Development of Millivolt Resolution X-ray Optics for Inelastic X-ray Scattering at NSLS-II

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  - → Xianrong Huang
  - → Marcelo Honnicke
  - → Jeff Keister
  - → Zhong Zhong *(MOU)*
  - → Scott Coburn, *Engineer*
  - → Leo Reffi, *Designer*
  - → Bill Struble, *Technician*

- **Collaborators** (Beamline Advisory Team)
  - → Clement Burn (Spokesperson), *WMU*
  - → Sow-Hsin Chen, *MIT*
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  - → John Hill, *BNL*
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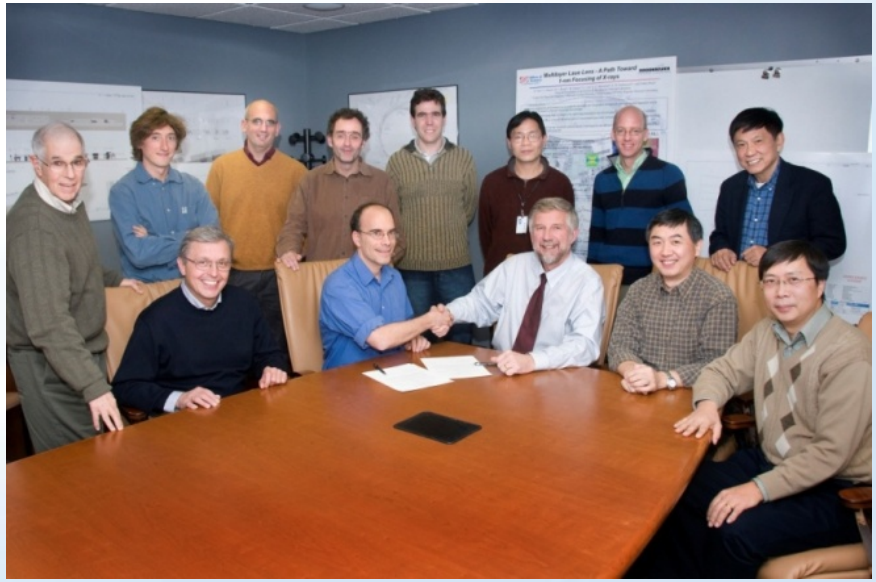
  - → Stephen Shapiro, *BNL*
  - → Yuri Shvyd'ko, *APS*



