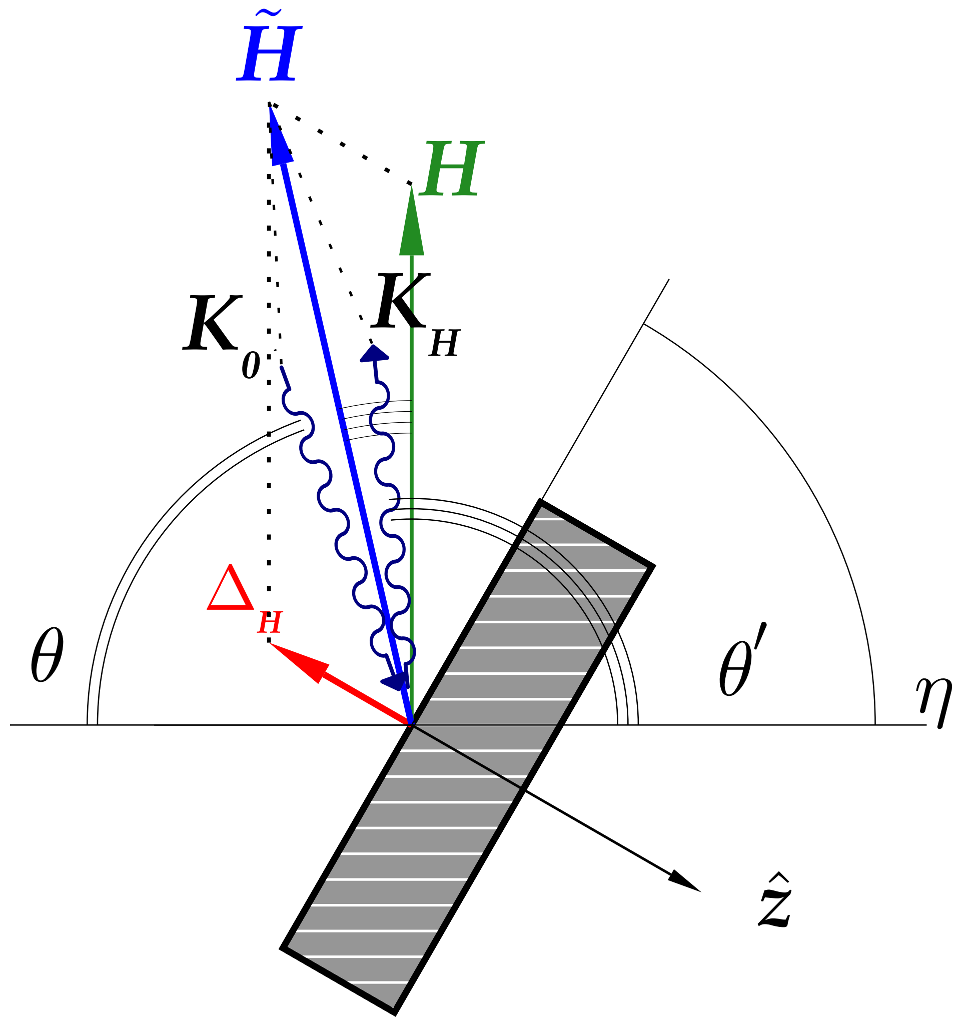
$$K_H = K_0 + \tilde{H}$$

$$\tilde{H} = H + \Delta_H$$

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

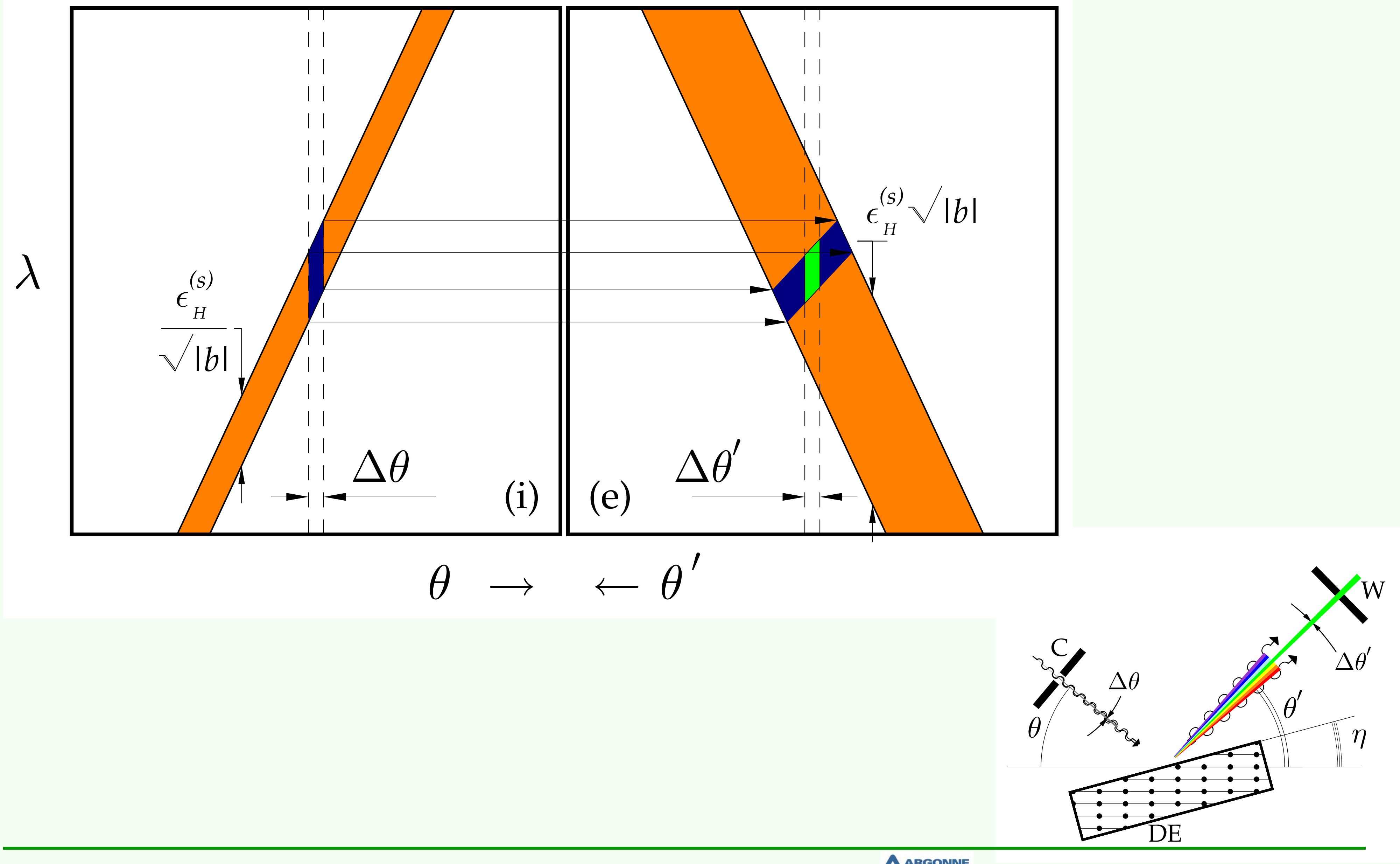
$\theta \simeq \pi/2$

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

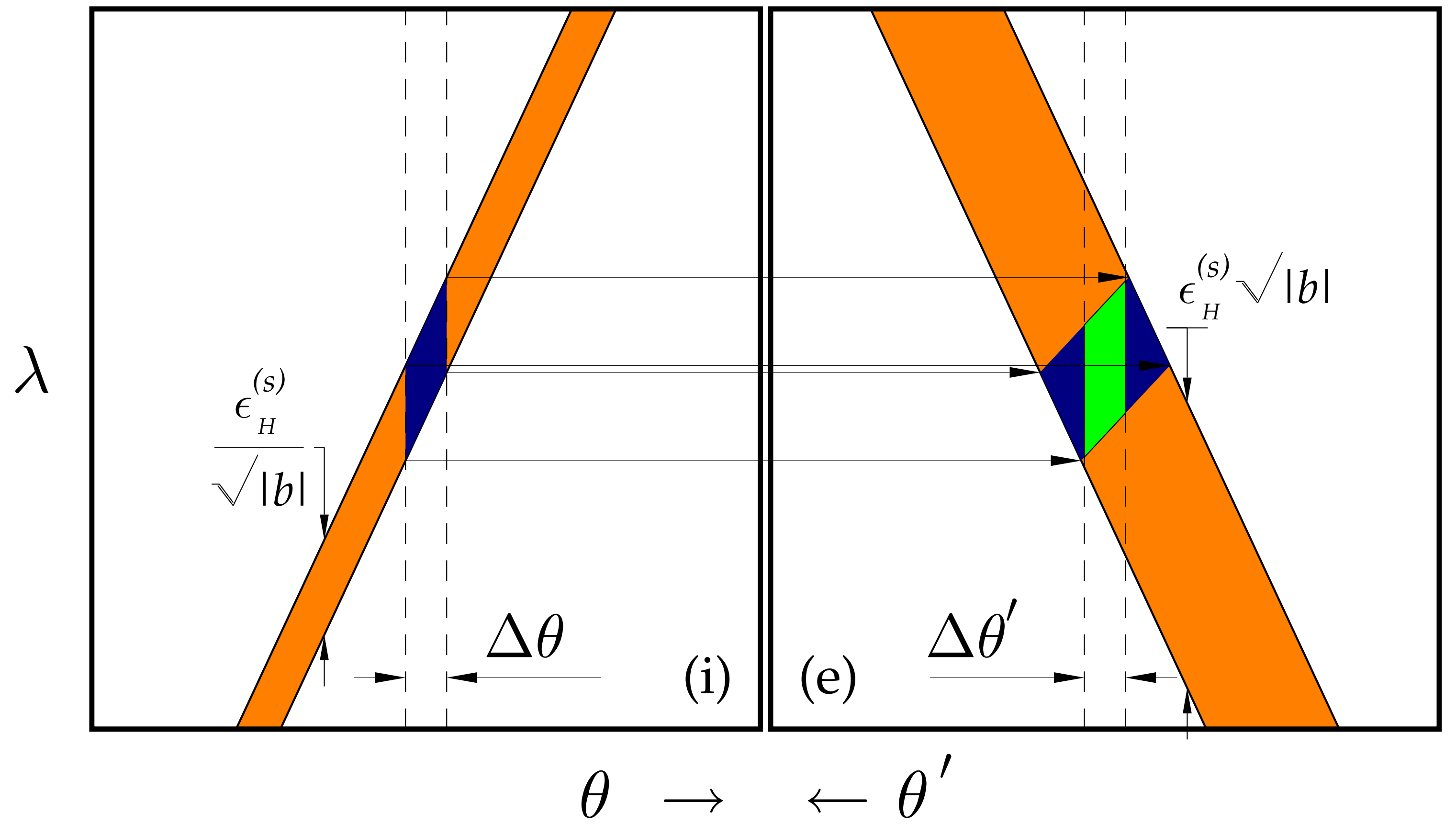


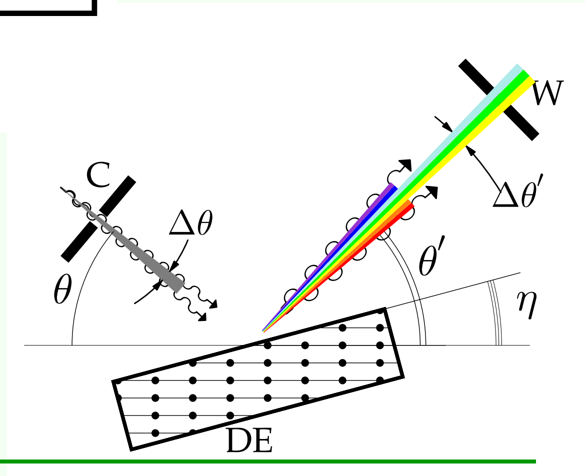
AND: Exact Backscattering is NOT at normal incidence!

Effect of angular dispersion (4)

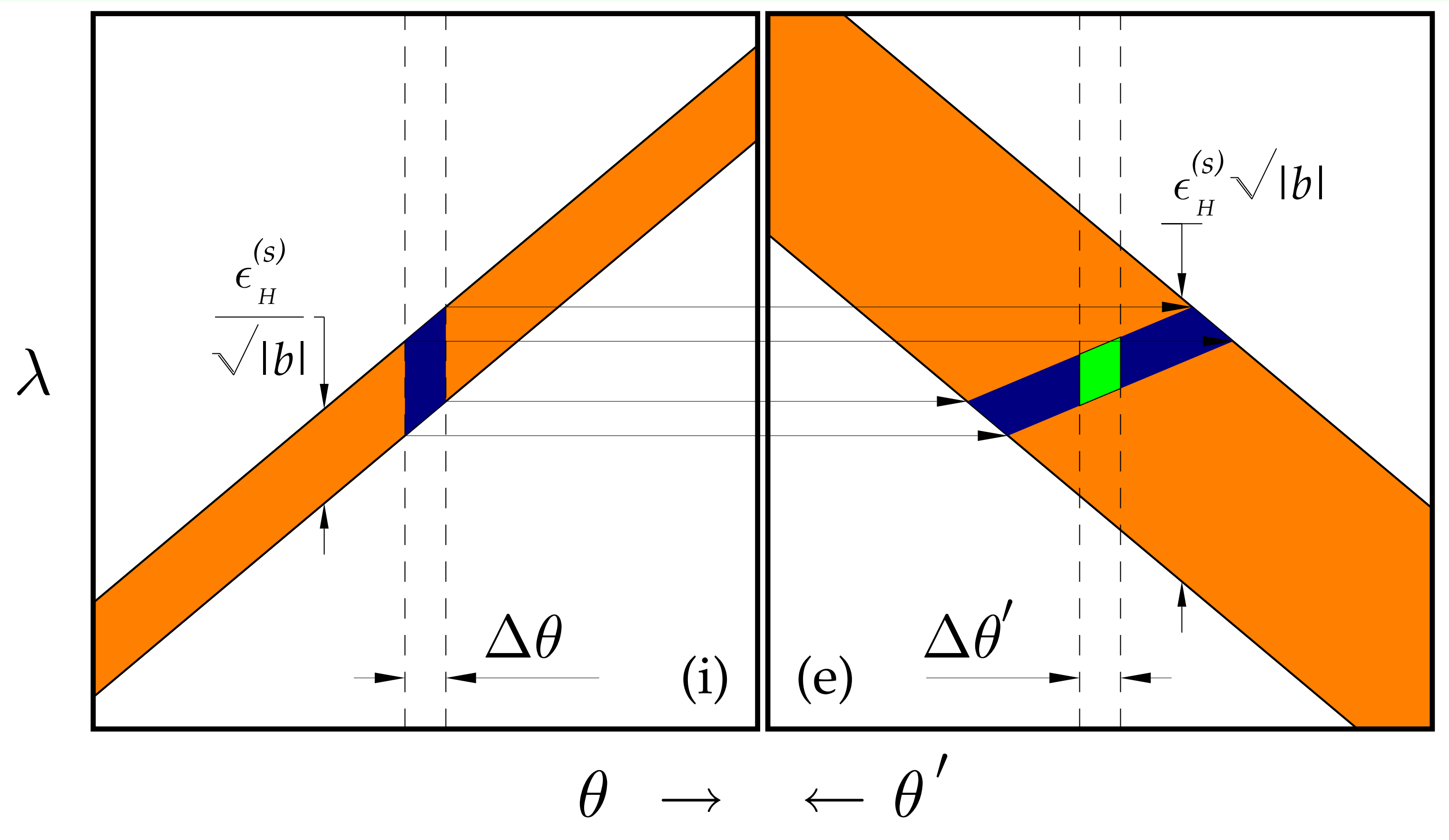


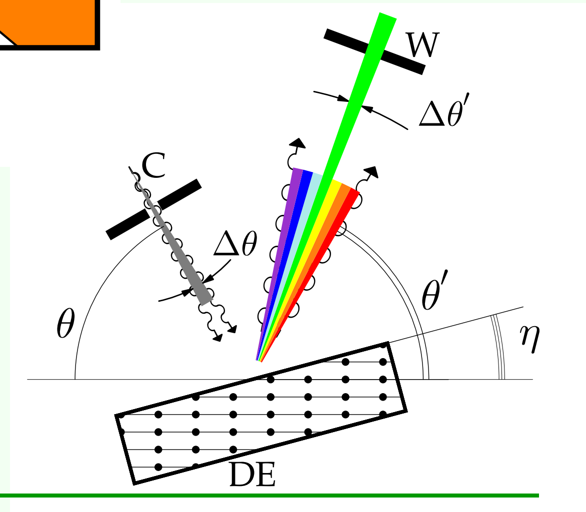
Effect of angular dispersion (5)





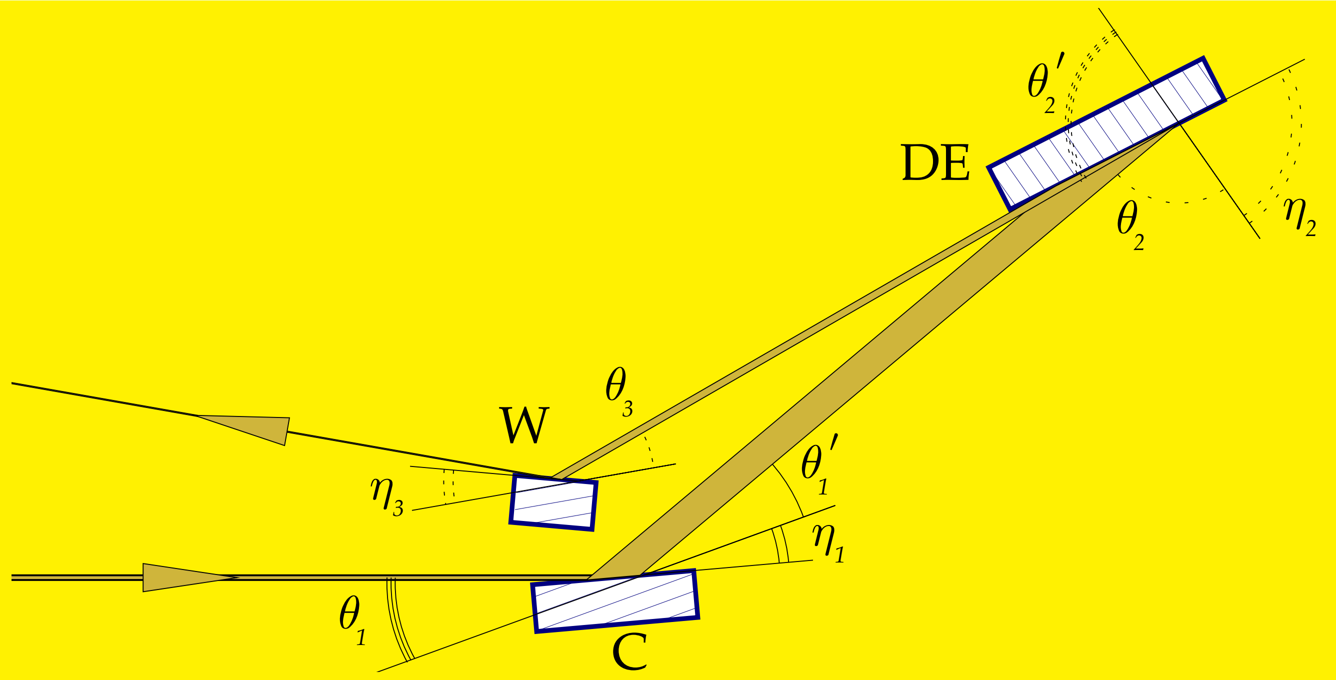
Effect of angular dispersion (6)





Use a small fraction of the Bragg reflection width!

$(+,+,-)$ Monochromator



DE - dispersing element C - collimator W - wavelength selector

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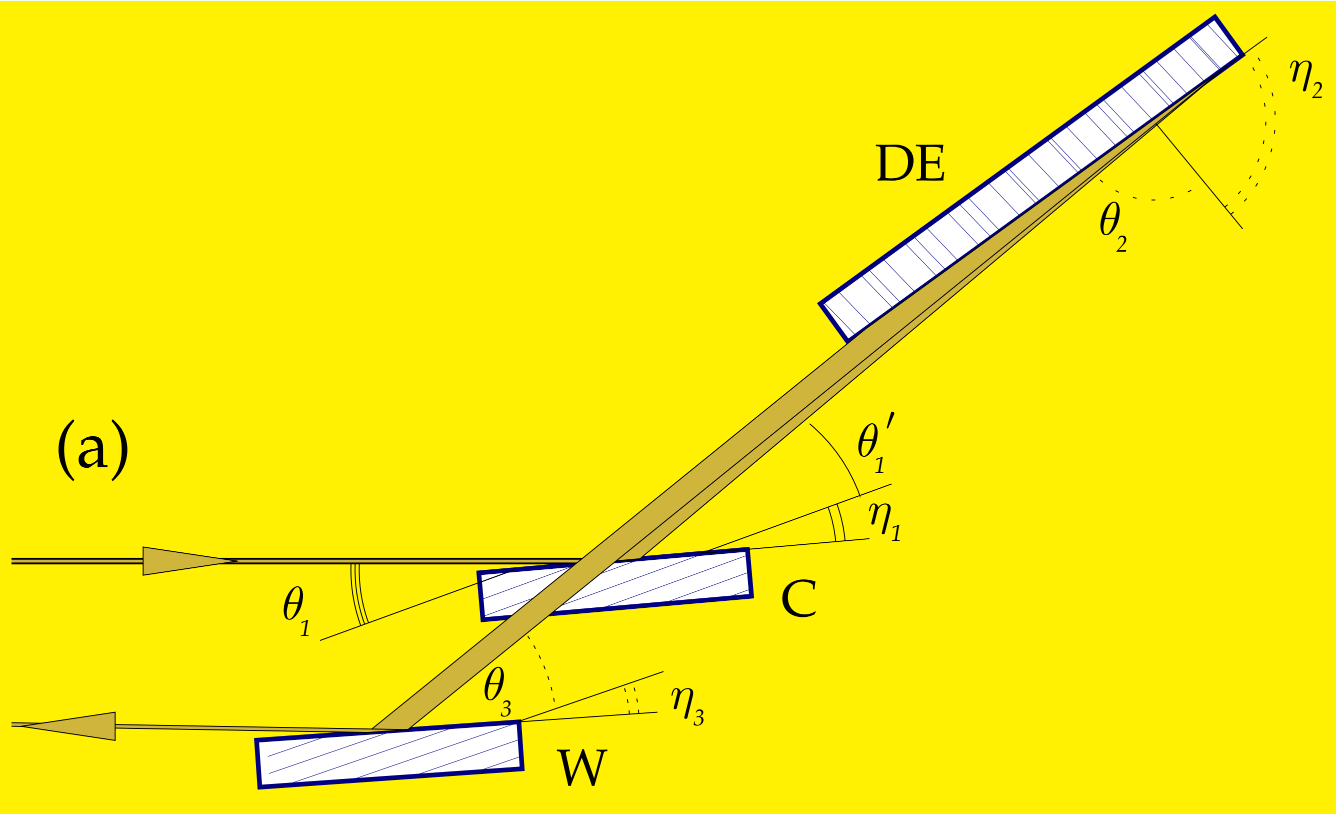

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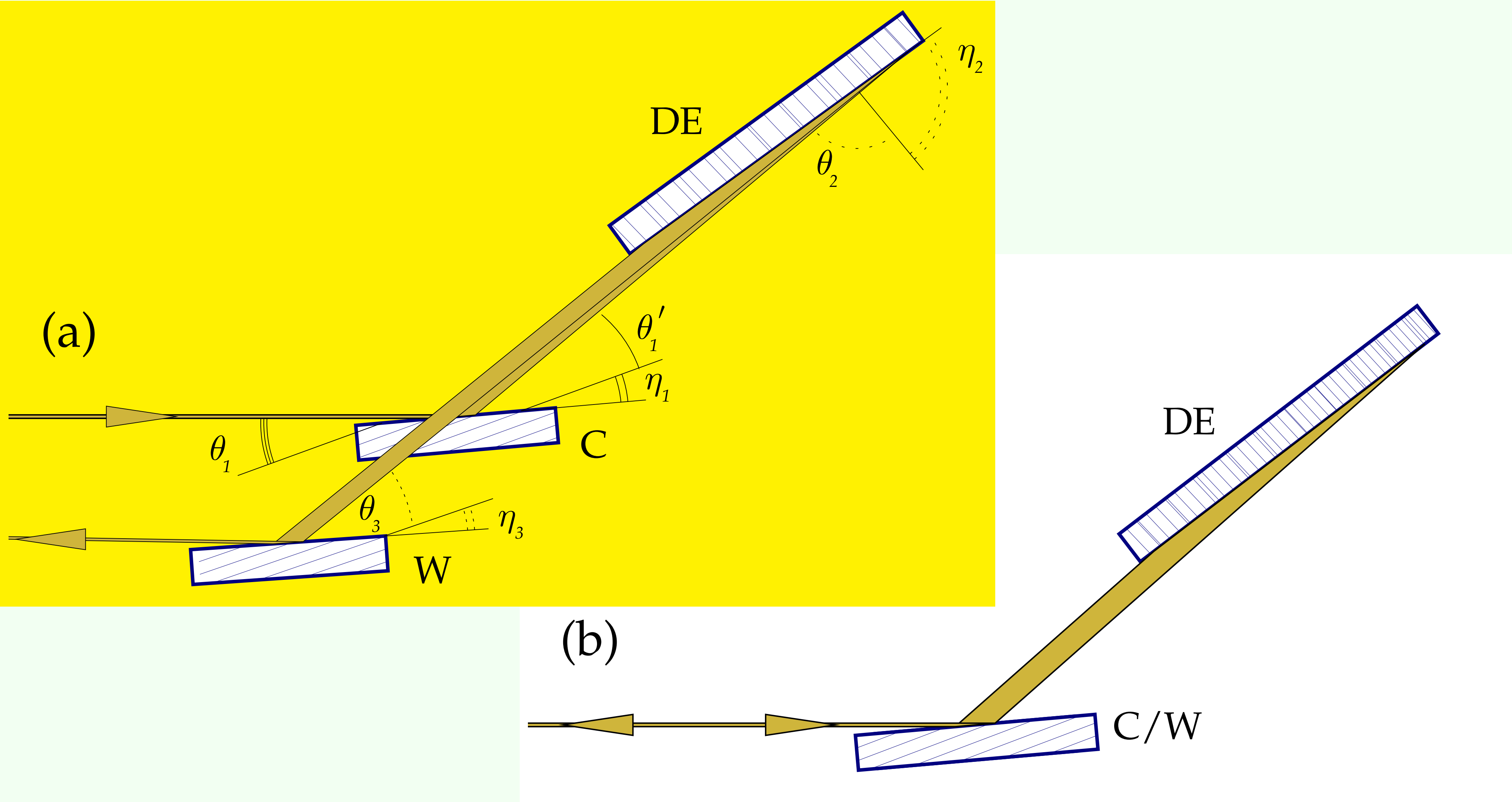
$(+,+,\pm)$ Monochromator



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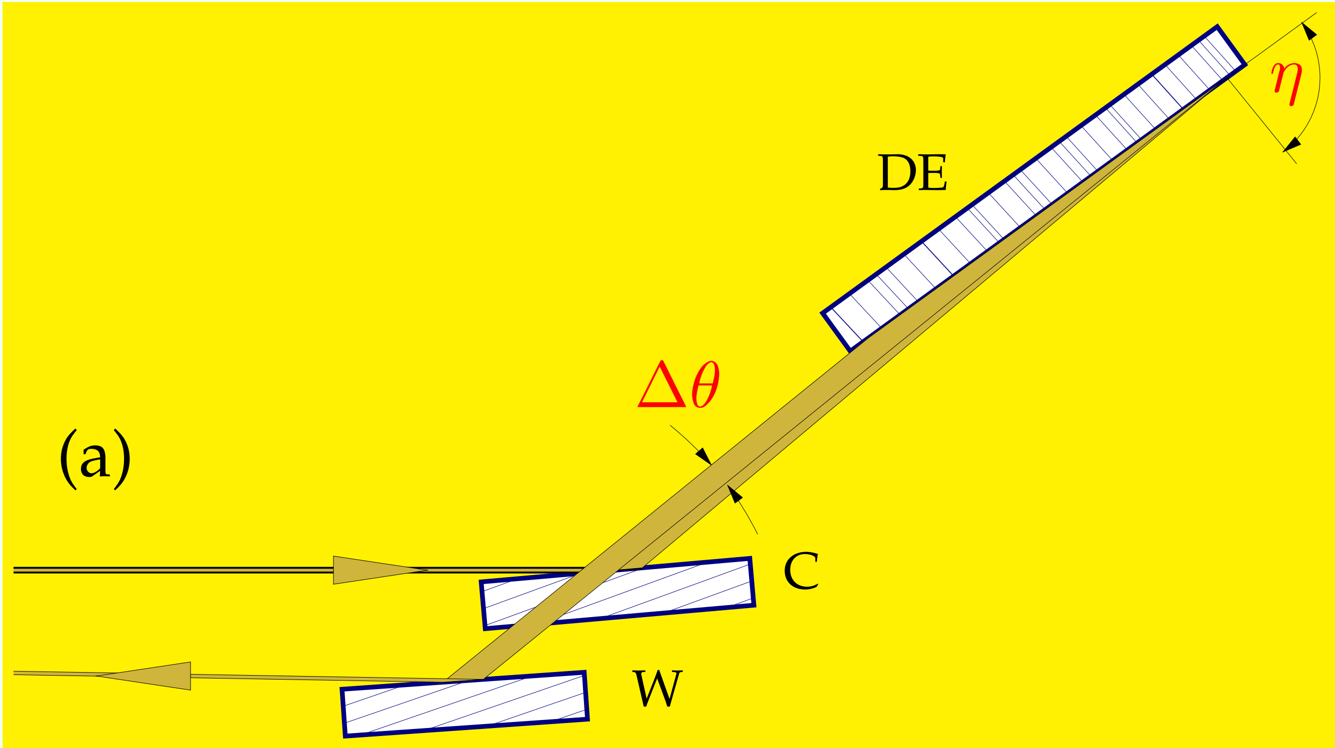
Spectral resolution of the $(+,+,\pm)$ monochromator

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



The smaller the photon energy E,

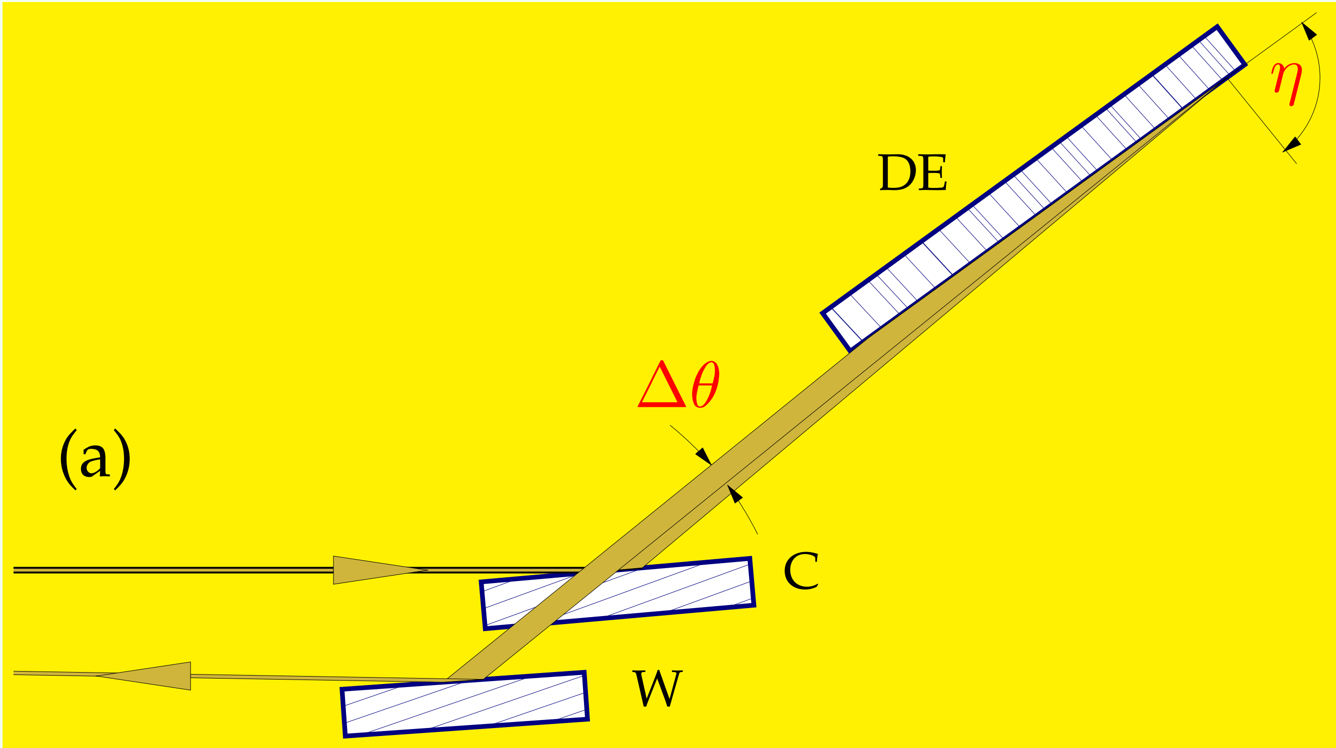
the smaller is the energy bandwidth ΔE (fortunately!).

Spectral resolution of the $(+,+,\pm)$ monochromator

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

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

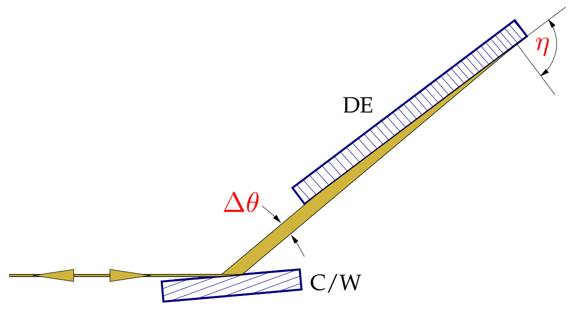
is feasible

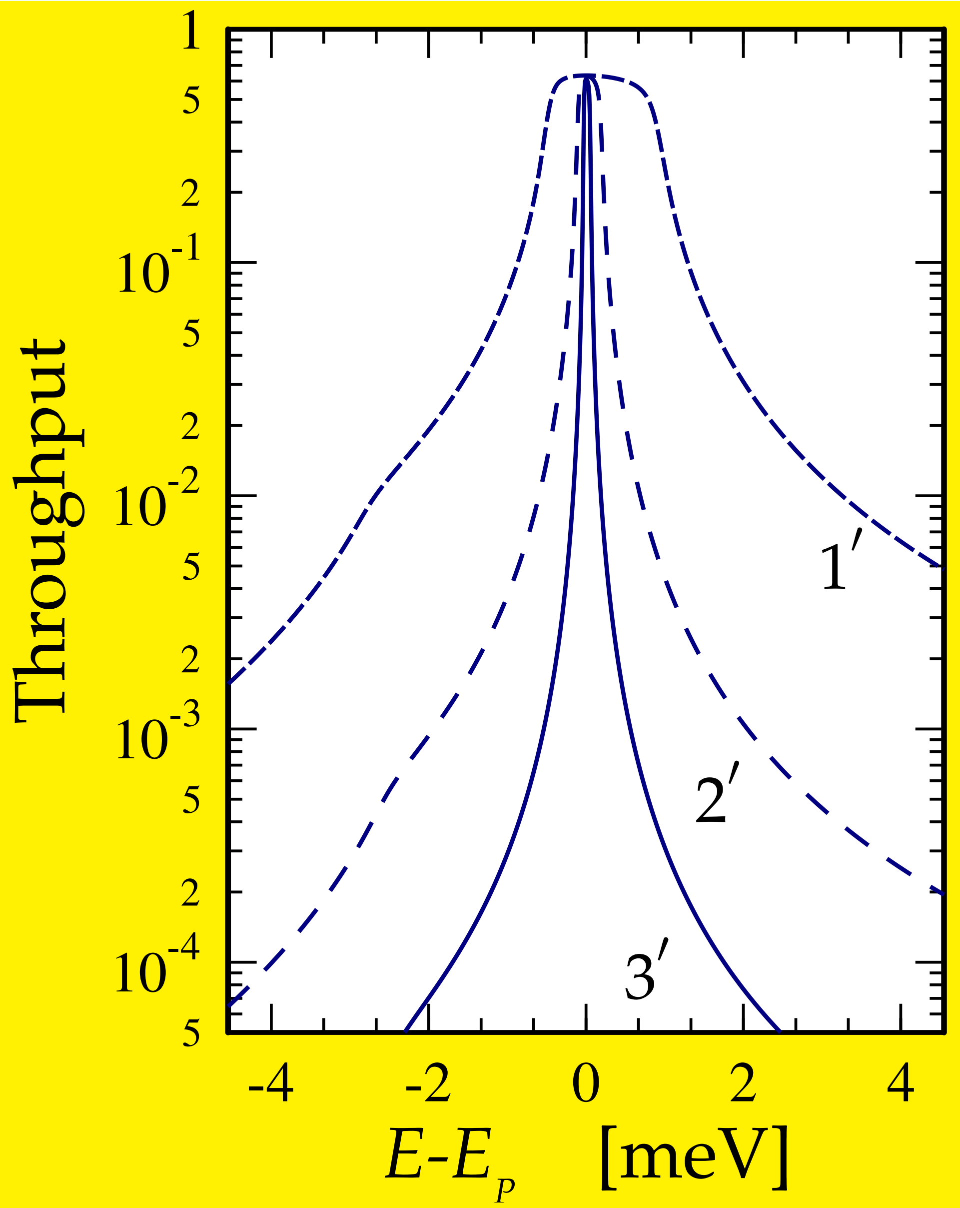


$E = 10$ **keV** $\Rightarrow \quad \Delta E = 10 - 0.1$ **meV**

$E = 5$ **keV** $\Rightarrow \quad \Delta E = 5 - 0.05$ **meV**

Throughput of the $(+,+,\pm)$ monochromator



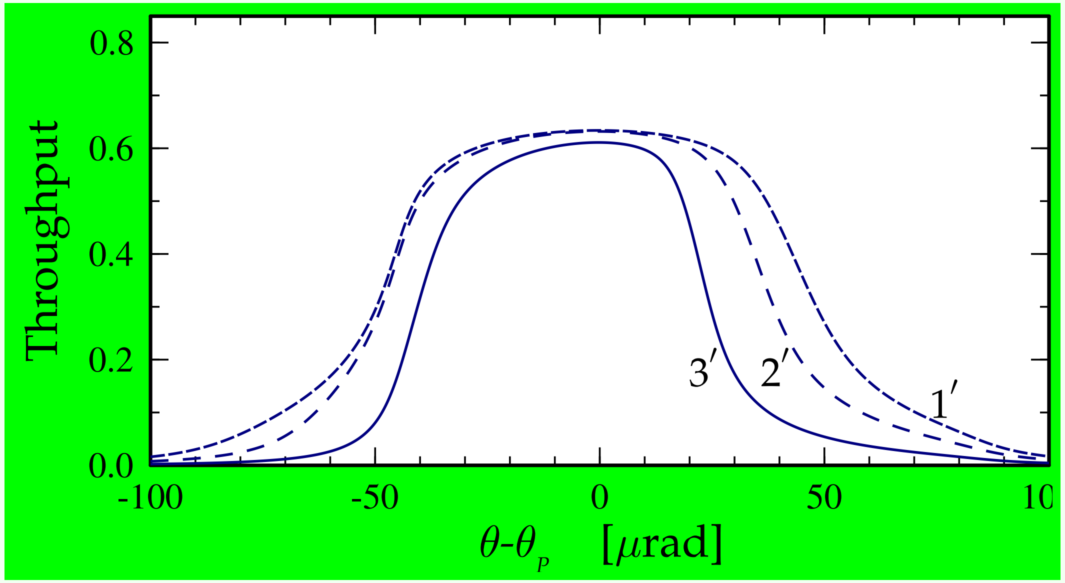


$E = 9.1$ keV

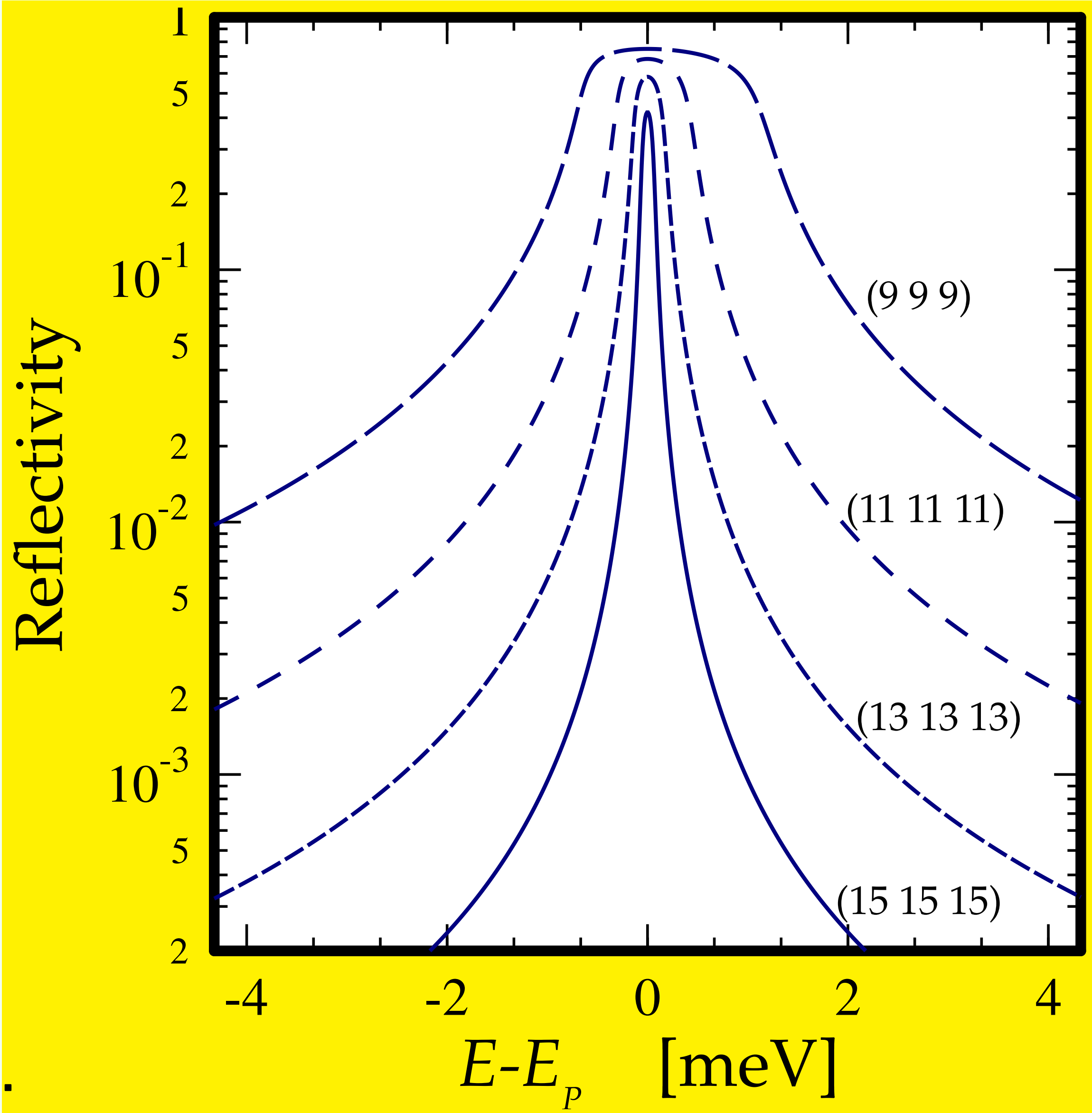
$1' : \Delta E = 1.5$ meV $(\eta = 85°)$

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

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



Energy dependence of the reflectivity in Si in backscattering



$(9\ 9\ 9)$, $E_P = 18$ *keV* :
$\Delta E = 1.9$ *meV*

$(11\ 11\ 11)$, $E_P = 22$ *keV* :
$\Delta E = 0.82$ *meV*

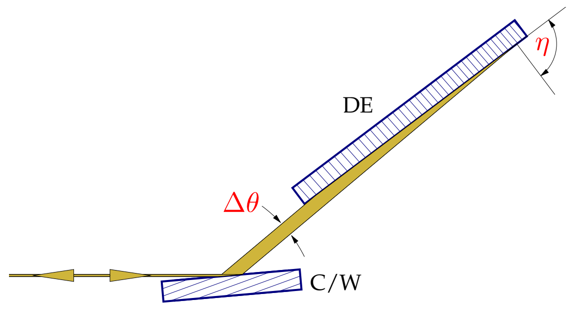
$(13\ 13\ 13)$, $E_P = 26$ *keV* :
$\Delta E = 0.35$ *meV*

$(15\ 15\ 15)$, $E_P = 30$ *keV* :
$\Delta E = 0.15$ *meV*

ΔE *decreases with increasing* E_P

..., but, also the reflectivity drops ...
from 0.74 *to* 0.42*, respectively.*

..