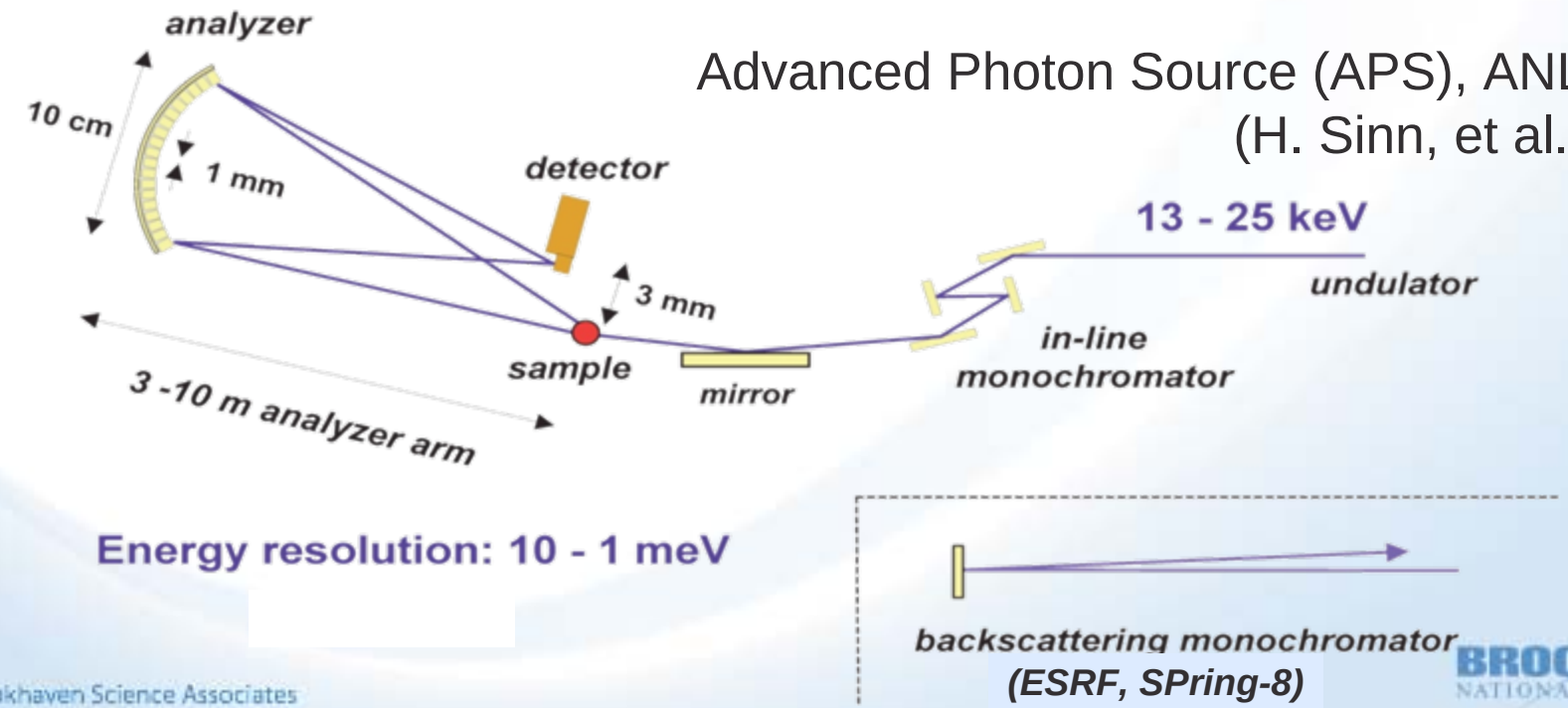
Outline

- Current Status of meV Inelastic X-ray Scattering
  - → Need for ultrahigh resolution in (E, Q)
- Optical Scheme for 0.1 meV and 0.01 nm^{-1}
- Spectrometer Design and Performance Issues
- Key Scientific Applications