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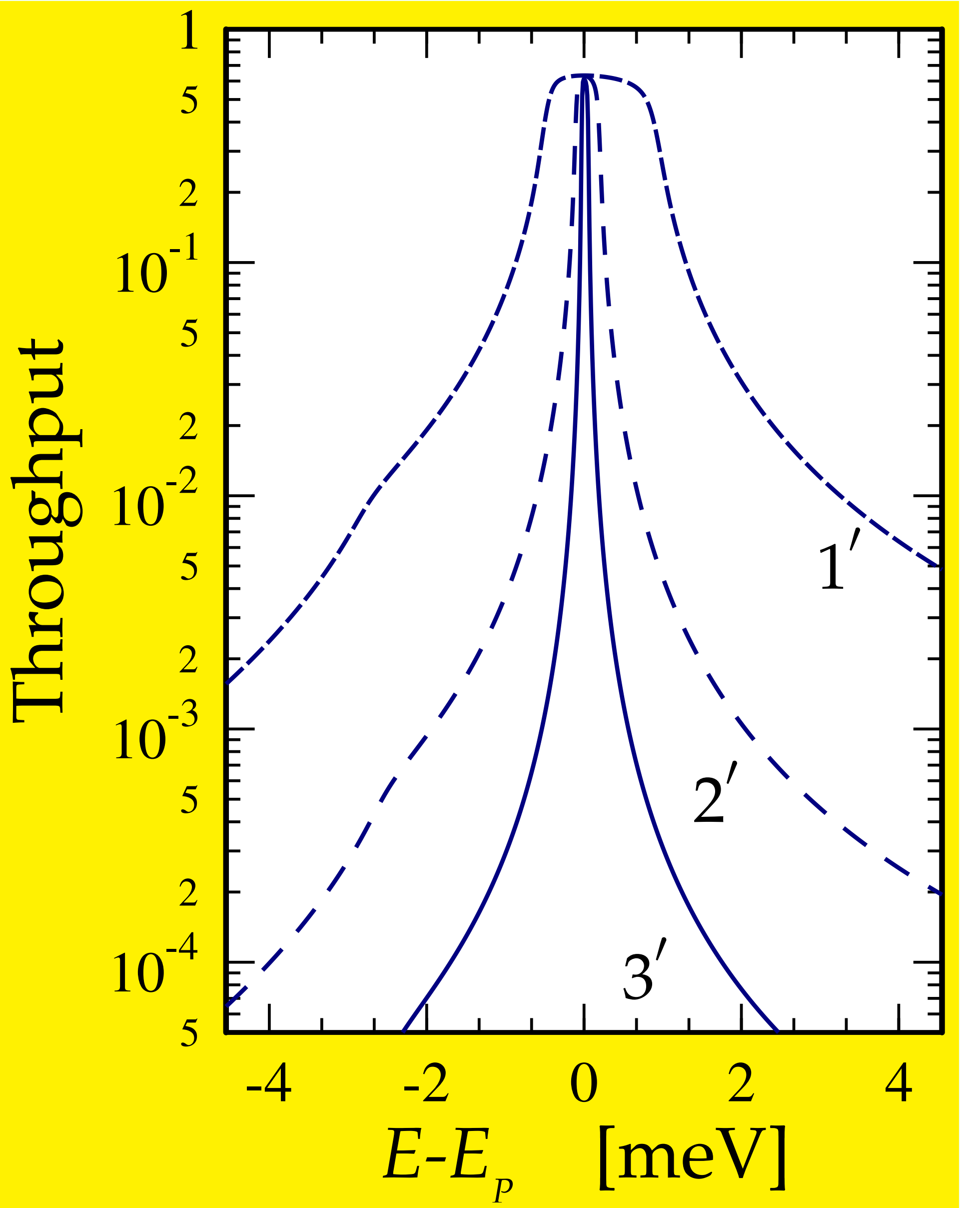

$E = 9.1$ **keV**

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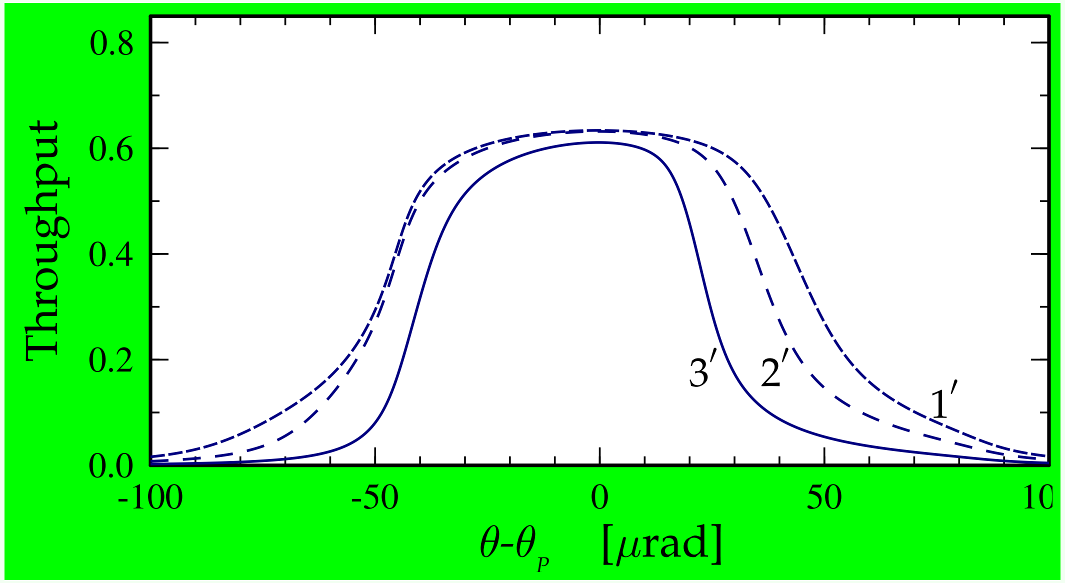
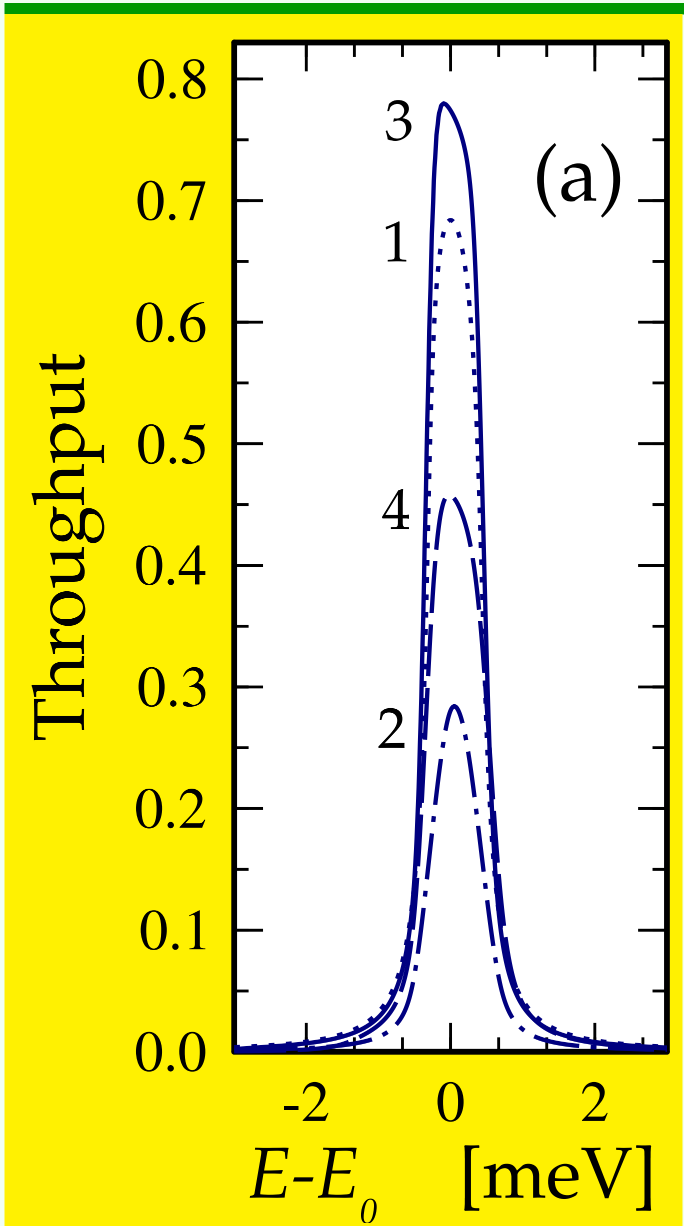
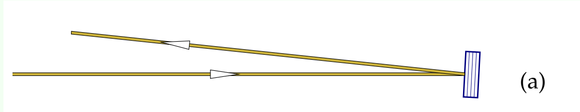


Spectral functions of different types of monochromators



1: single-bounce

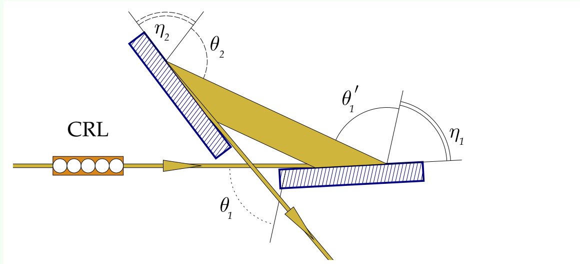
$\Delta E = 0.82$ meV
$E_0 = 21.7$ keV



Graef, Materlik (1982)
Verbeni et al. (1996)

2: $(+,+)$

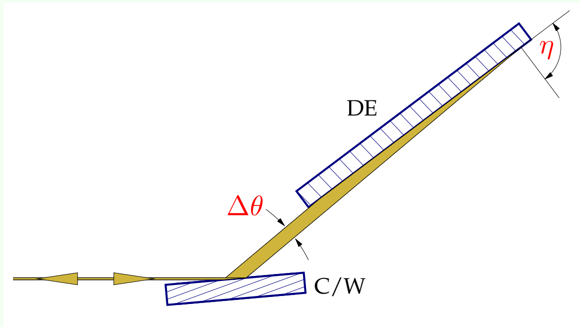
$\Delta E = 0.80$ meV
$E_0 = 14.4$ keV



Chumakov et al. (1996)
Toellner et al. (1997)

3: $(+,+,\pm)$

$\Delta E = 0.83$ meV
$E_0 = 9.1$ keV



Shvyd'ko (2004)

4: $(+,-,-,+)$

$\Delta E = 0.93$ meV
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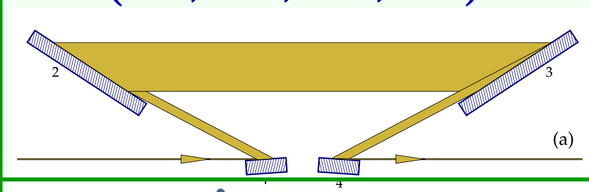

Yabashi et al. (2001)
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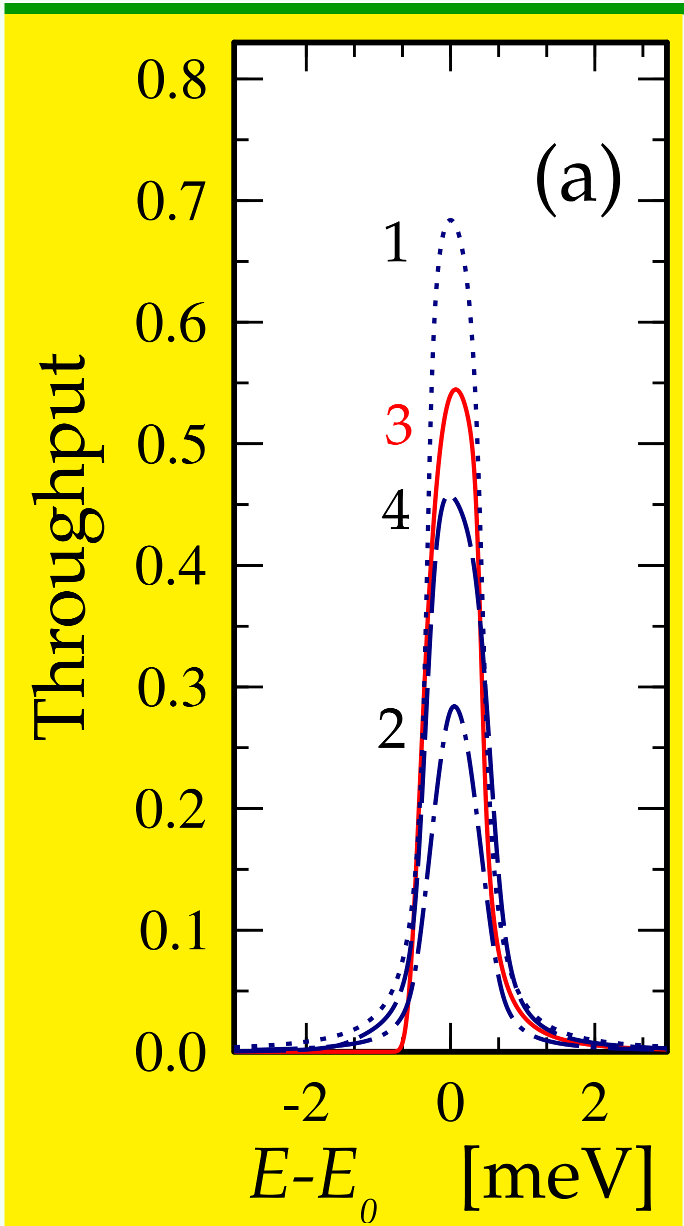

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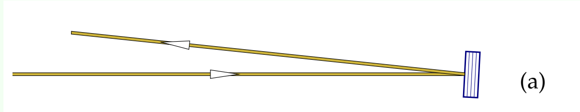

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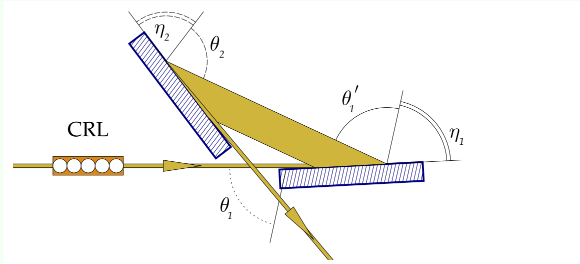

Chumakov et al. (1996)
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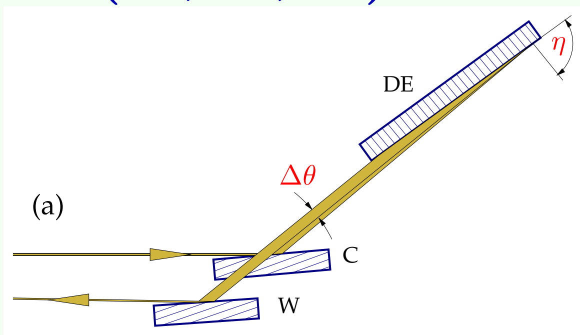

Shvyd'ko (2004)

4: (+, −, −, +)

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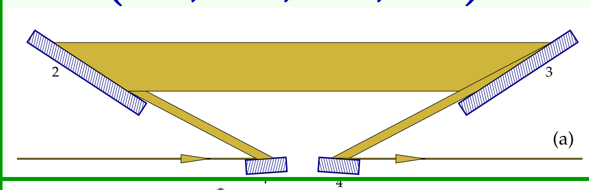

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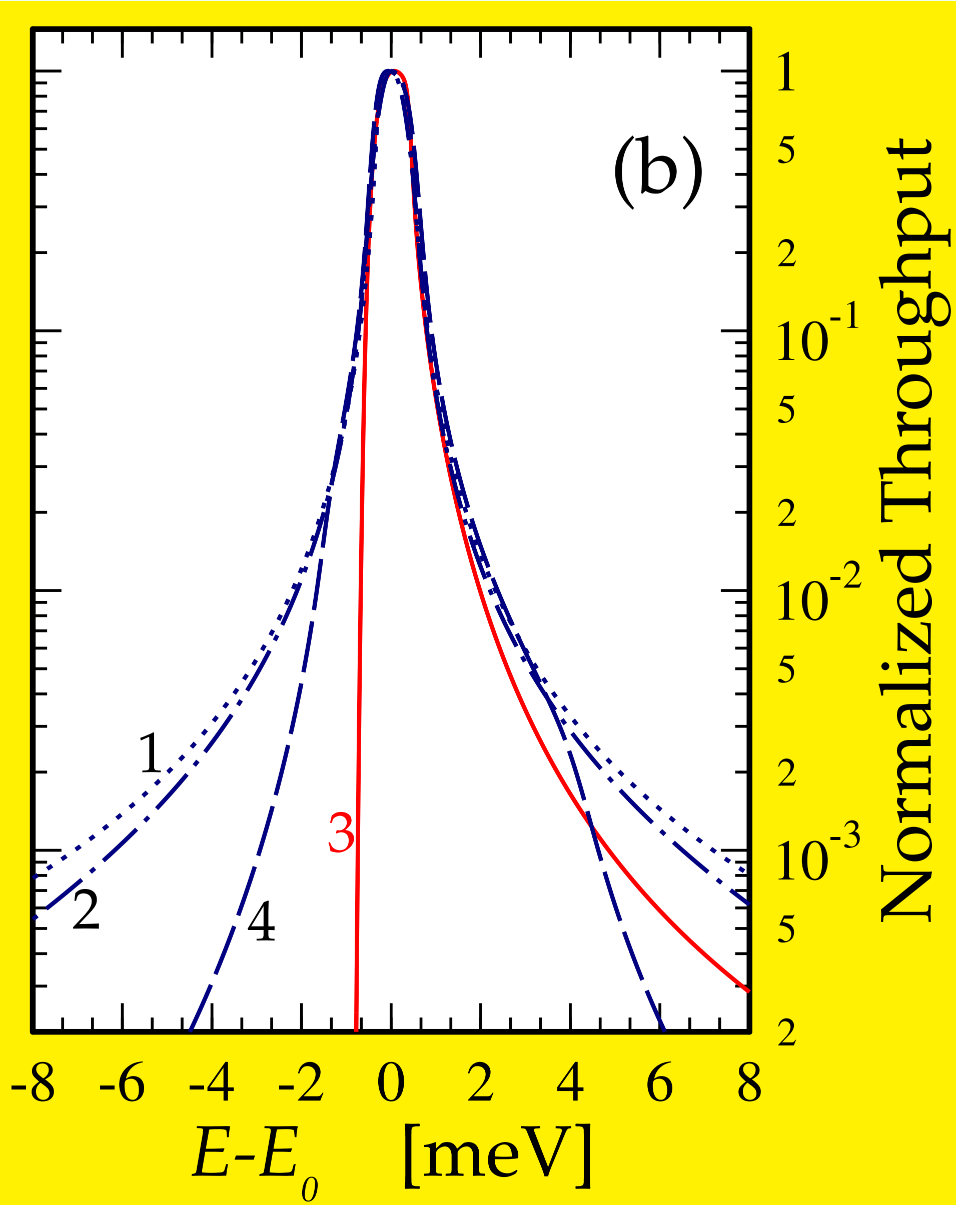

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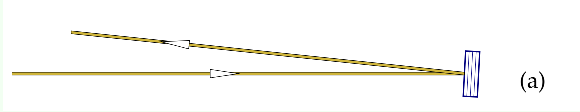

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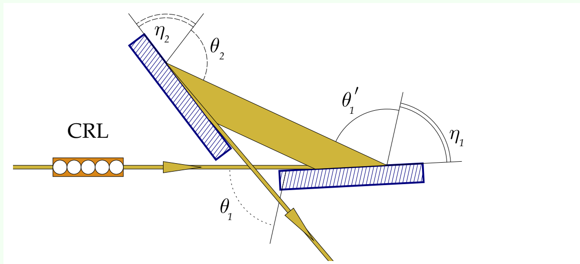

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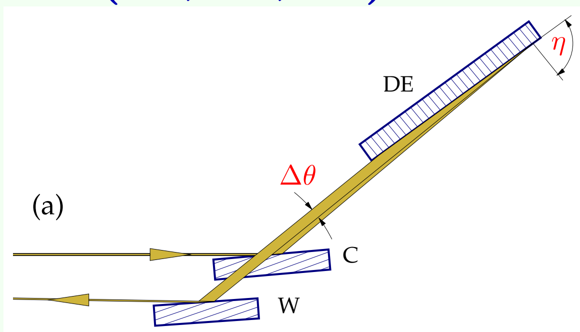

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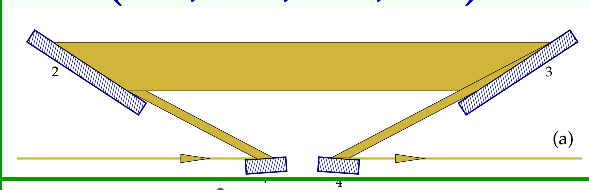

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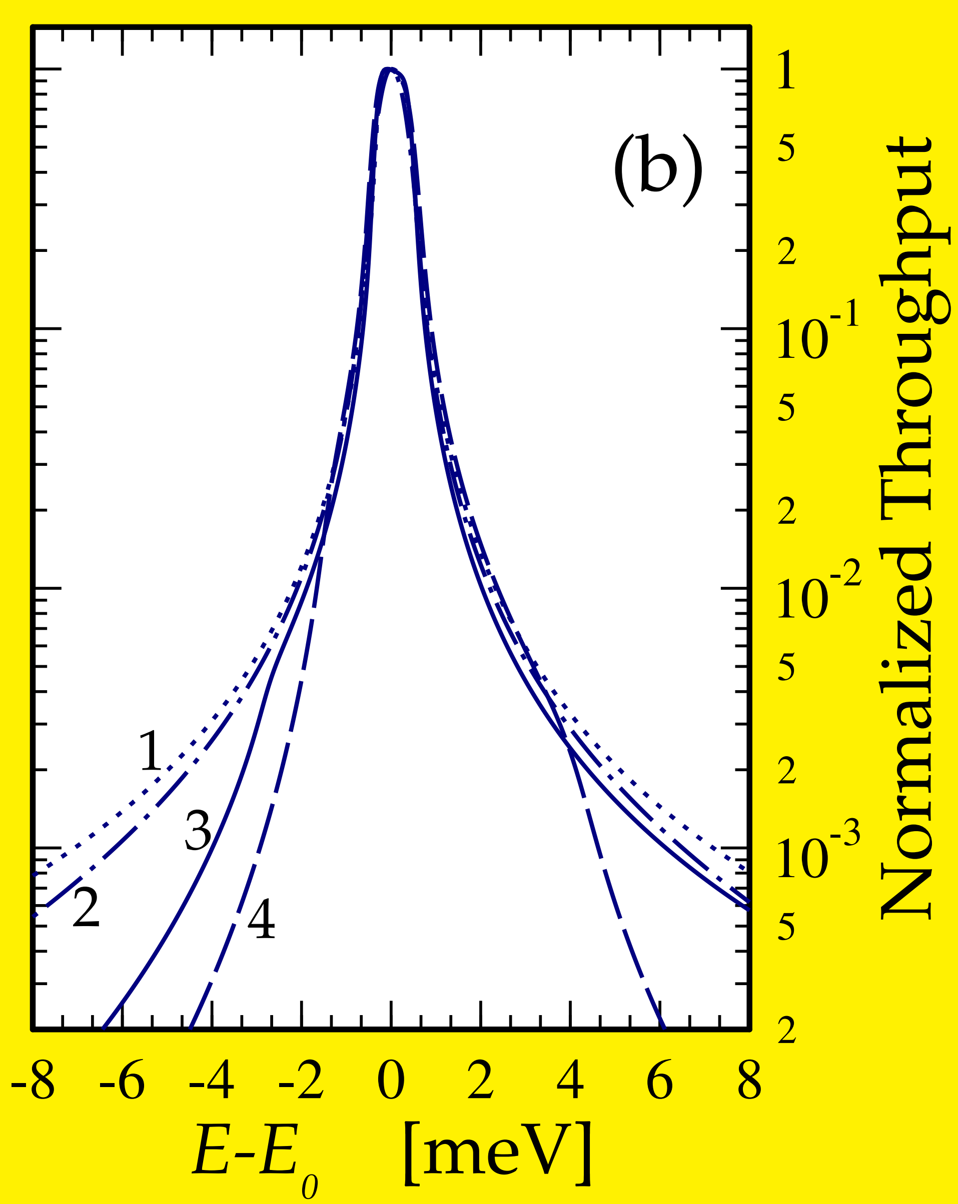

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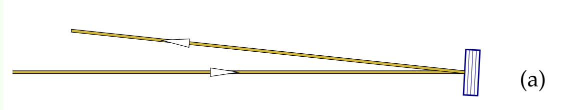
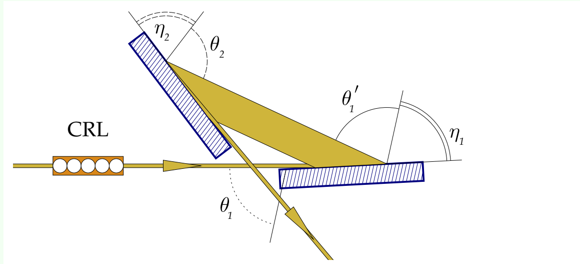


Graef, Materlik (1982)
Verbeni et al. (1996)

2: (+, +) $\Delta E = 0.80$ meV, $E_0 = 14.4$ keV



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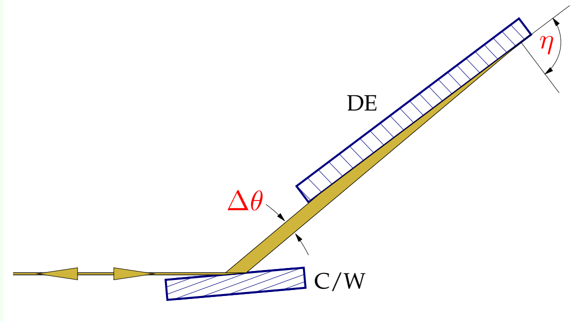