meV IXS: Current Status

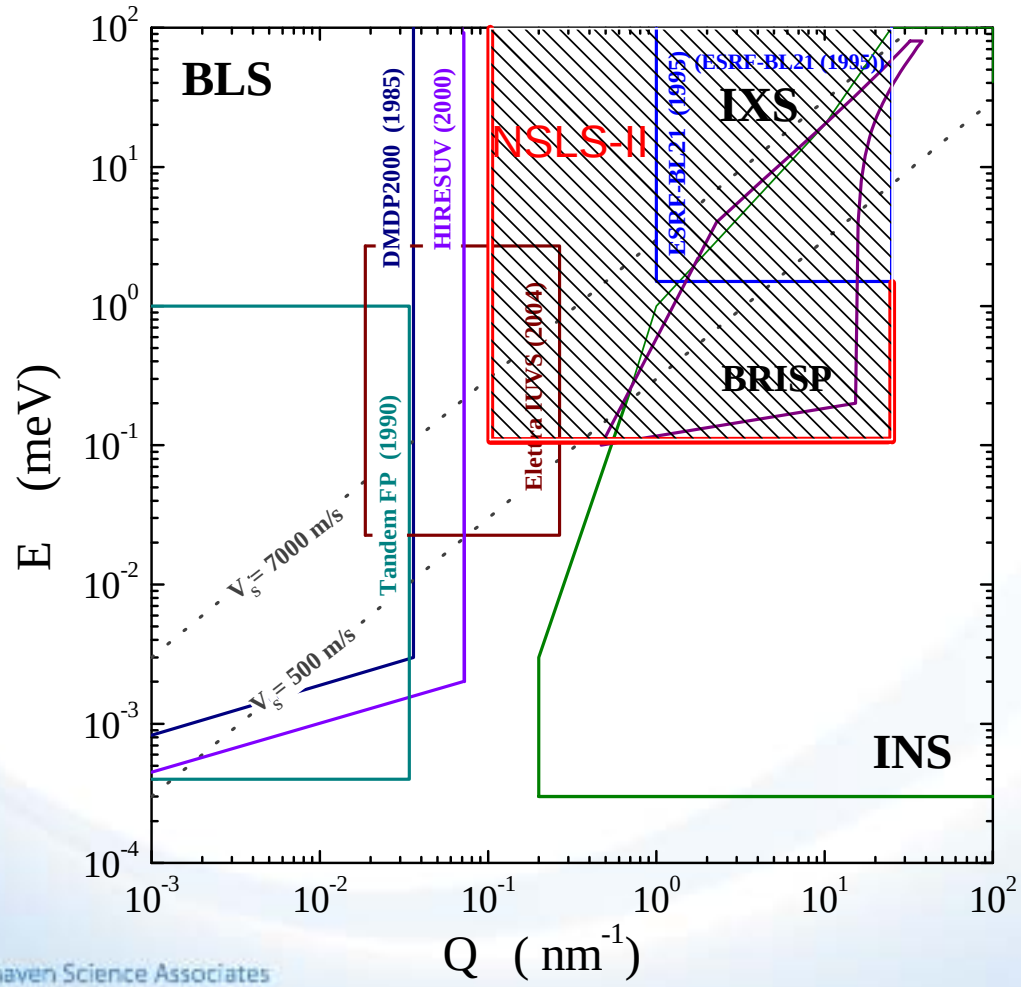
- Spectrometer optics (mono and/or analyzer) design based on high-order symmetric Bragg back reflections of Si(nnn) (n = 7 - 13)
- Successful implementation at all high energy synchrotrons (APS, ESRF, and SPring-8), routinely operated at < 2 meV and ~ 0.5 nm^{-1} with ~10^9 photons/sec above 20 keV
- Powerful tool to study phonons in solids, vibration modes and relaxation processes in liquids and disordered systems

Advanced Photon Source (APS), ANL
(H. Sinn, et al.)



NSLS-II: Filling in the Gap

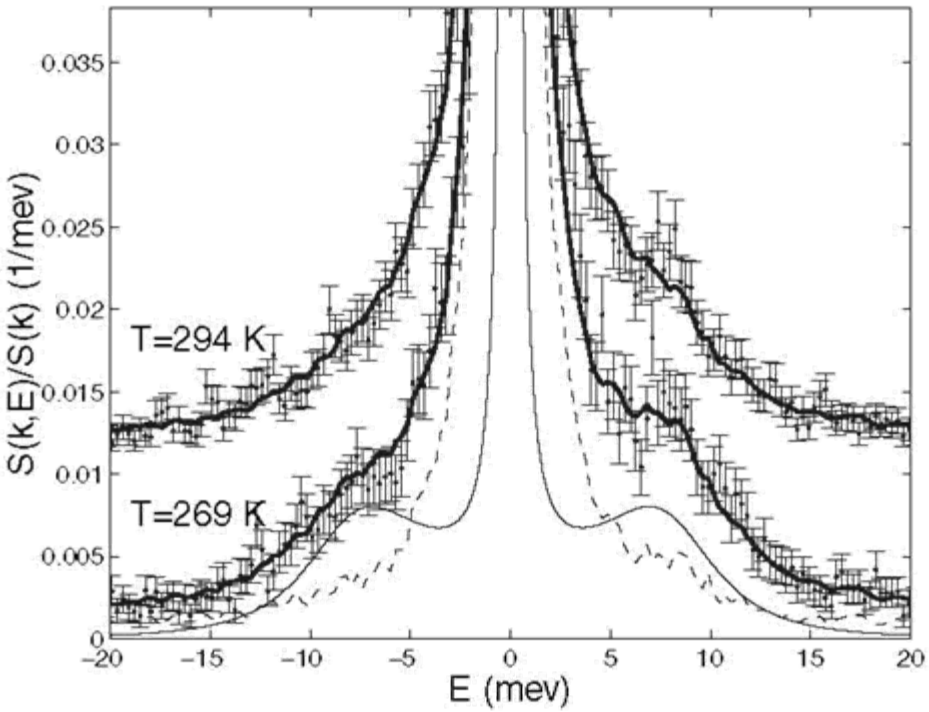
- New NSLS-II IXS spectrometer aims to achieve an ultimate resolution of 0.1 meV and 0.01 nm^{-1}, partially bridging the dynamic gap between the existing high and low frequency probes