Shvyd'ko (2004)

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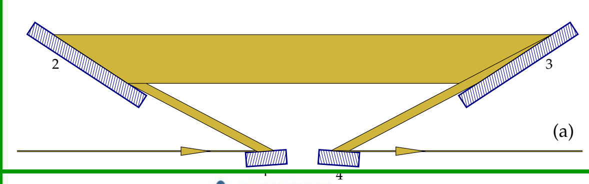
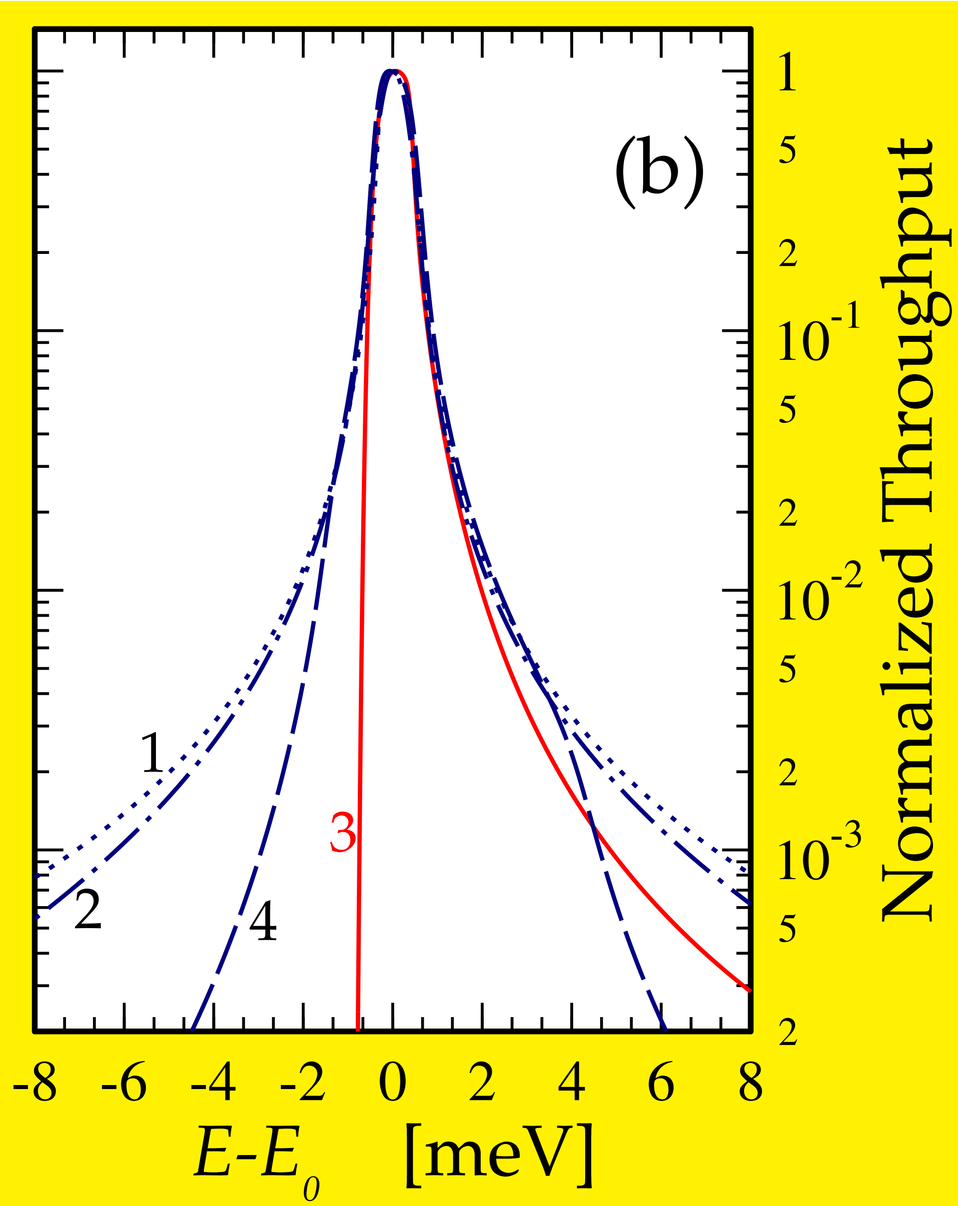


Yabashi et al. (2001)
Toellner et al. (2001)

Spectral functions of different types of monochromators



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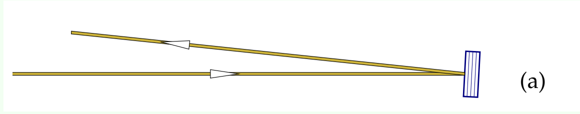

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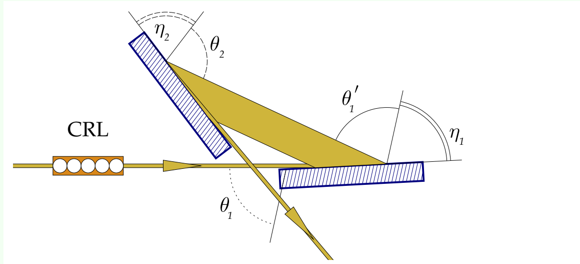

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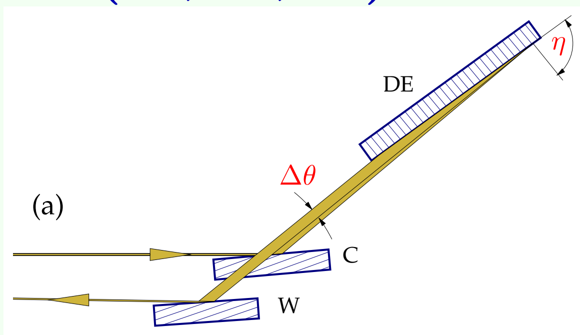

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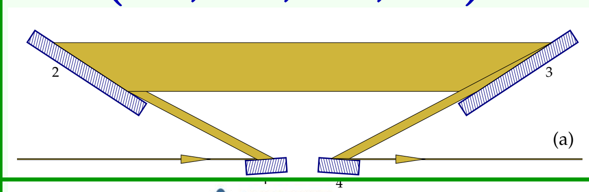

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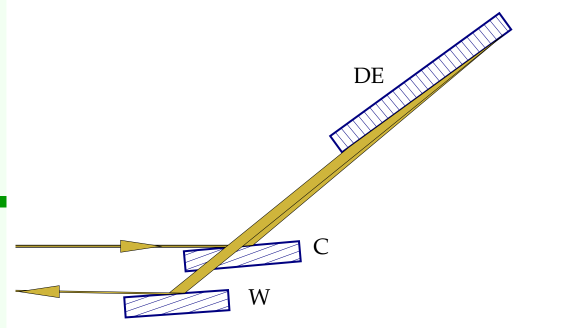
Features of the $(+,+,\pm)$ monochromators



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.

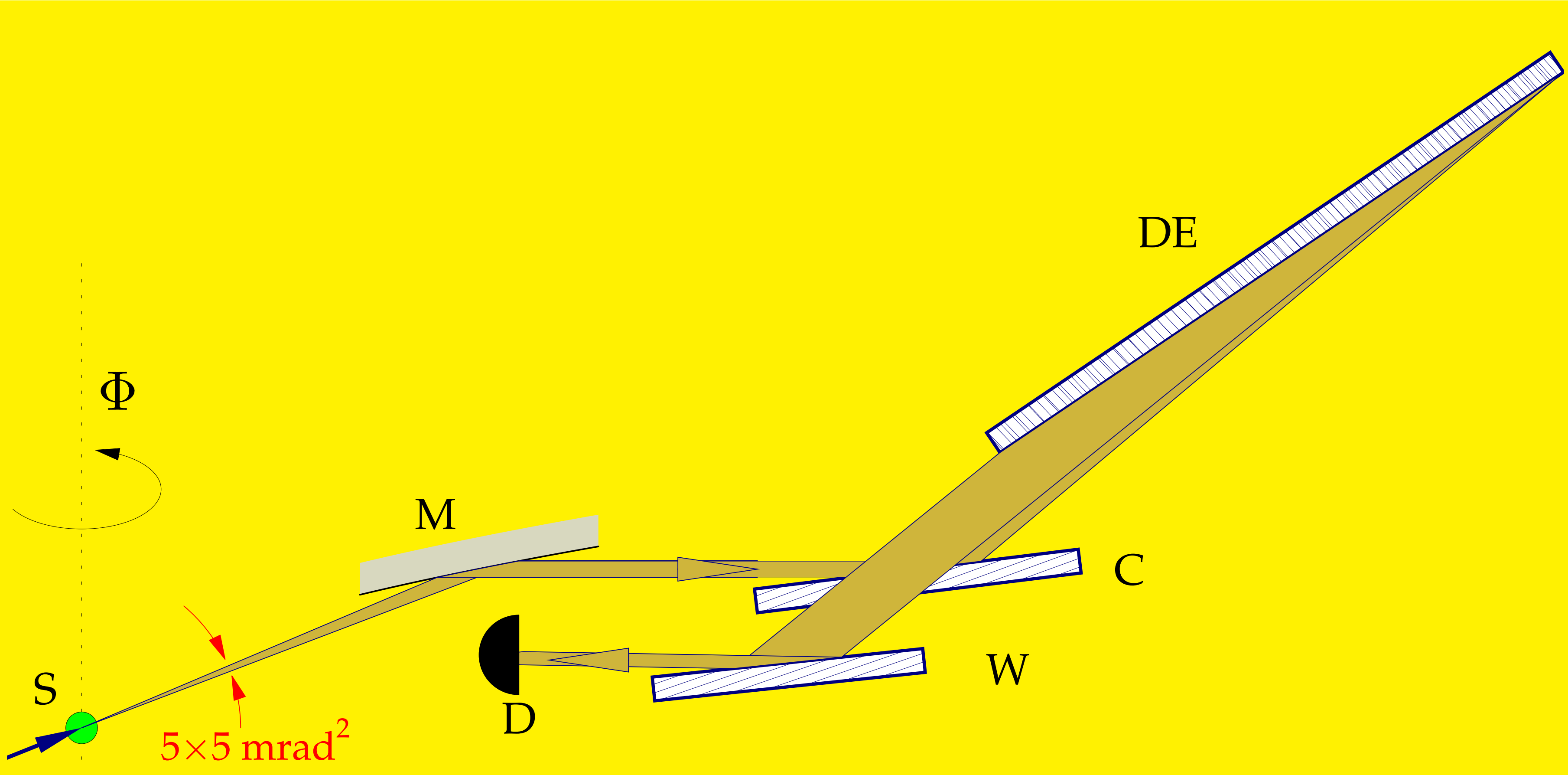
3. ΔE can be varied by changing η (E is fixed).

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).

$(+,+,\pm)$ Analyzer



M - paraboloidal mirror

DE - dispersing element

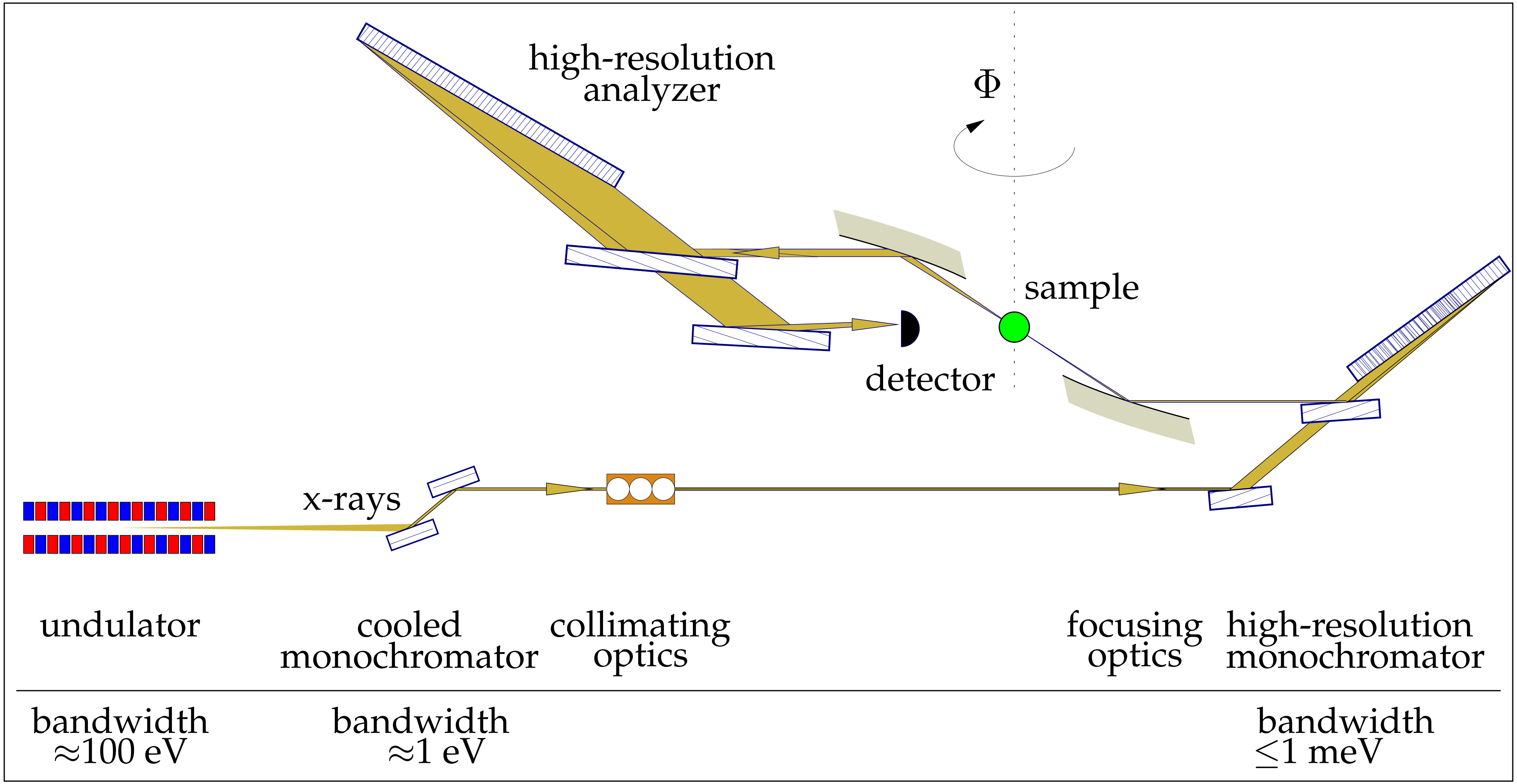
D - detector

S - sample

W - wavelength selector

C - collimator

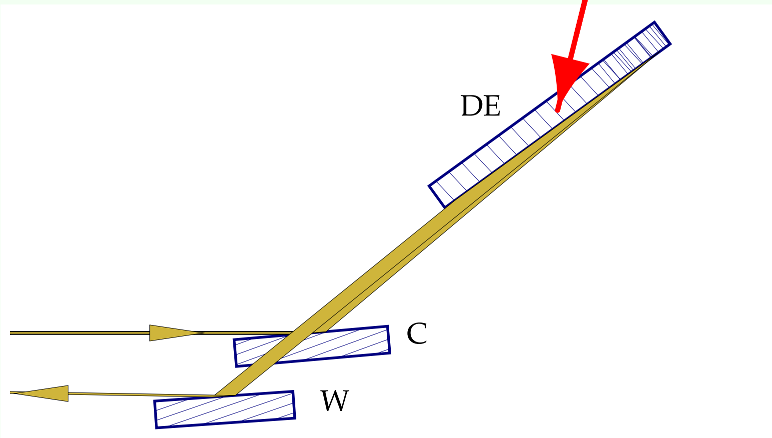
$(+,+,\pm)$ IXS spectrometer (1)



Energy tuning of the $(+,+,\pm)$ monochromator

... by changing the temperature of the dispersing element (DE)

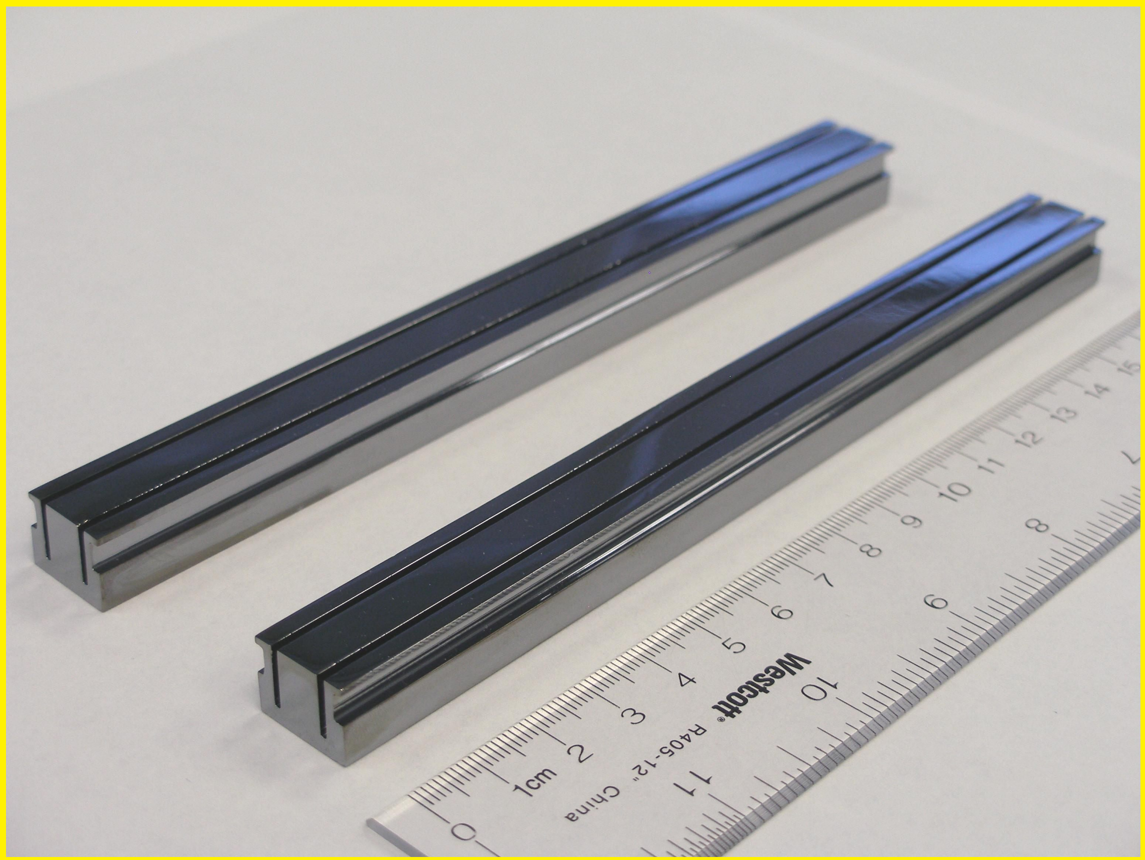
Bragg back-reflection Si(008) @ 9.13 keV