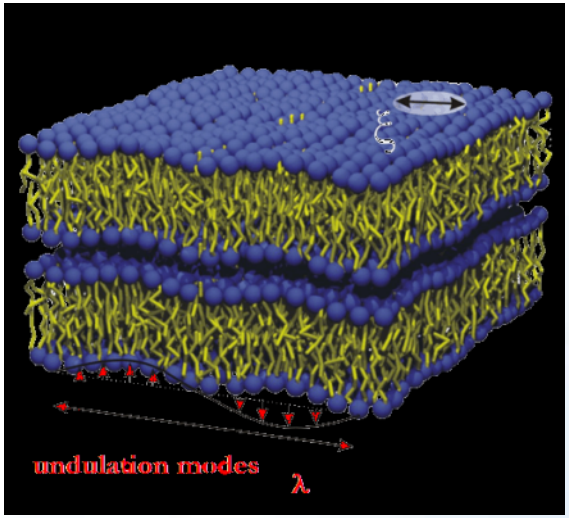


- Immediately useful for a few classes of 0.1 meV experiments:
  - → Visco-elastic crossover behavior of disordered systems and fluids
  - → Collective modes in soft matter and biomolecular systems (e.g., lipid membrane)

Collective motions in lipid membranes

ΔE = 1.5 meV, 21.75 keV



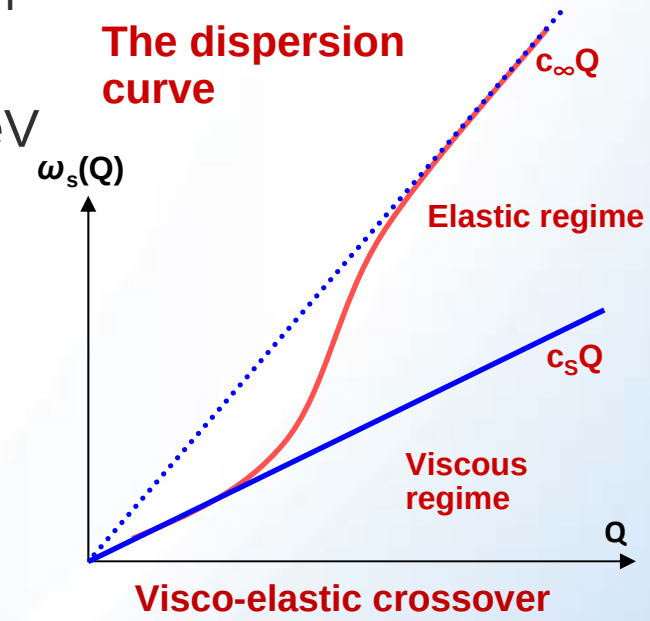


Chen et al, Phys. Rev. Lett. (2001)

- Better resolution is needed
- Advantages of X-rays: small sample (0.2mm), no deuteration, scattering dominated by heavy atoms as compared to inelastic neutron scattering

NSLS-II IXS Beamline Overview

- Two spectrometers based on new optics scheme
  - → E ~ 9 keV well matched to undulators
  - → Ultrahigh (0.1meV) and high (0.5-1meV) resolution
  - → Improved resolution tails
- Incident flux at sample > 10^9 photons/sec/0.1meV
- Q range / resolution:
  - → Liquids, glassy systems: 0.02 ~ 10nm^{-1} / 0.01nm^{-1}
  - → Soft matter: 0.1 ~ 40nm^{-1} / 0.1nm^{-1}
  - → Hard matter 0.1 ~ 80nm^{-1} / 0.1nm^{-1} or lower
- Focus:
  - → Vertical focus < 5µm
  - → 1µm (V) x 3µm (H) desirable for high pressure
- Sample environments
  - → Compatible with high-P, low-T (4K), high-T (1000K), and single crystals



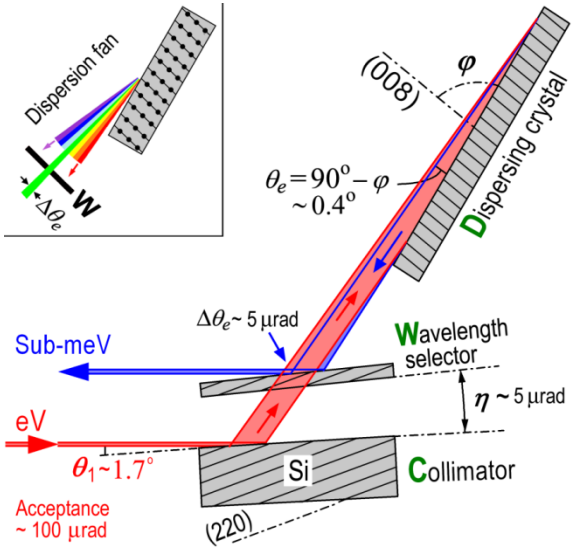
→ **New optics scheme requires considerable R&D effort !**

0.1meV Crystal Optics

- CDW and CDDW optics schemes at 9.1 keV proposed by Yu. Shvyd'ko, verified by our dynamical theory calculations (Xianrong Huang)

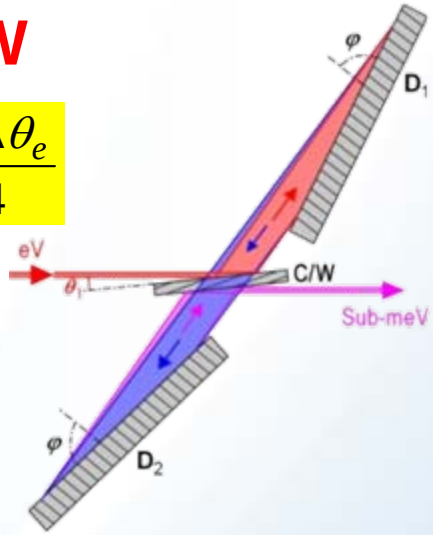
CDW

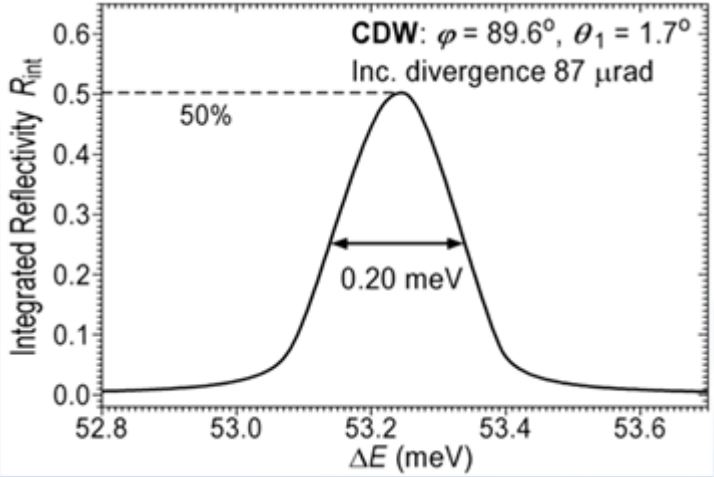
$$\frac{\Delta E}{E} \approx \frac{\Delta\theta_e}{2\tan\varphi} \approx \frac{\theta_e \Delta\theta_e}{2}$$