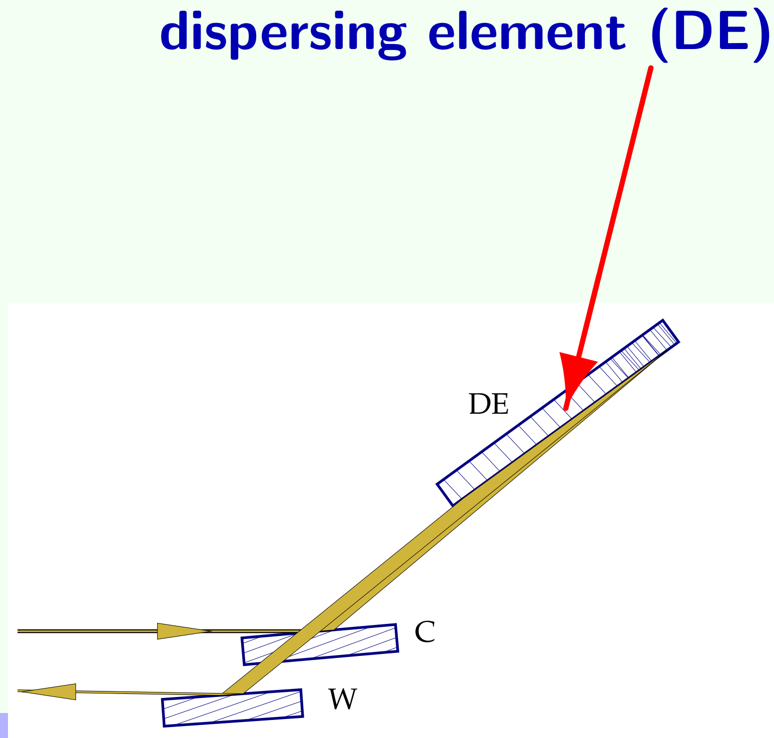


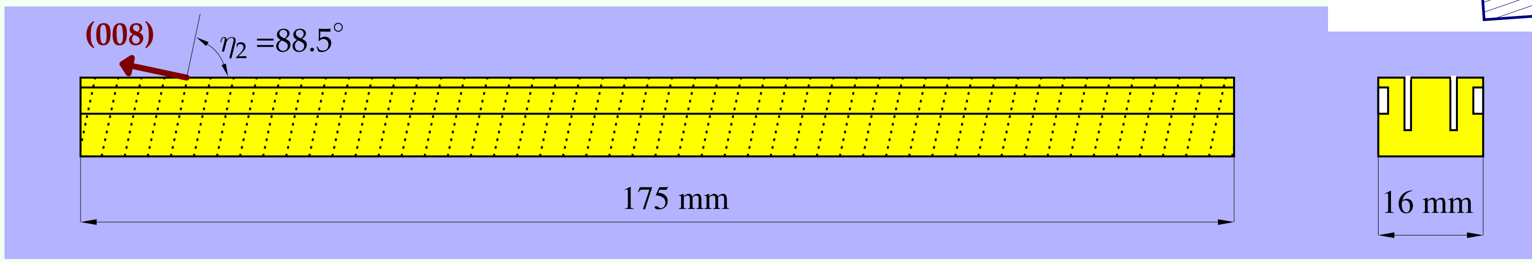
dE/dT=-23 meV/K

1 meV $\Rightarrow$ 43 mK

Dispersing Silicon Crystal Elements = Long Crystals







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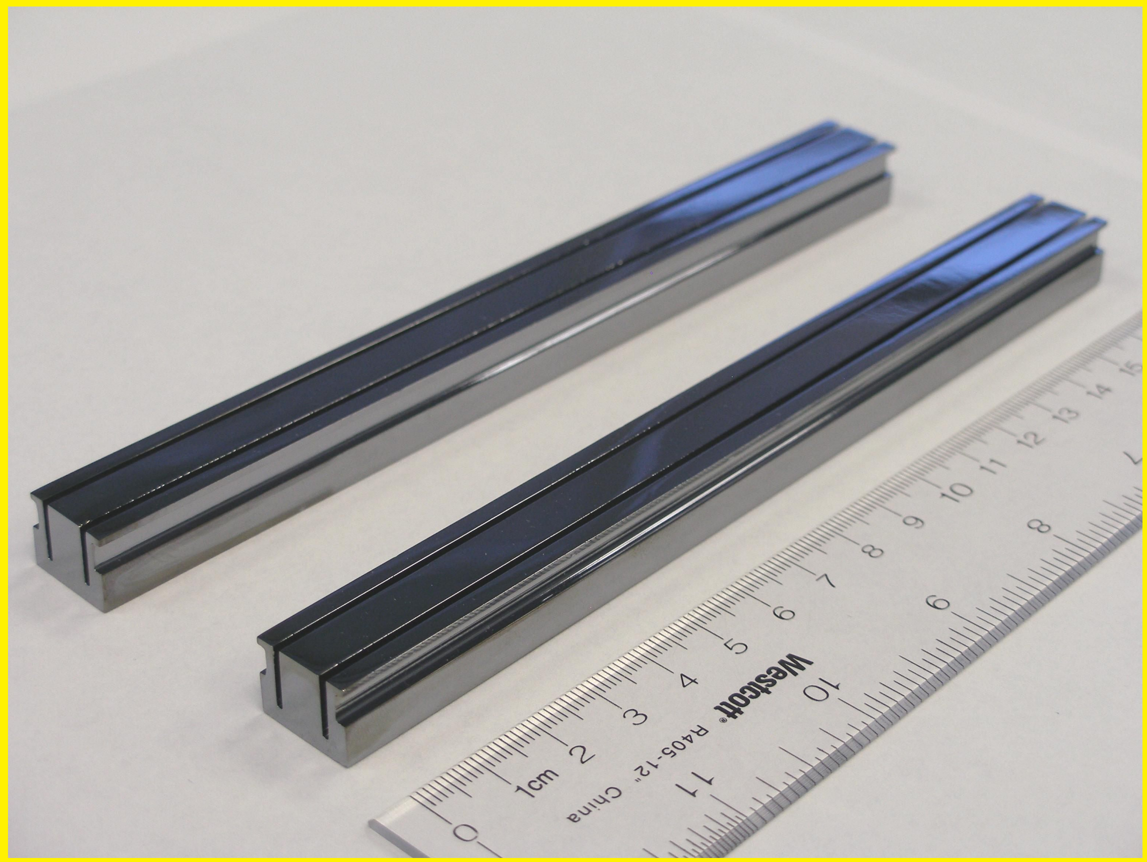
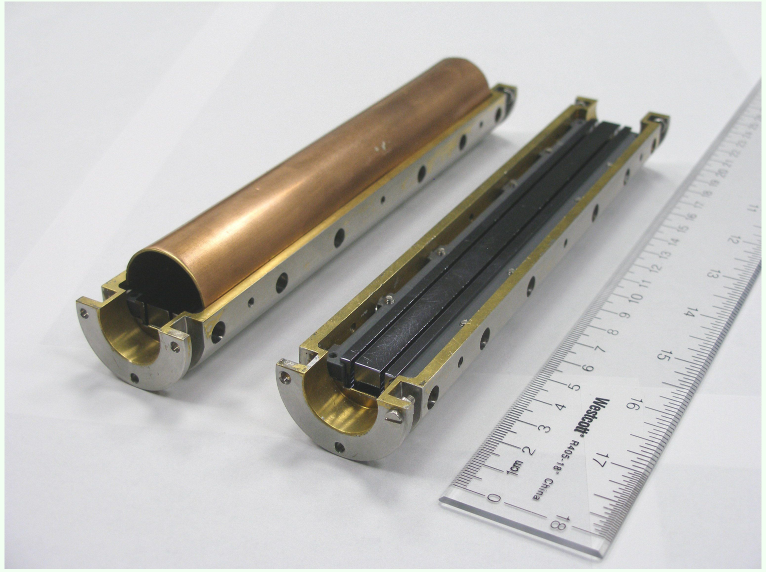
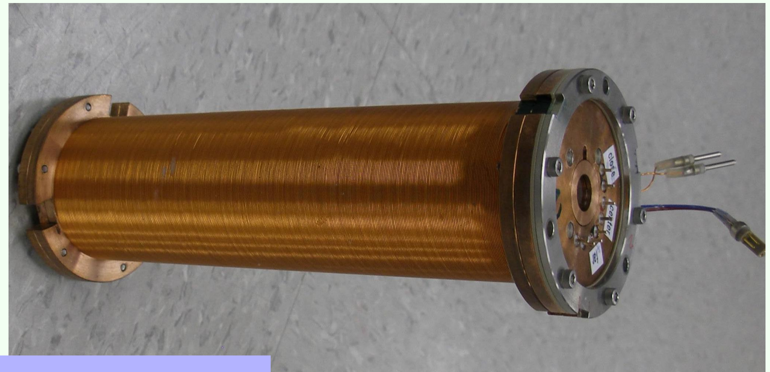
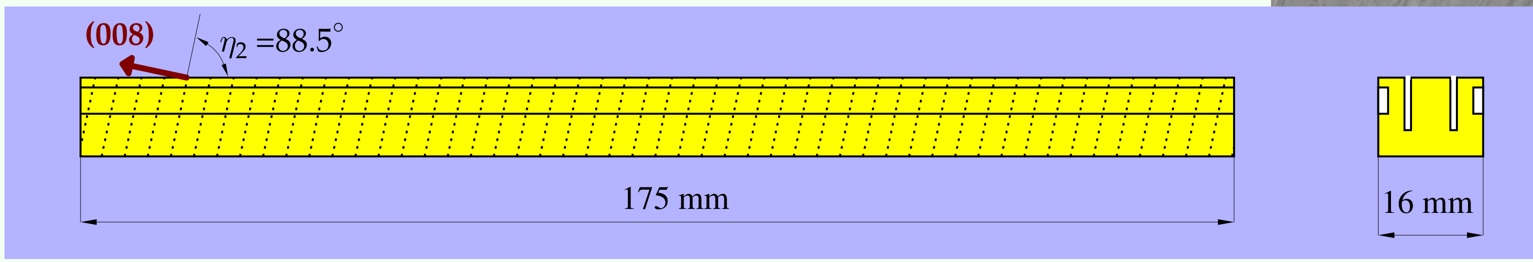
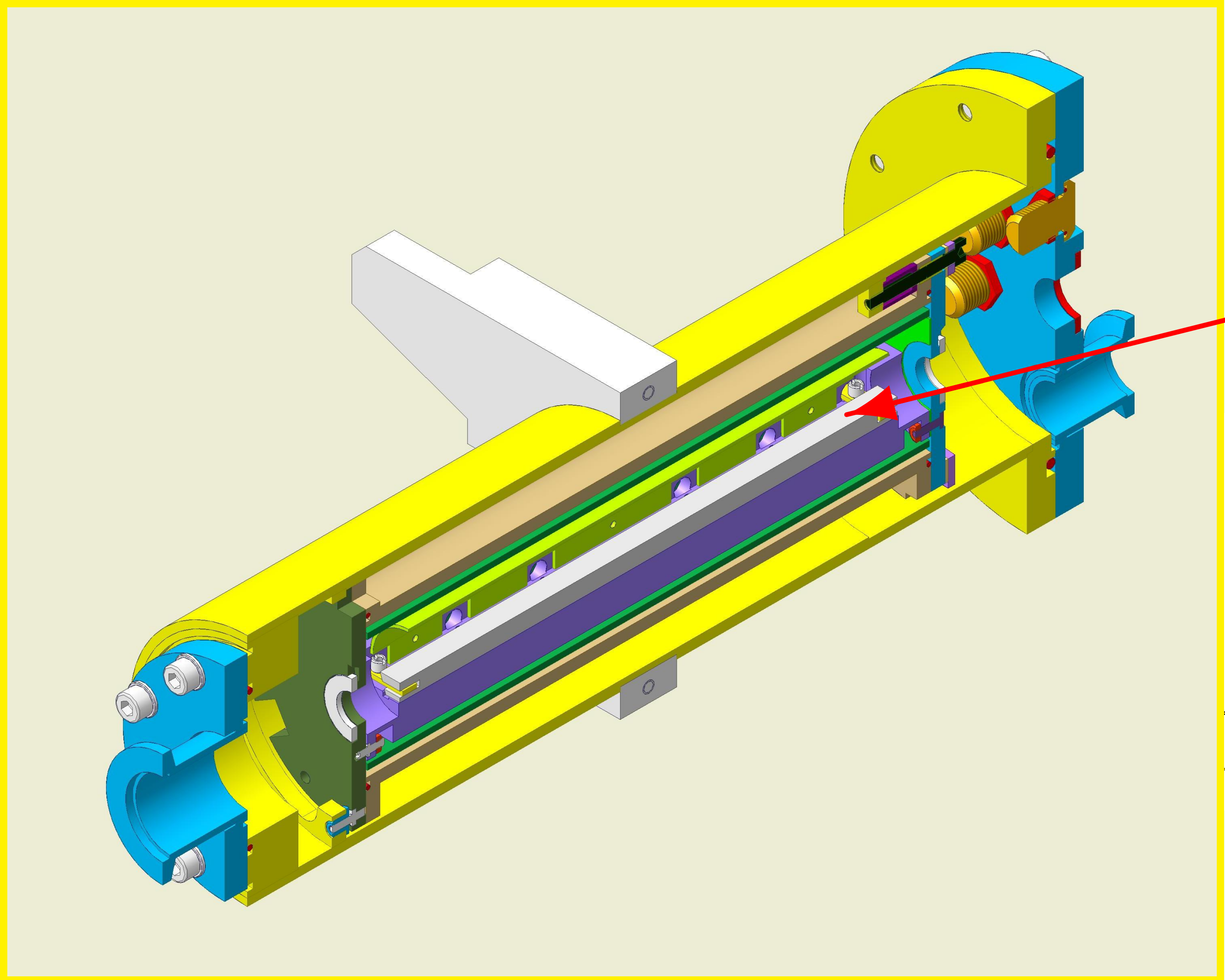




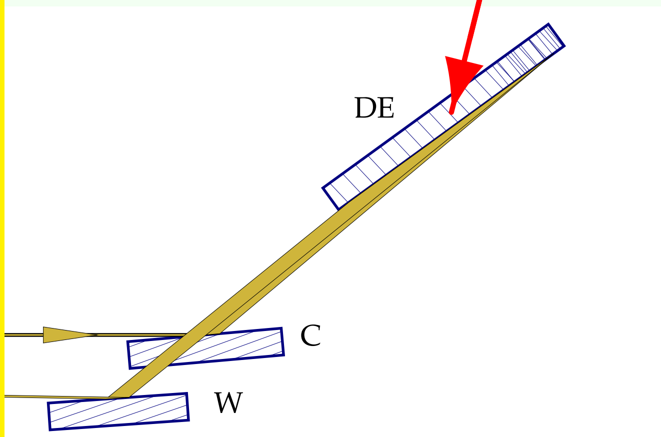




Energy tuning of the $(+,+,\pm)$ monochromator



... by changing the temperature of the dispersing element (DE)



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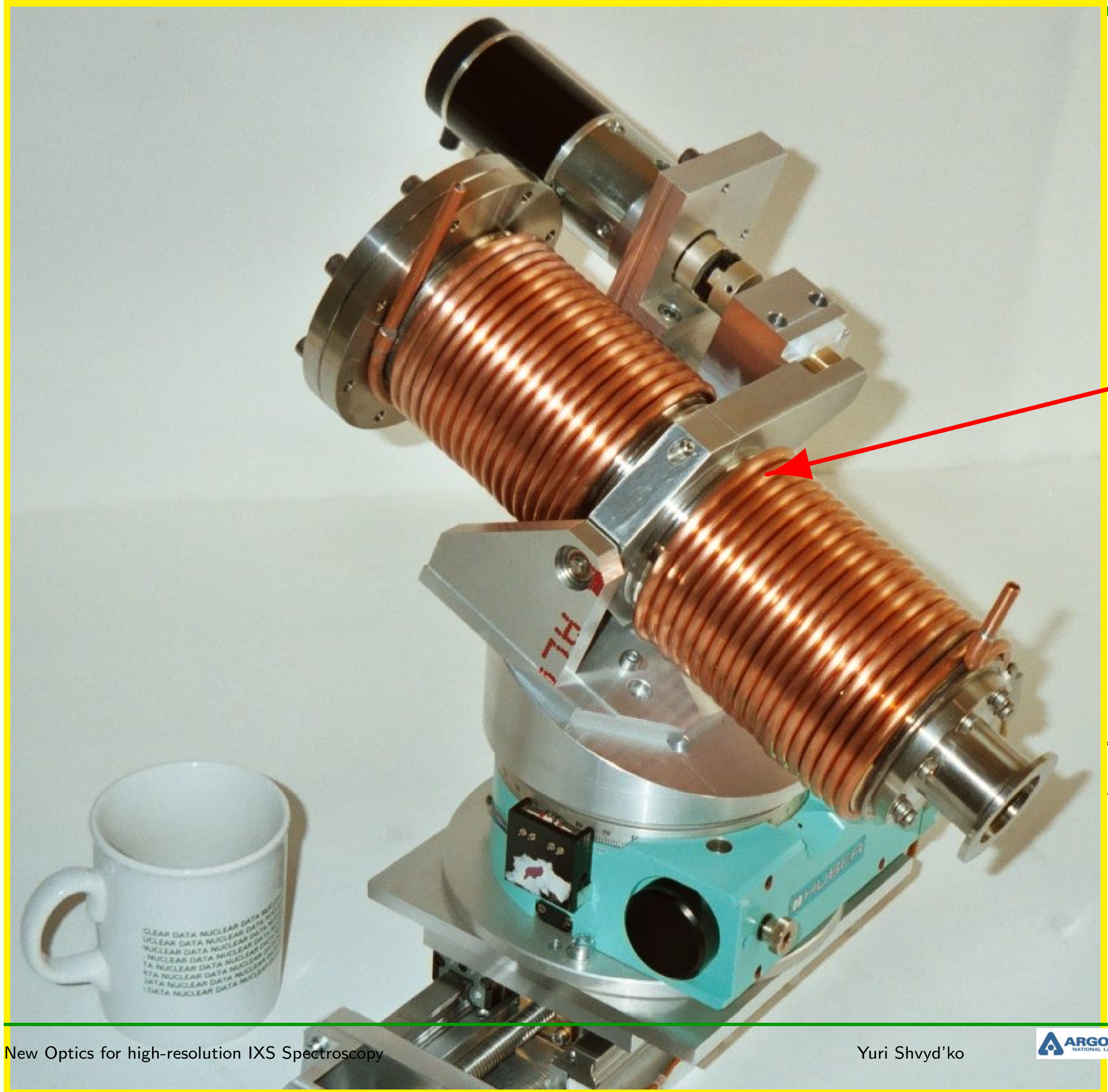

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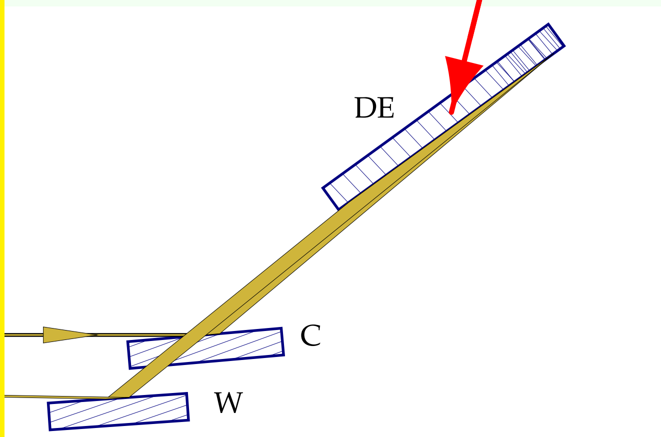

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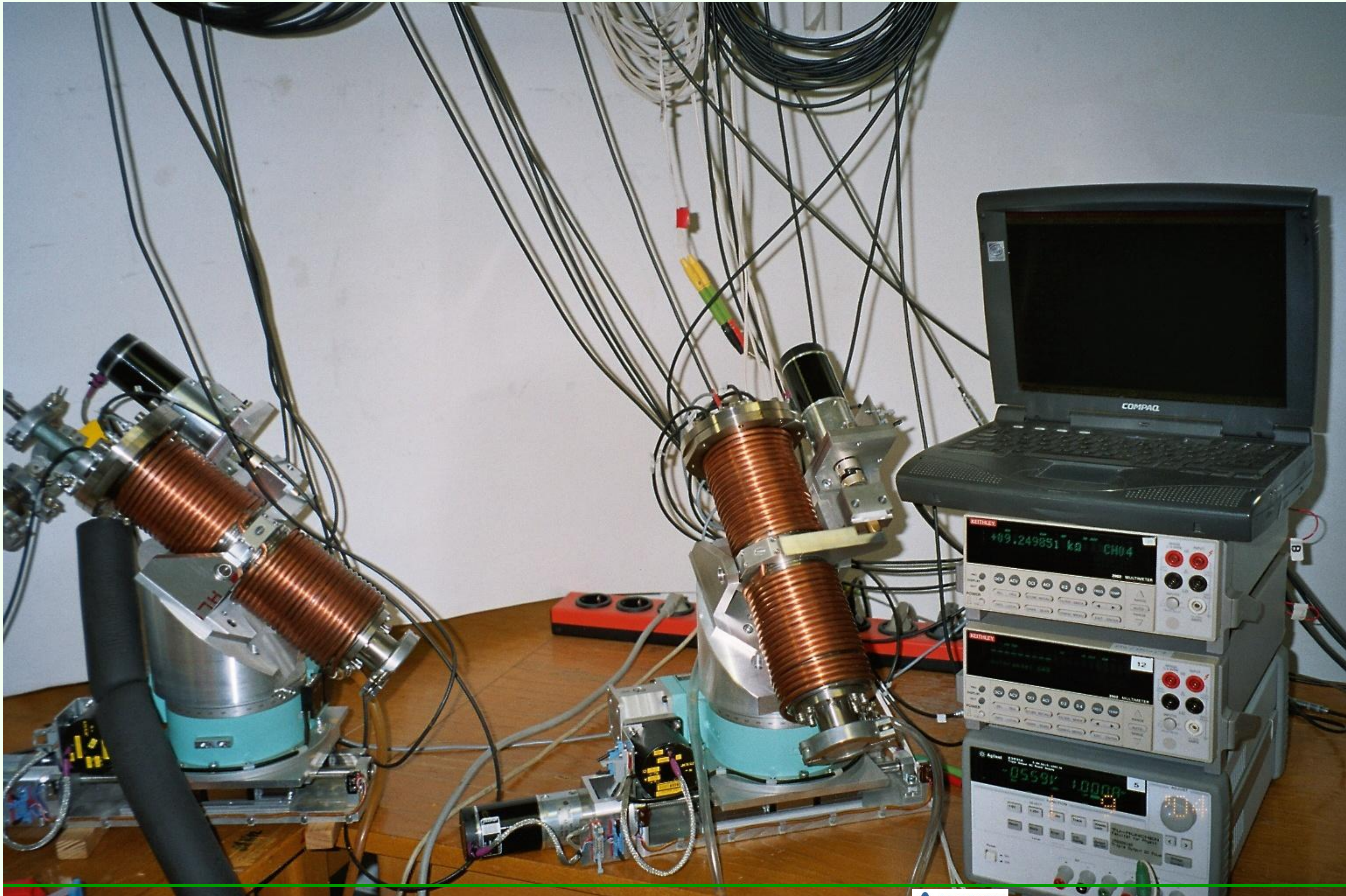
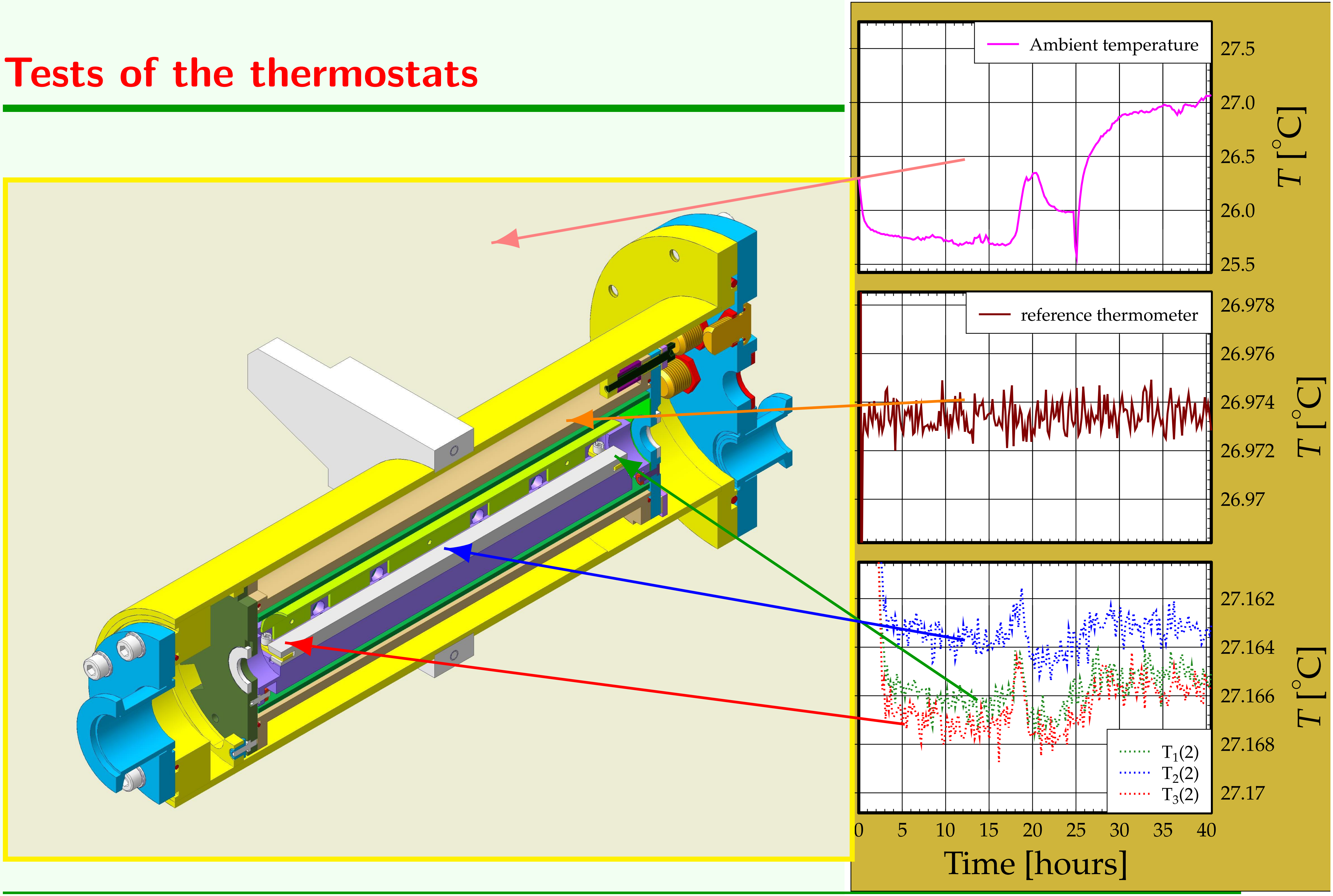
Thermostats for long crystals



Tests of the thermostats



Collimator and Wavelength-Selector (C-W)