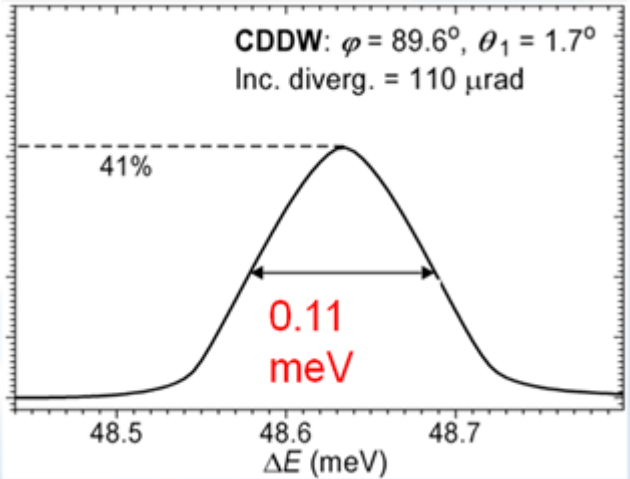


CDDW

$$\frac{\Delta E}{E} \approx \frac{\theta_e \Delta\theta_e}{4}$$







→ Large angular acceptance (>100 µrad)

→ High efficiency (~50%)

→ Sharp tails (Borrmann effect), more important than resolution!

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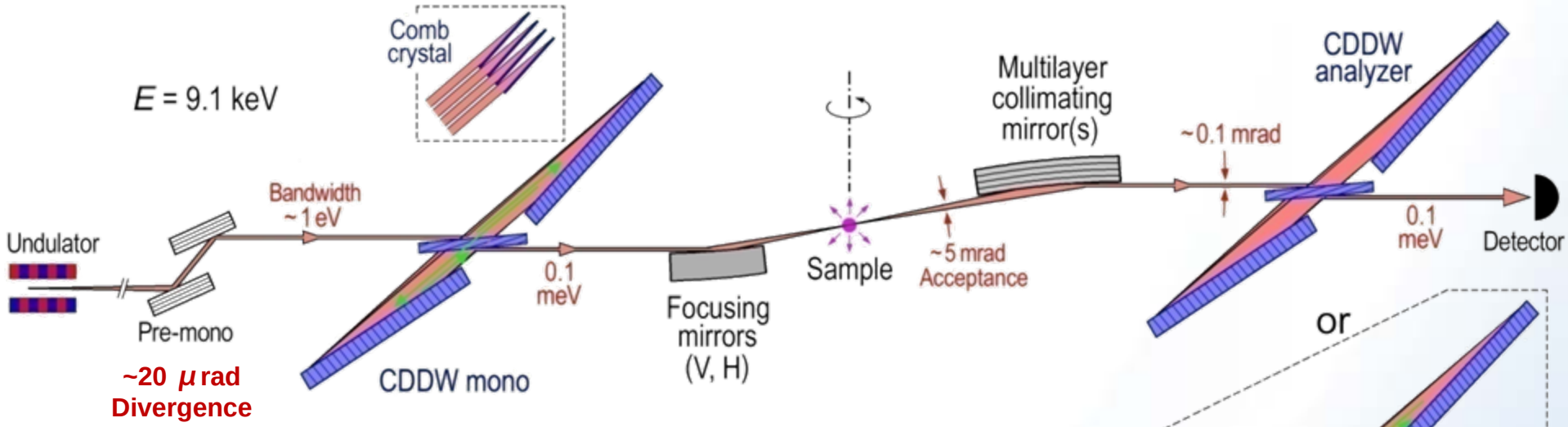
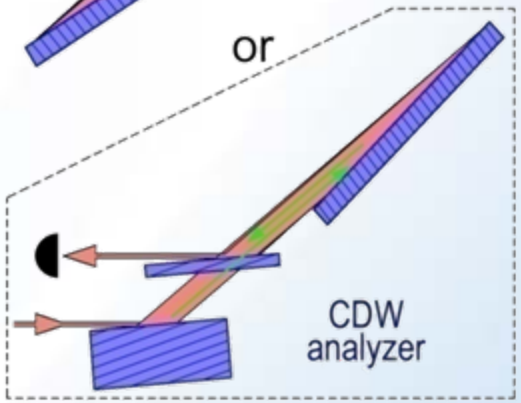
Beamline and Spectrometer Optics Layout