Bragg reflection: **Si(220)**

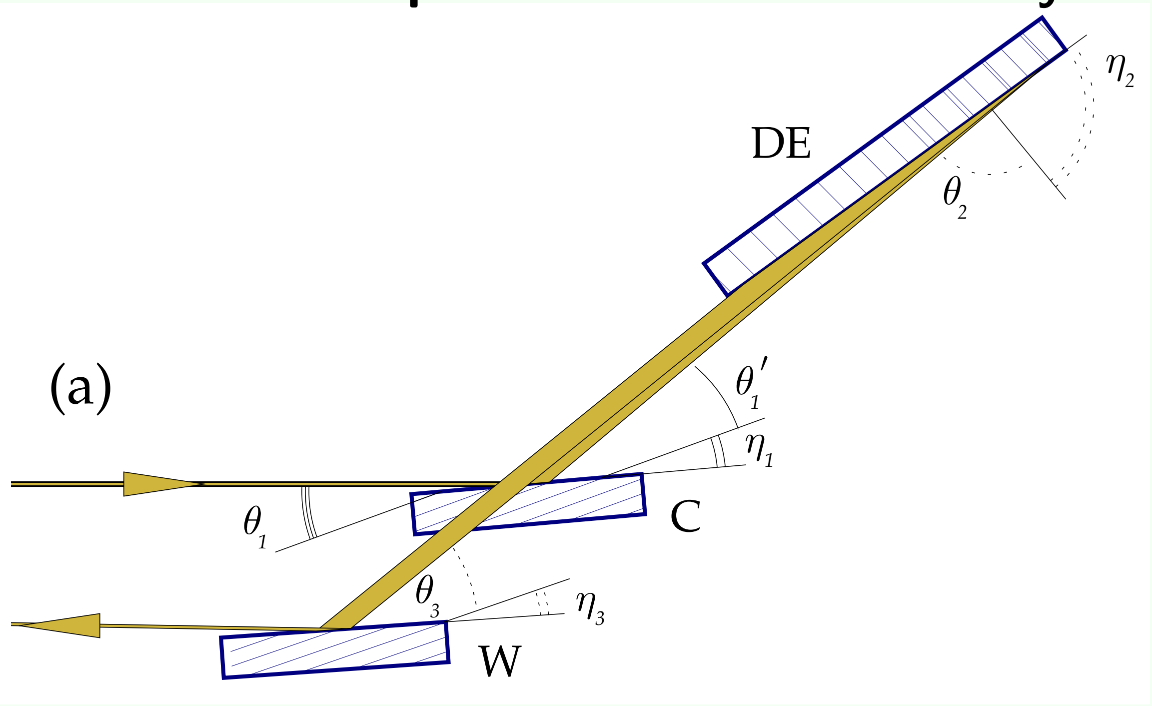
Asymmetry angles: $\eta_1 = \eta_3 = 19^\circ$

Angular acceptance: $\Delta\theta_1 = 106\ \mu\text{rad}$

Angular divergence: $\Delta\theta_1' = 5\ \mu\text{rad}$

Angular acceptance: $\Delta\theta_3 = 5\ \mu\text{rad}$

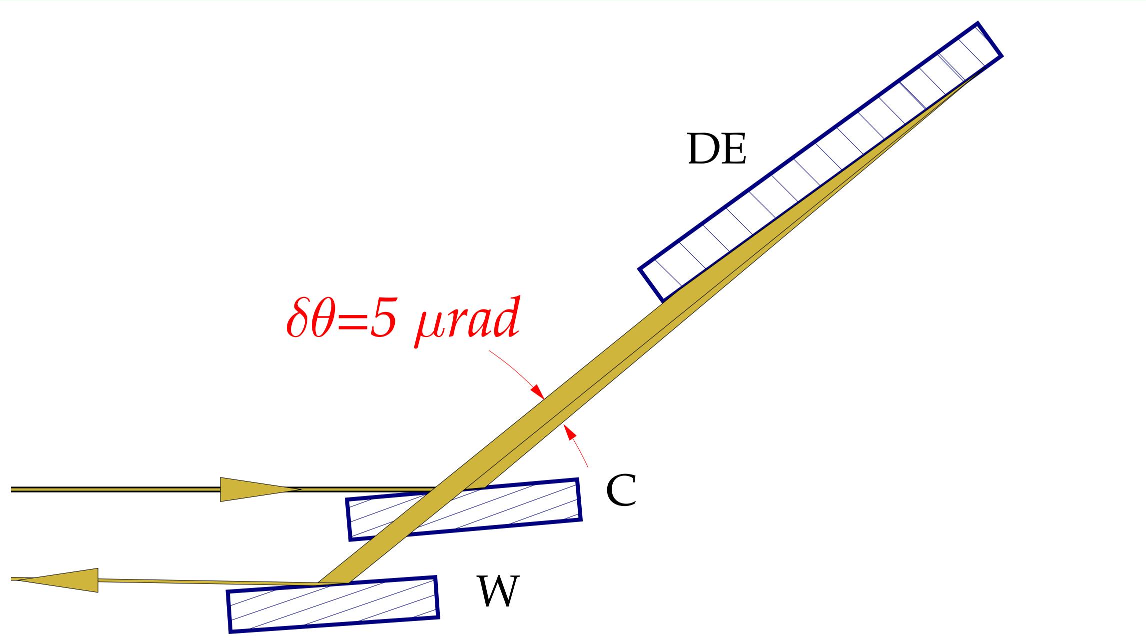
Problem: **absorption in the collimator crystal**

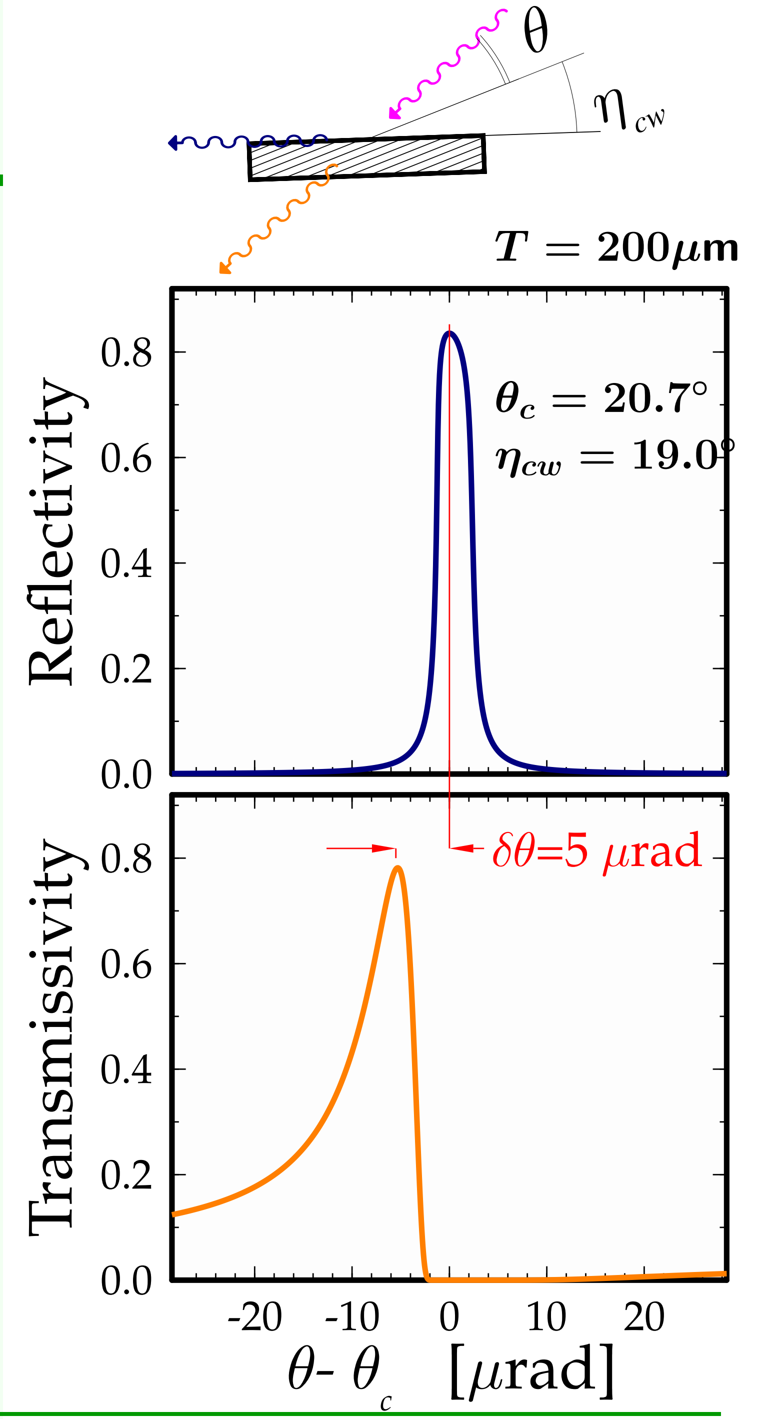


Collimator and Wavelength-Selector

Problem: absorption in the collimator crystal

Solution: penetration in the anomalous transmission mode





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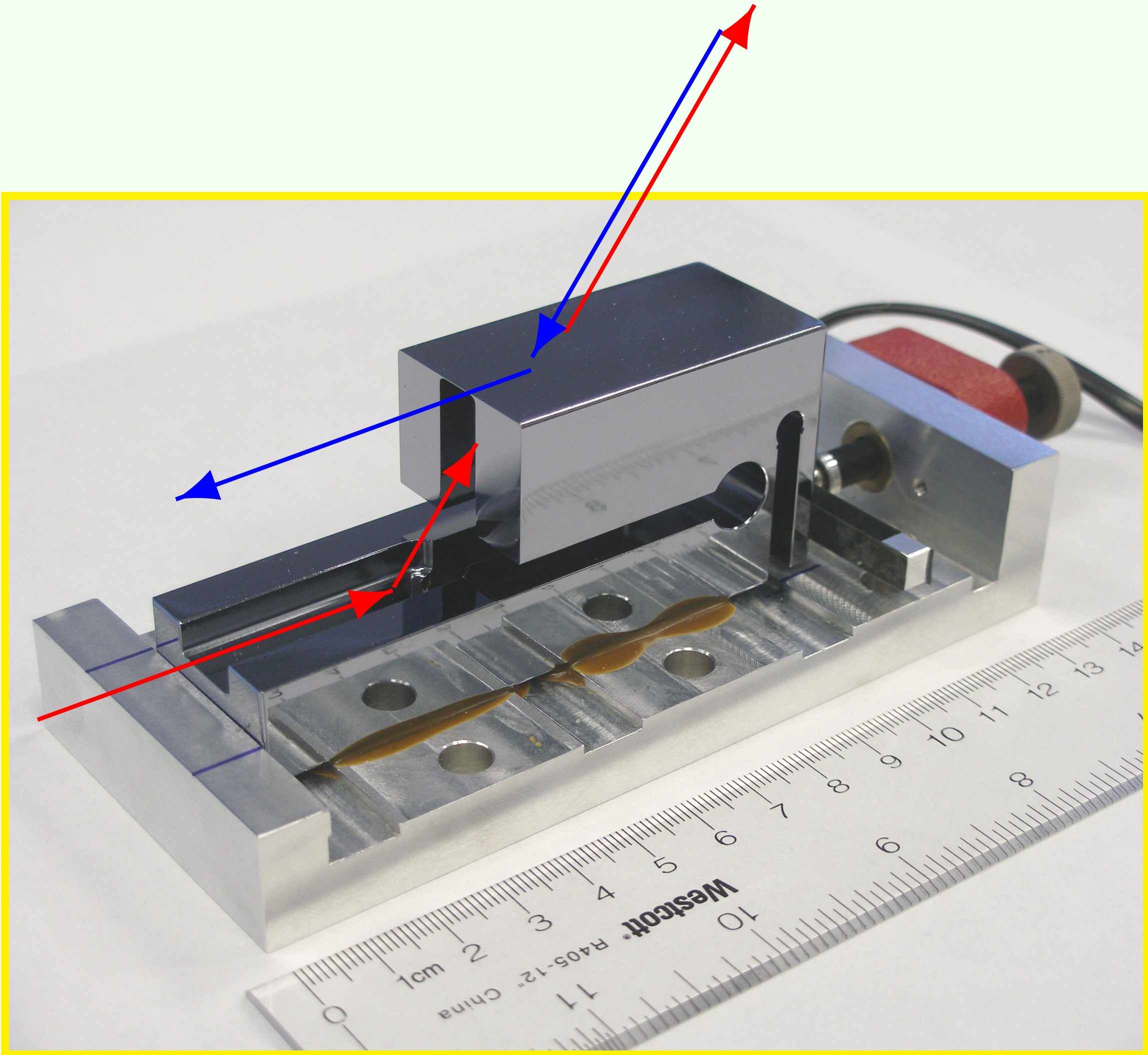

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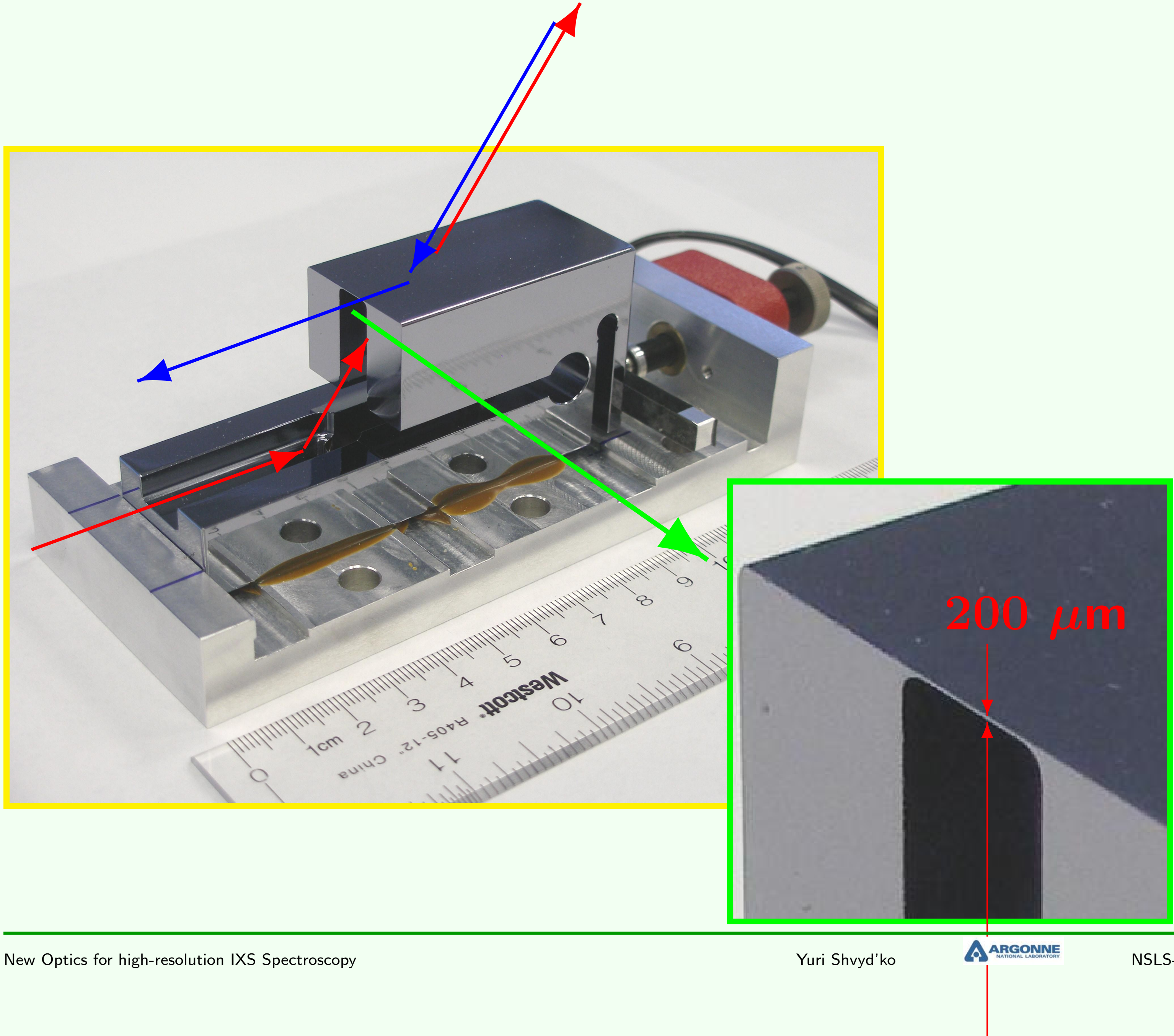

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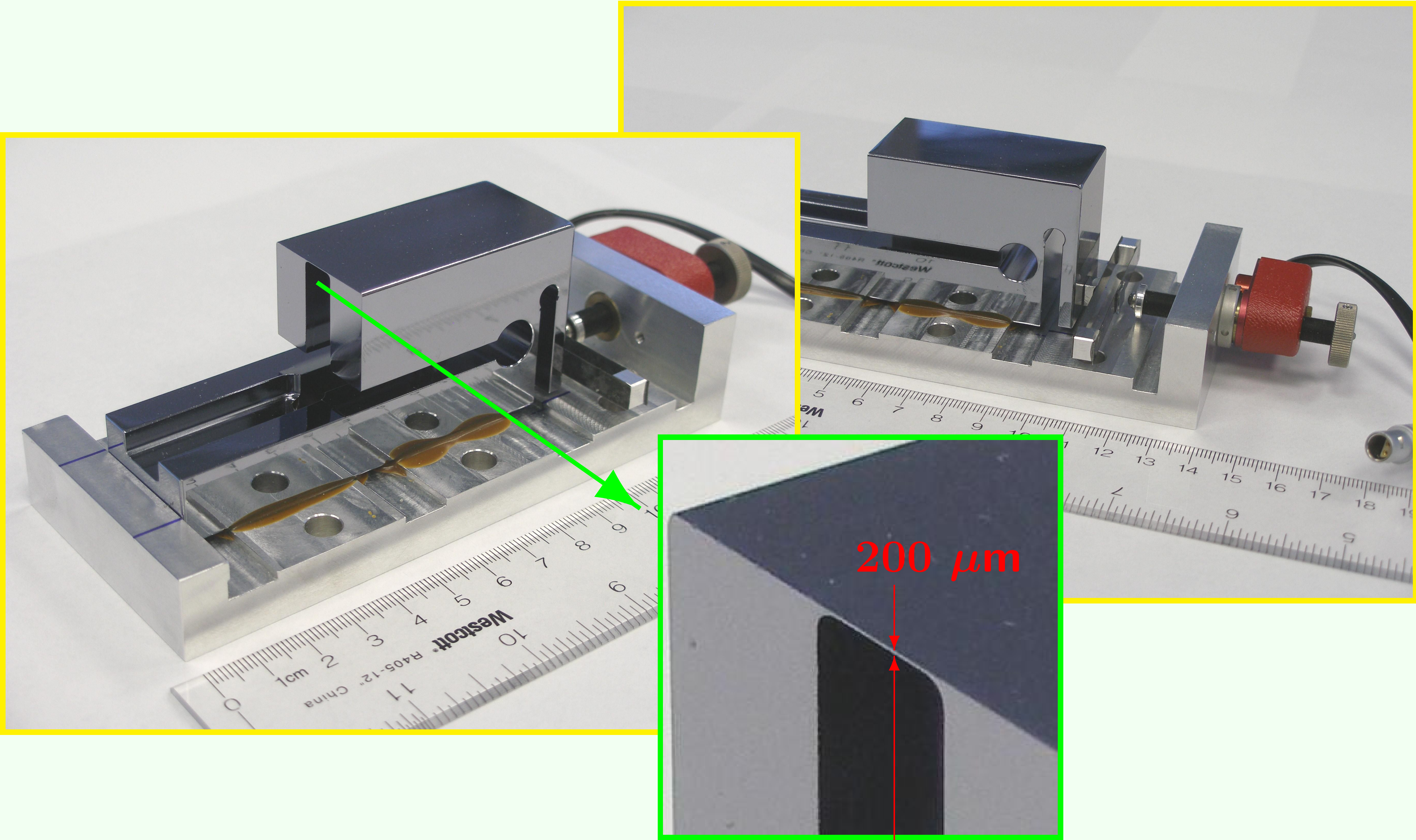

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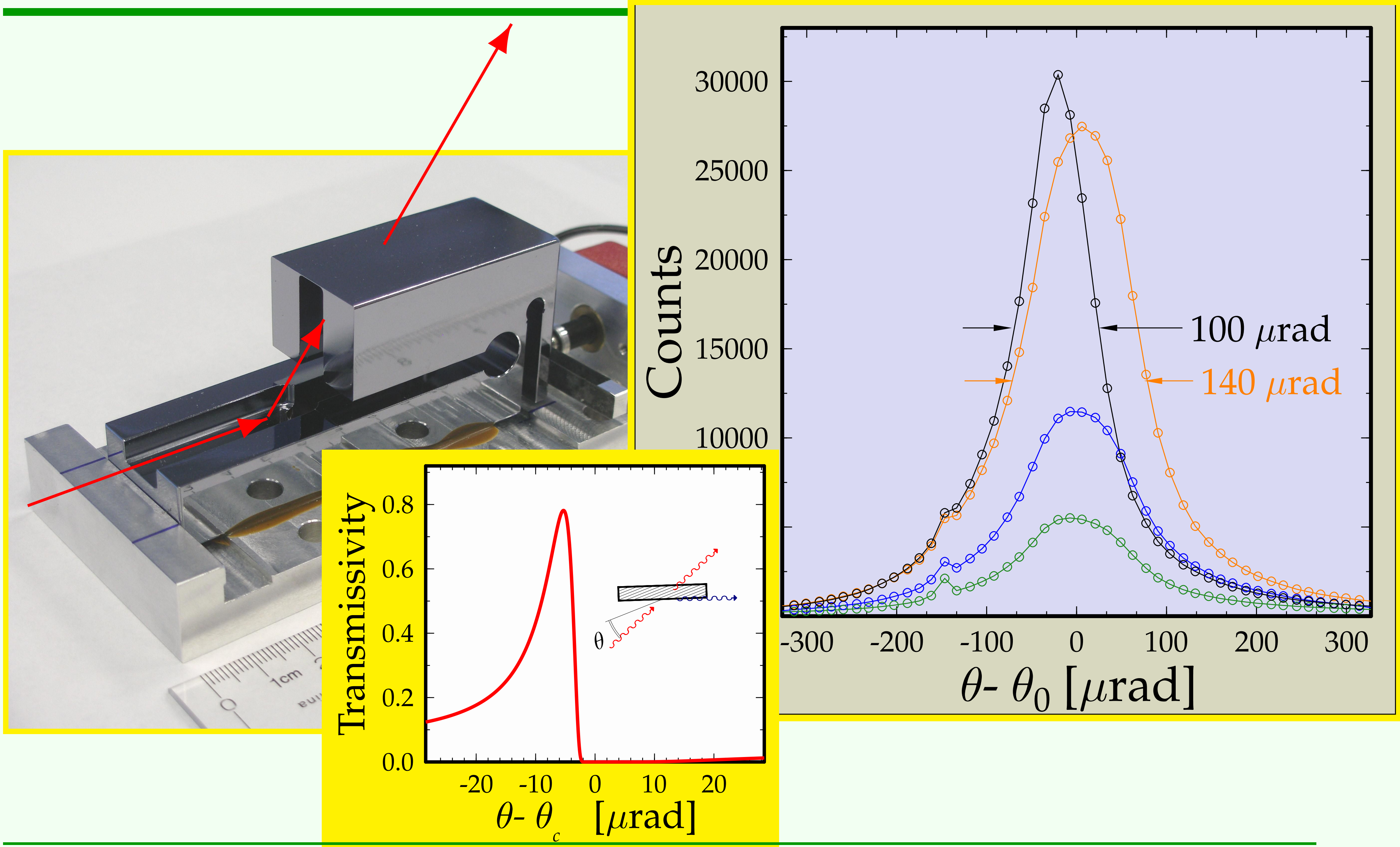
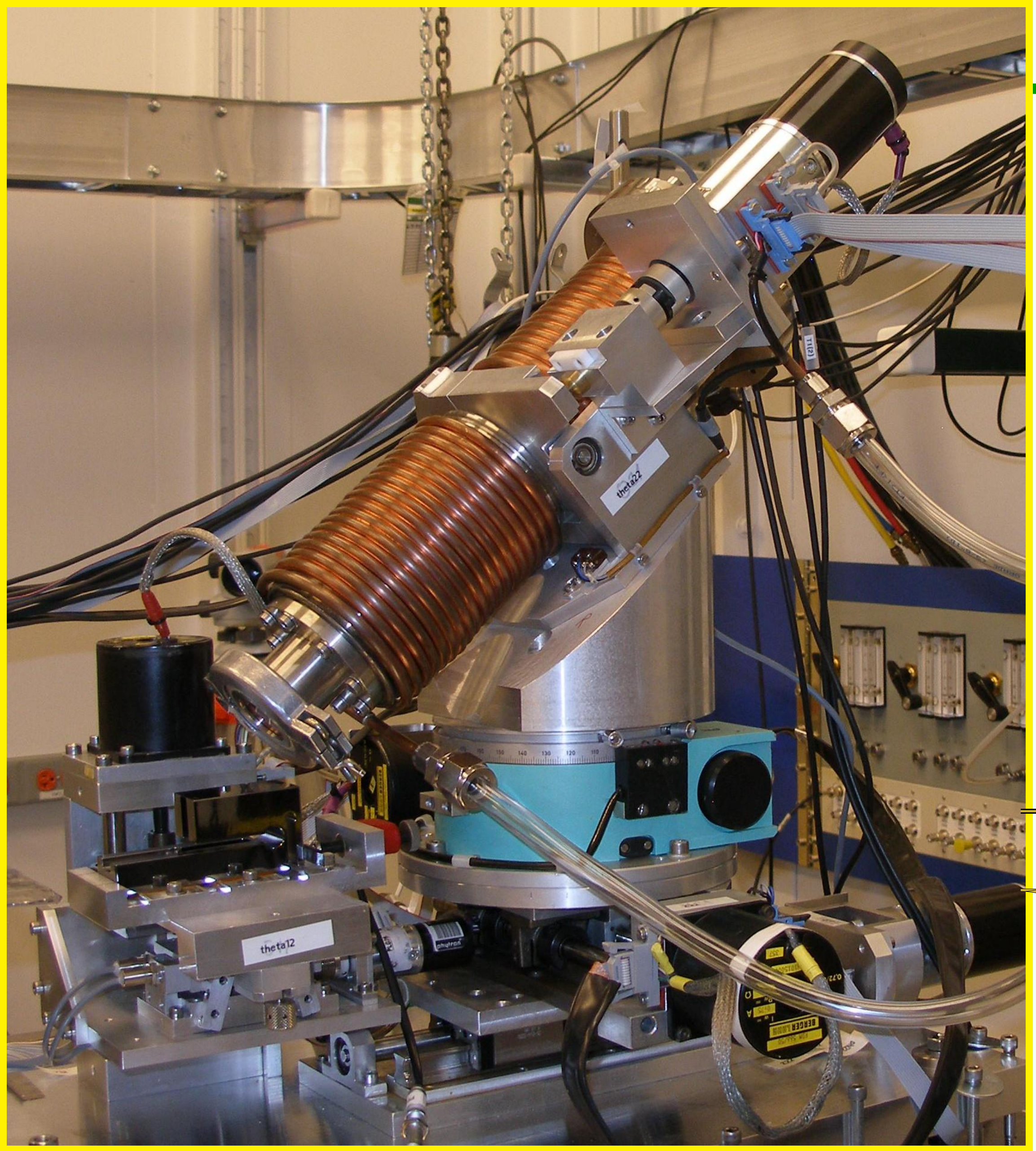
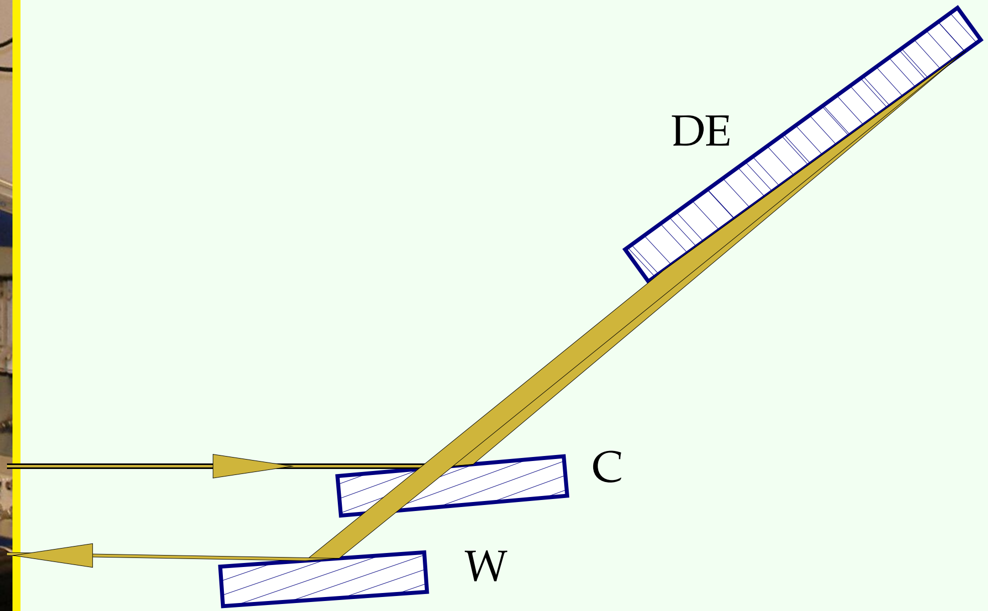
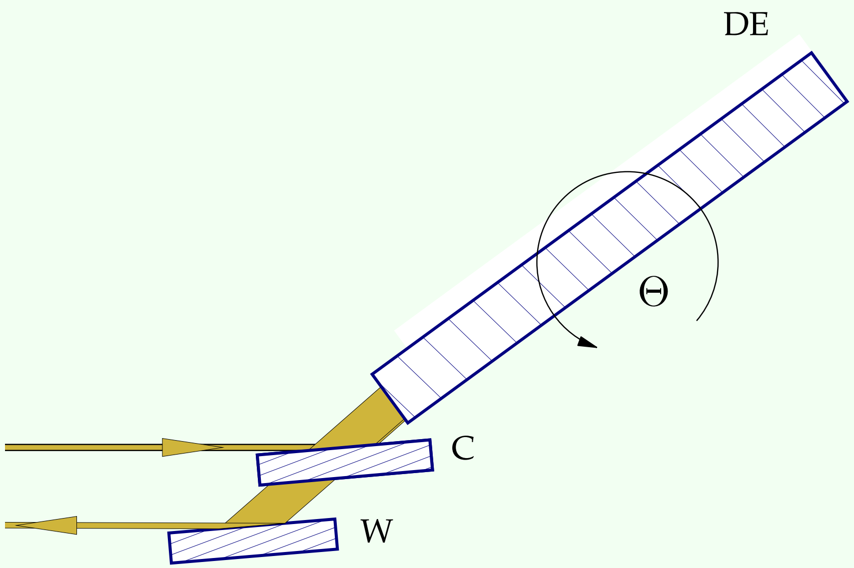


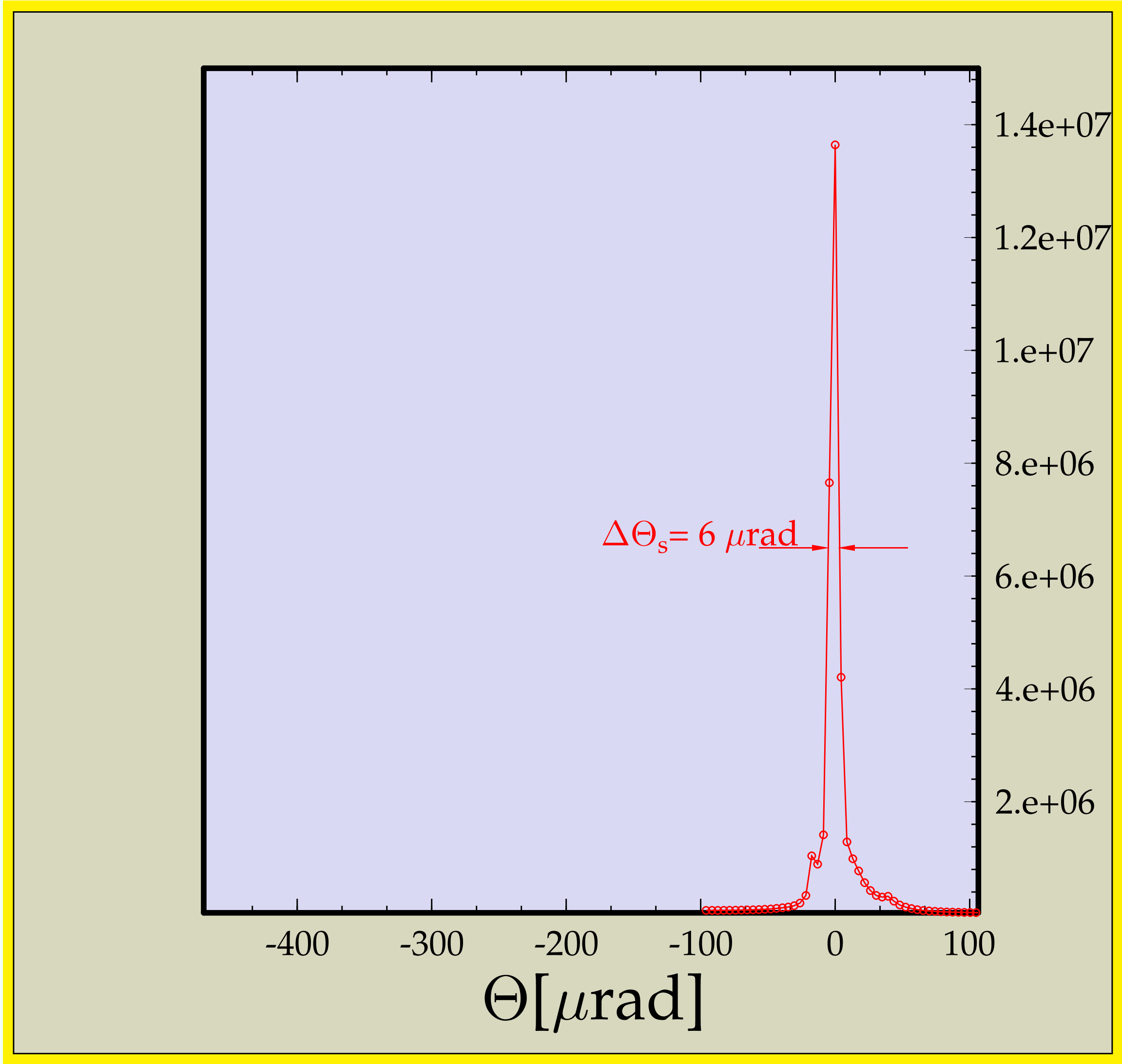
Experimental Set-up @APS.3ID.D



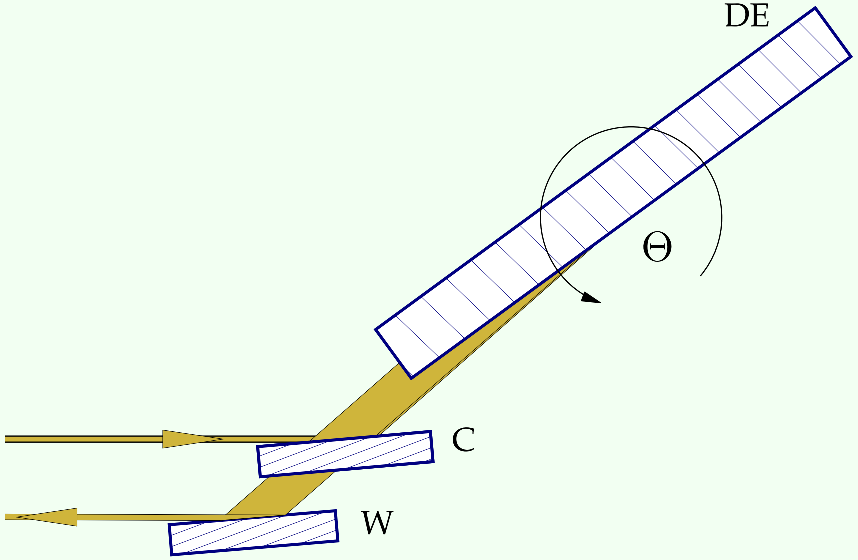


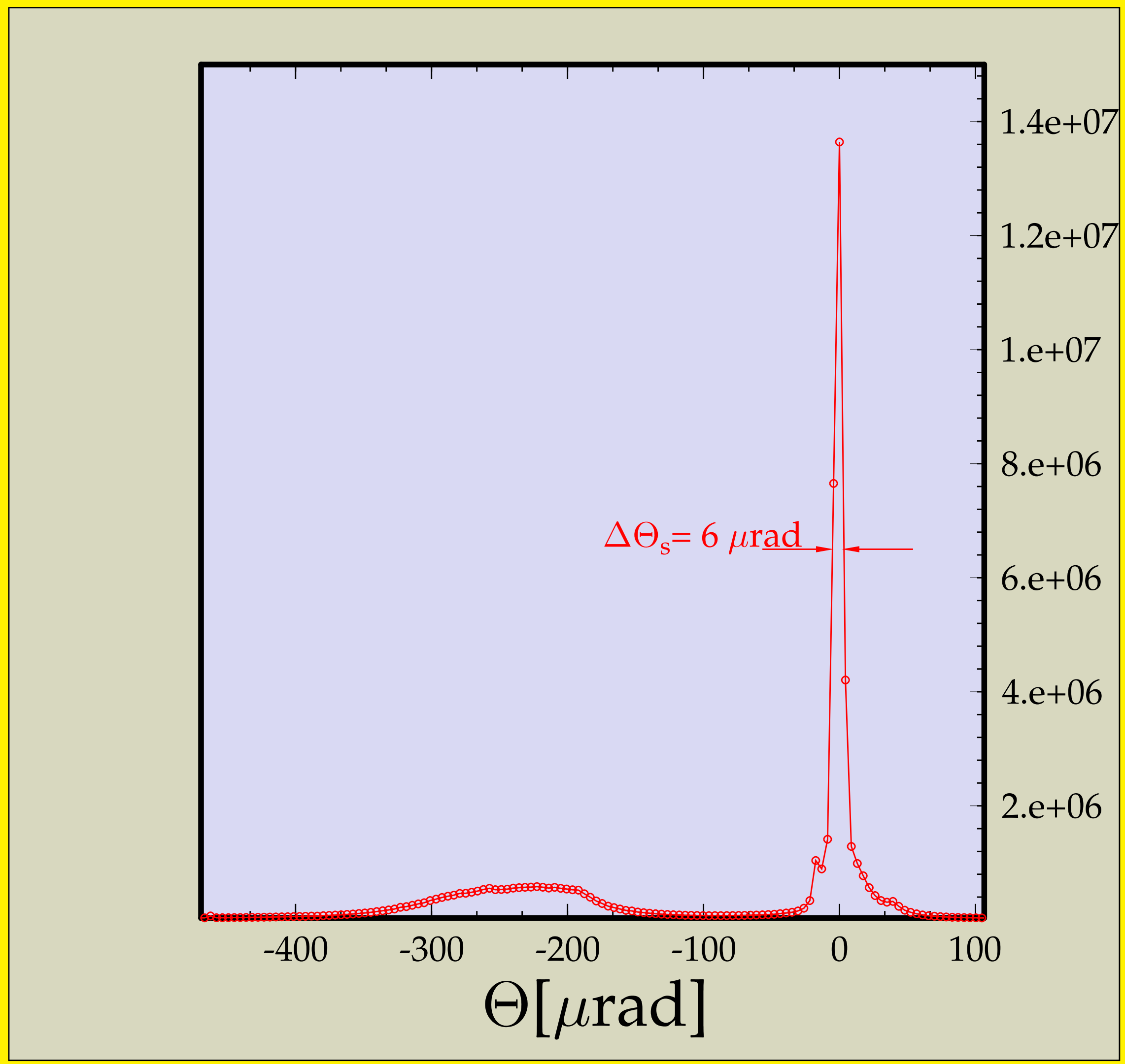
Bragg Backscattering from Symmetrically Cut Crystal





Bragg Backscattering from Asymmetrically Cut Crystal



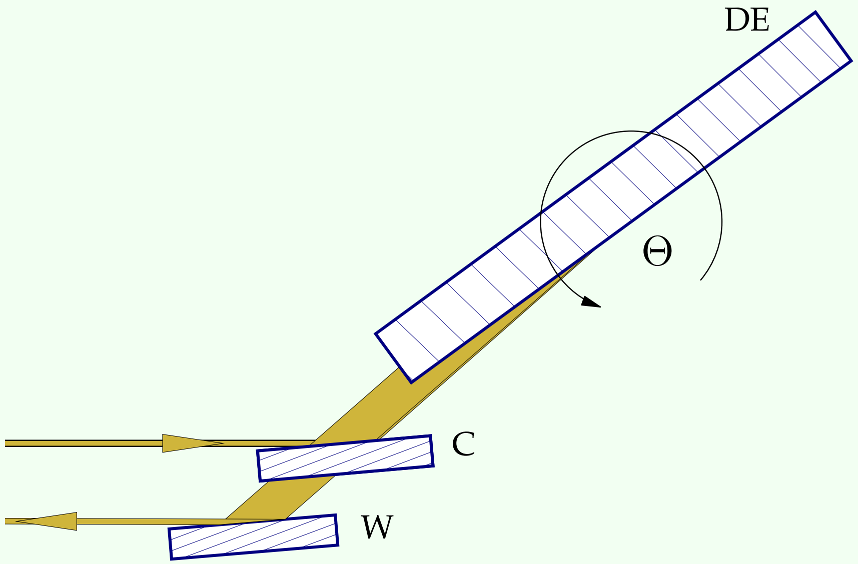


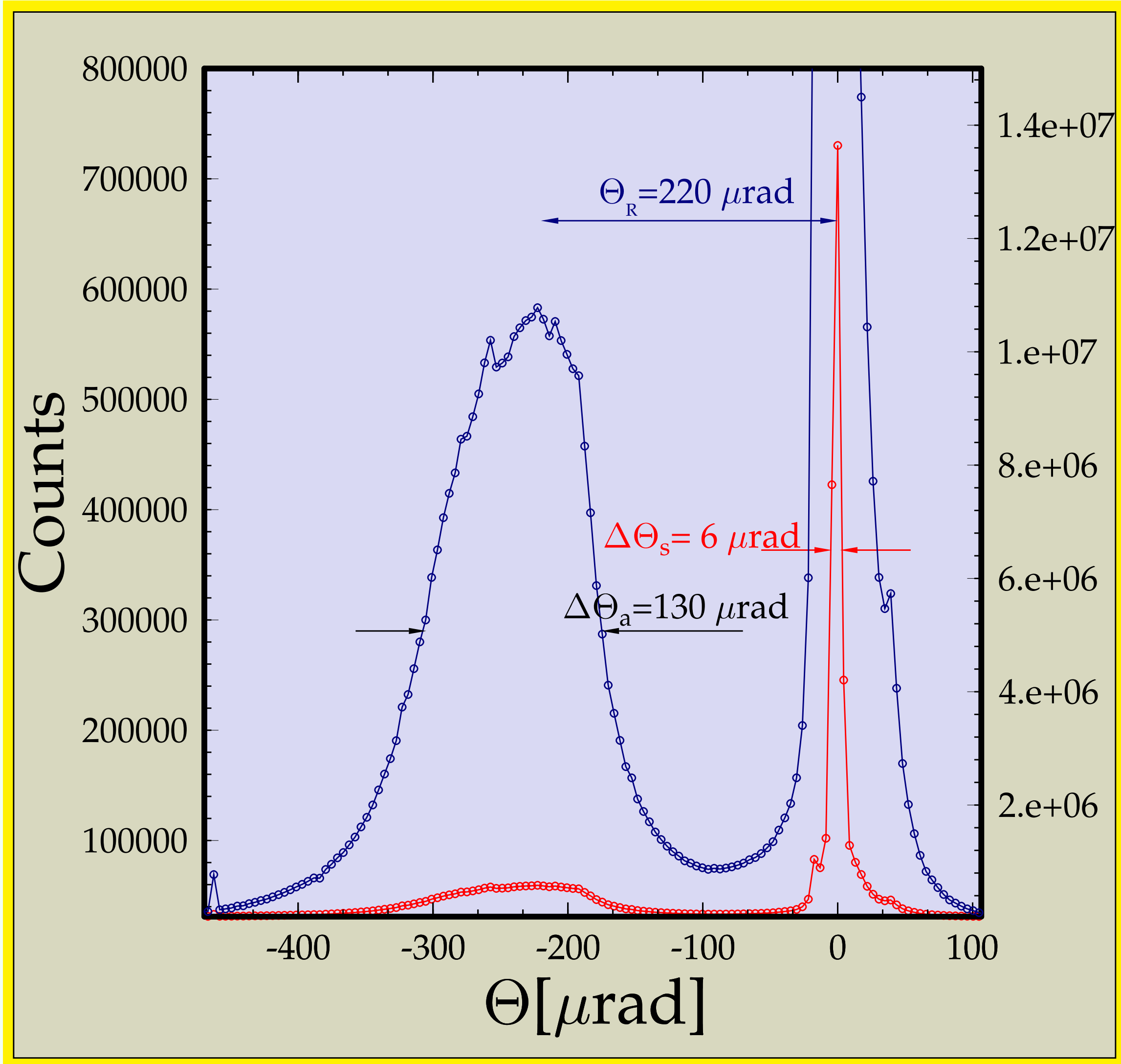
Bragg Backscattering from Asymmetrically Cut Crystal

What do we observe?

1. **Exact Backscattering takes place NOT at normal incidence to atomic planes: angular shift $\Delta\Theta_R = 220\ \mu$rad**

2. **Angular dispersion: $2 \times \Delta\Theta_a = 260\ \mu$rad**



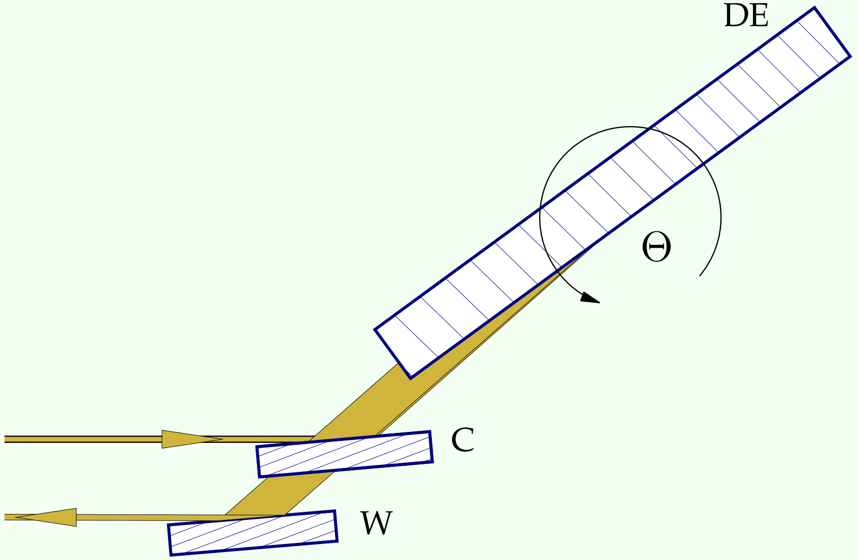


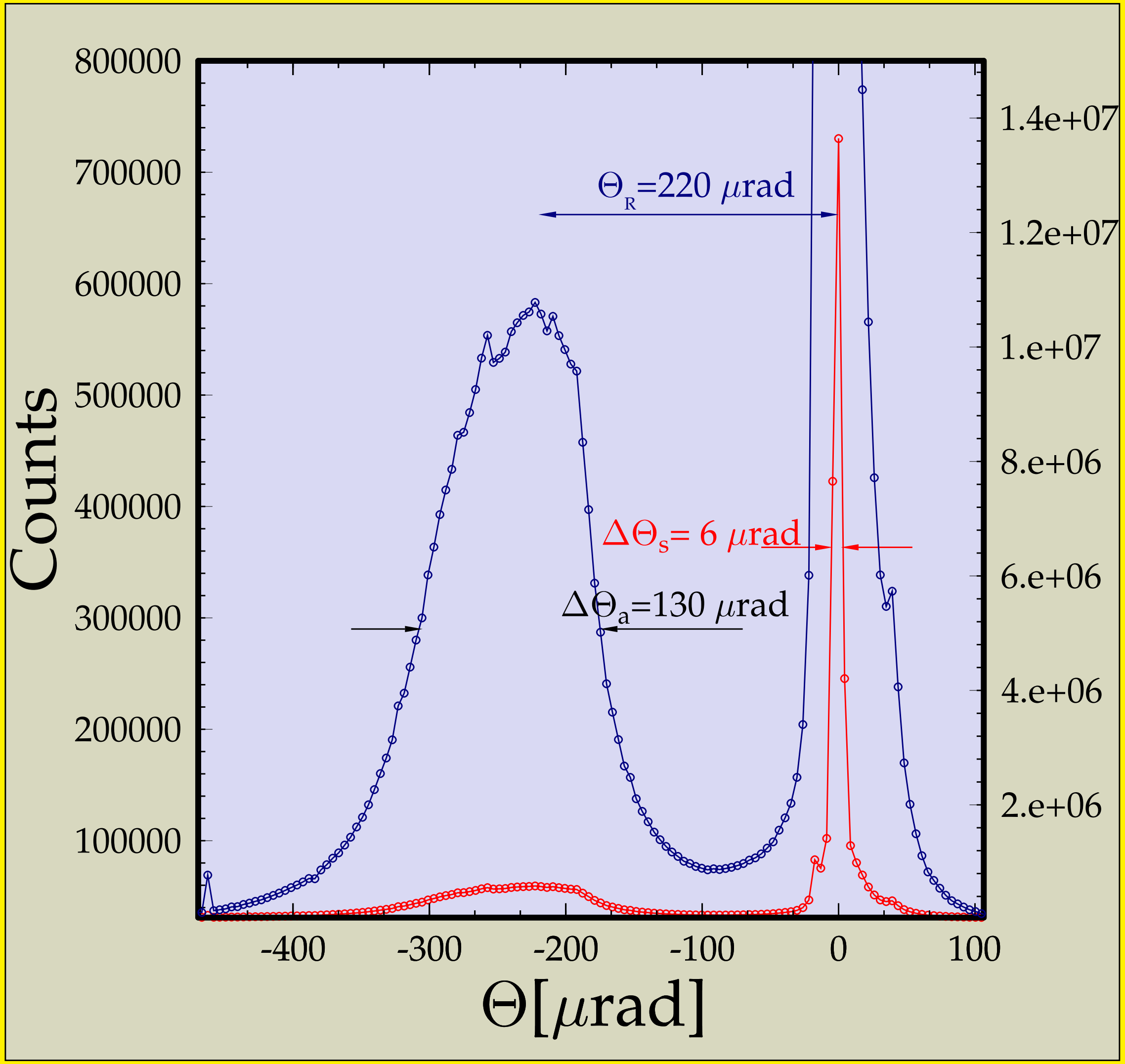
Bragg Backscattering from Asymmetrically Cut Crystal

THEORY:

1. **Angular shift:**
   $\Theta_R = w_H \tan\eta = 224\ \mu\text{rad}$
   $w_H = 2.65 \times 10^{-6}; \qquad \eta = 88.5^\circ$

2. **Angular dispersion:**
   $\Delta\theta = 2\epsilon_H \tan\eta = 225\ \mu\text{rad}$
   $\epsilon_H = 2.95 \times 10^{-6}$





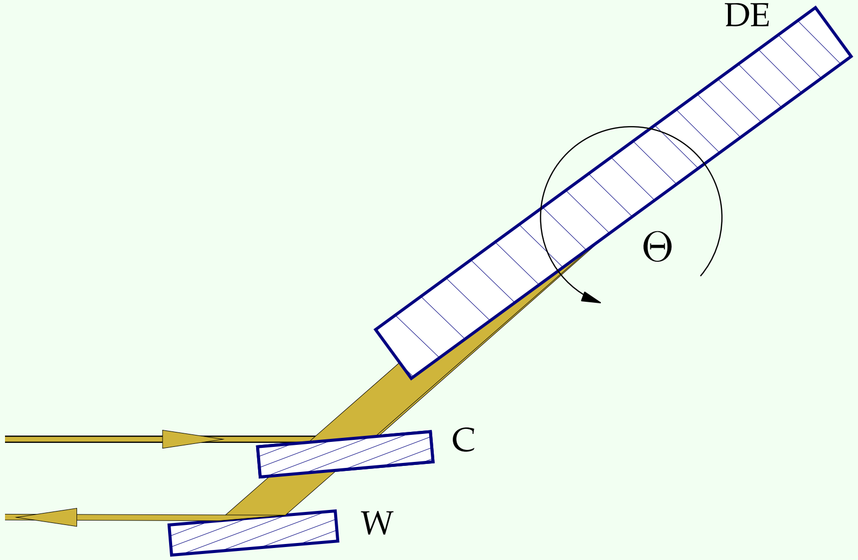
Estimation of the Energy Resolution

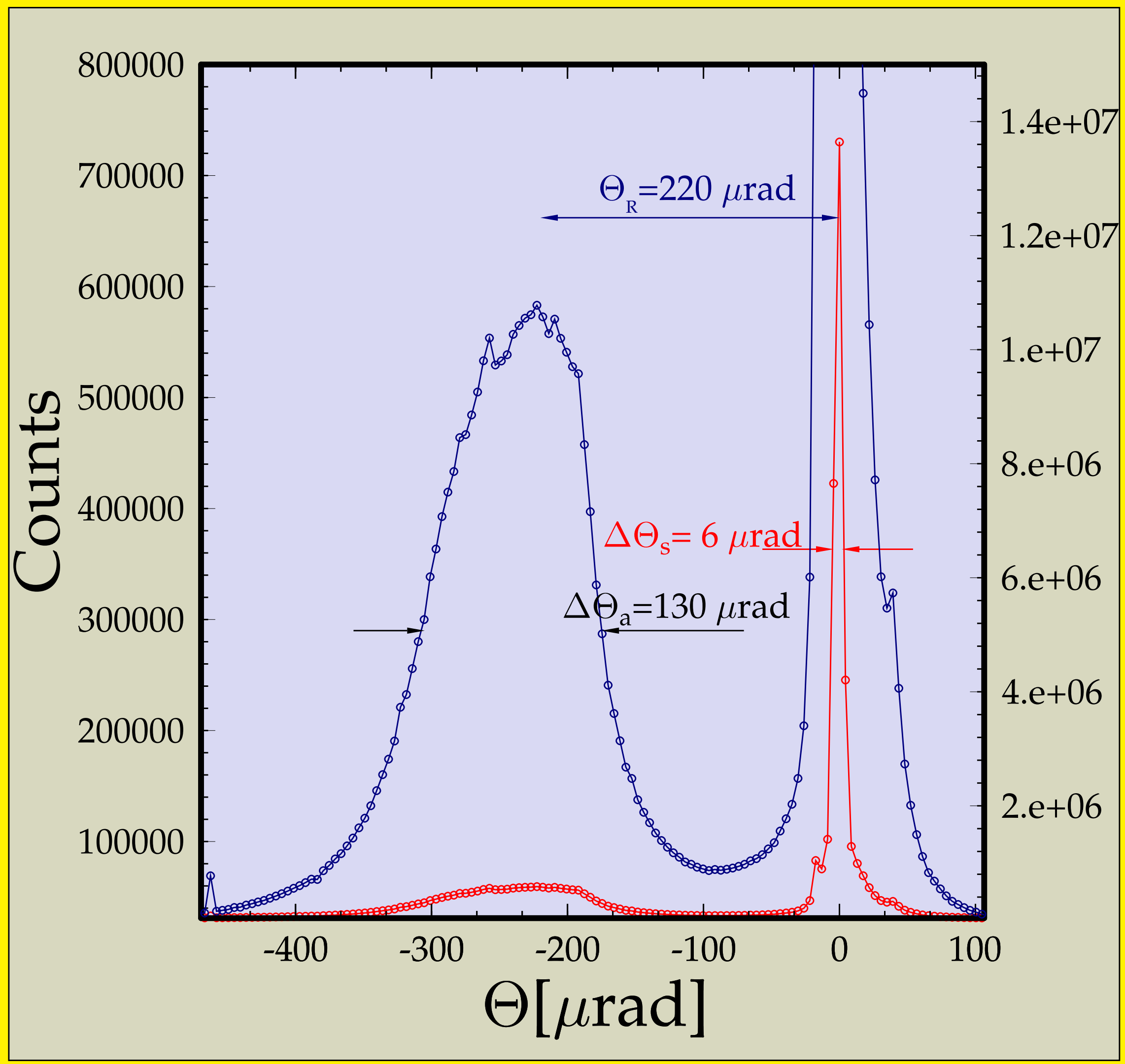
Energy width of Back Bragg reflection:

$\Delta E = 27$ **meV**

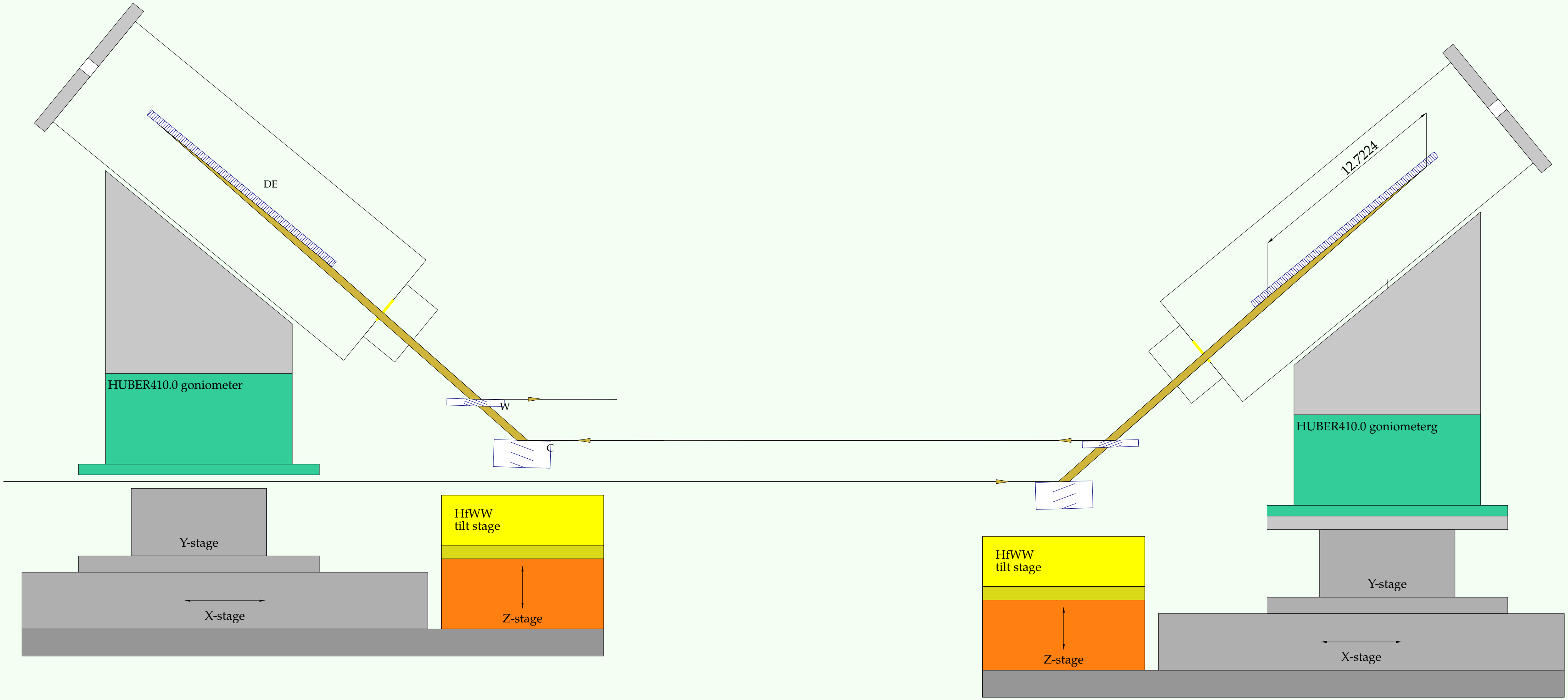
Wavelength selector selects:

$\simeq \Delta E\, \delta\Theta_s/(2\Theta_s) =$ **0.6 meV**

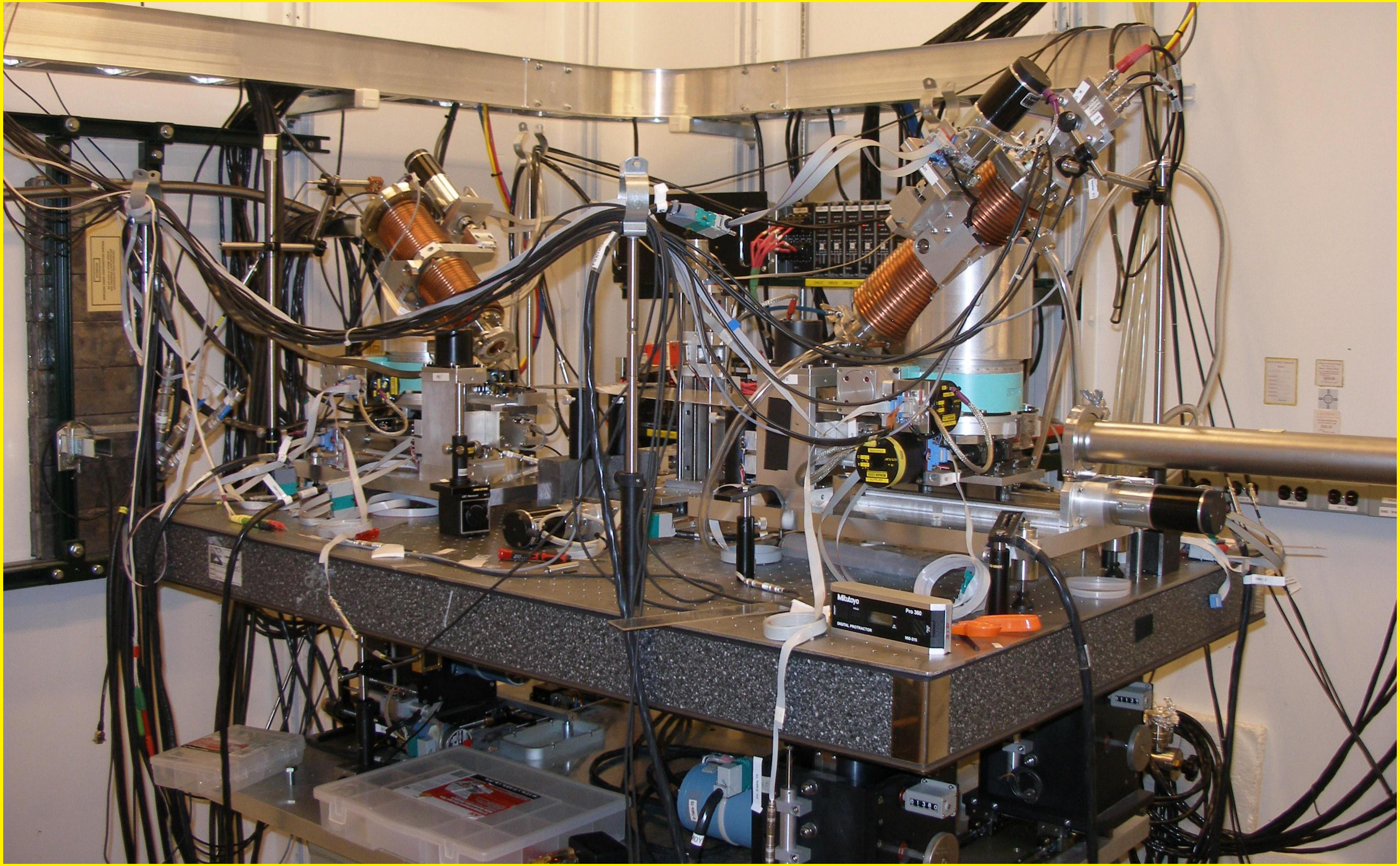




Energy Resolution - Direct Measurements



Experimental Set-up: @APS, 3ID-D



Summary of properties

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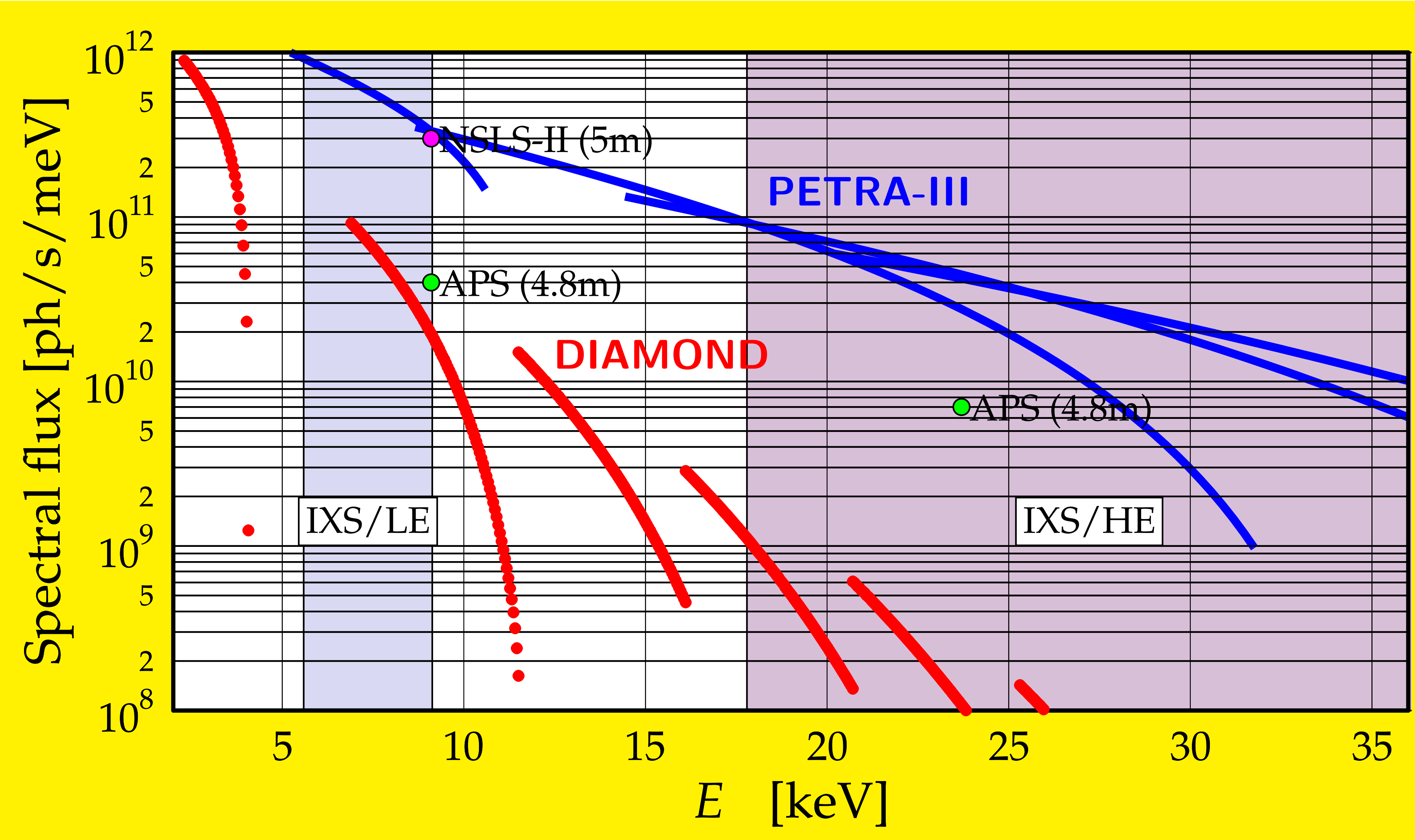
Undulator spectrum



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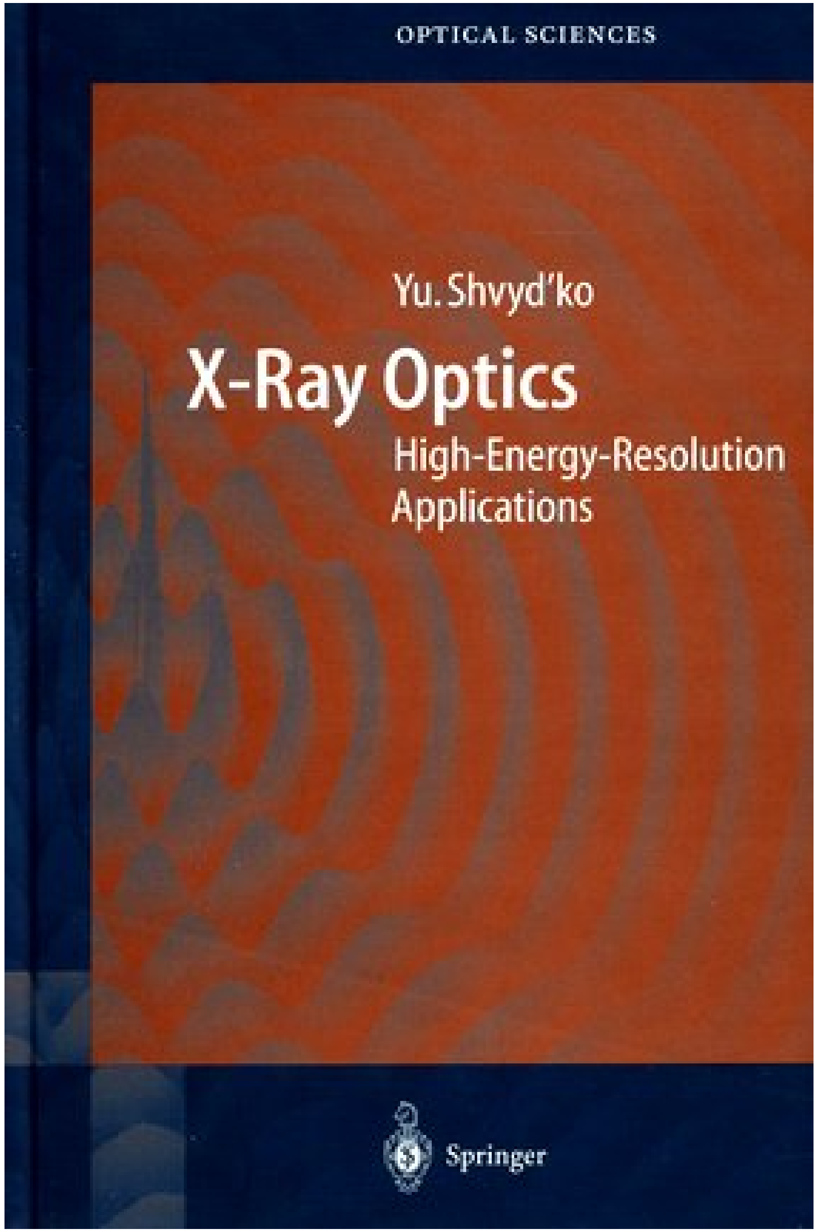
- Steep wings in the spectral function.
- Proximity to K-absorption edges of the important transition metals.

More details in:



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