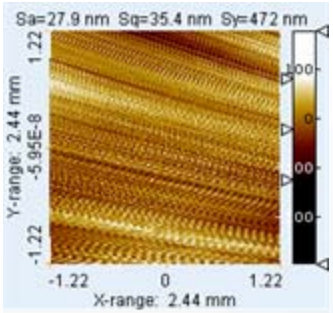
- Major parameters determined and verified for E = 9.13 keV, $\Delta\theta_e$ = 5 μrad, and h = 0.5 mm

ΔE of CDW	ΔE of CDDW	Θ_e (= 90° – φ)	Length of D
2 meV	1 meV	4.5°	120 mm
0.7 meV	0.3 meV	1.5°	380 mm
0.2 meV	**0.1 meV**	**0.4°**	**1400 mm!**

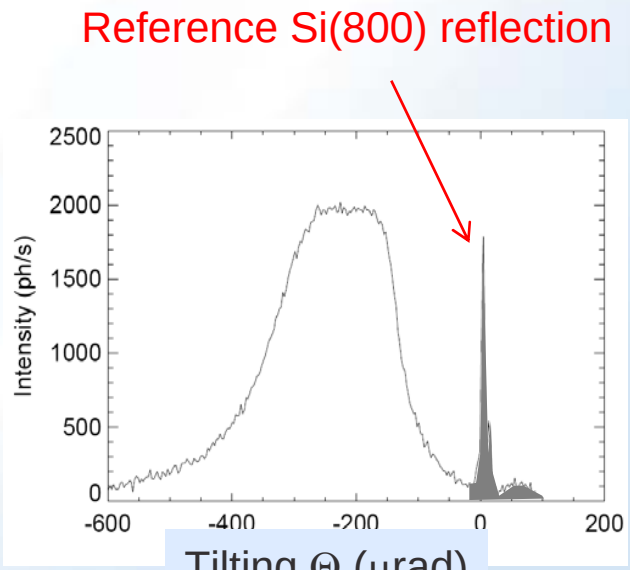
- Comb crystal proposed by Yu. Shvyd'ko: a possible solution to the long D crystal length, but a major challenge to fabricate (in progress)

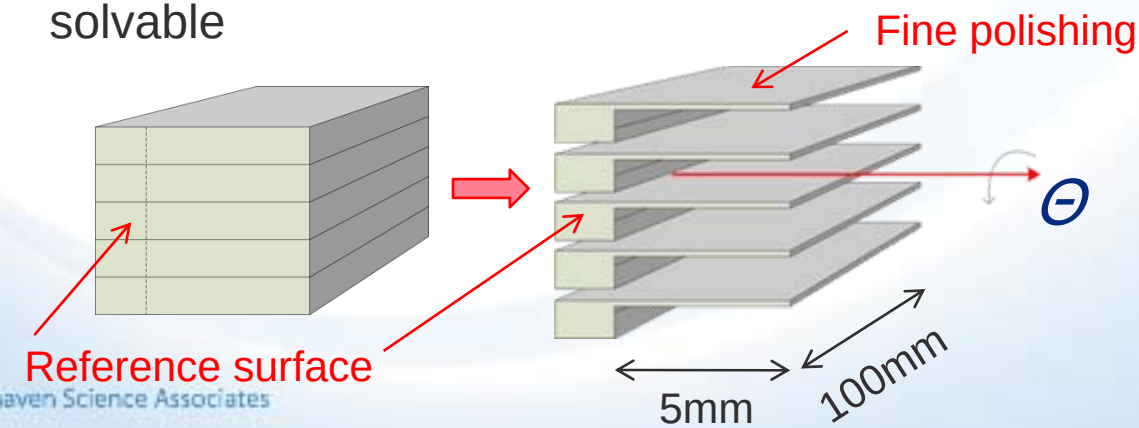
Comb Crystal Fabrication

- Current approach employed by APS:
  - → Cut directly from a monolithic crystal.
  - → Polishing the diffraction surface a major challenge.
- Alternative approach to be explored:
  - → Prepare reference Si(100) surface from a monolithic block
  - → Cut individual fins allowing fine polishing of diffraction surface
  - → Align individual blocks using reference surface on symmetric backscattering Si(800) reflection
  - → Mechanical positioning a challenge, but more solvable

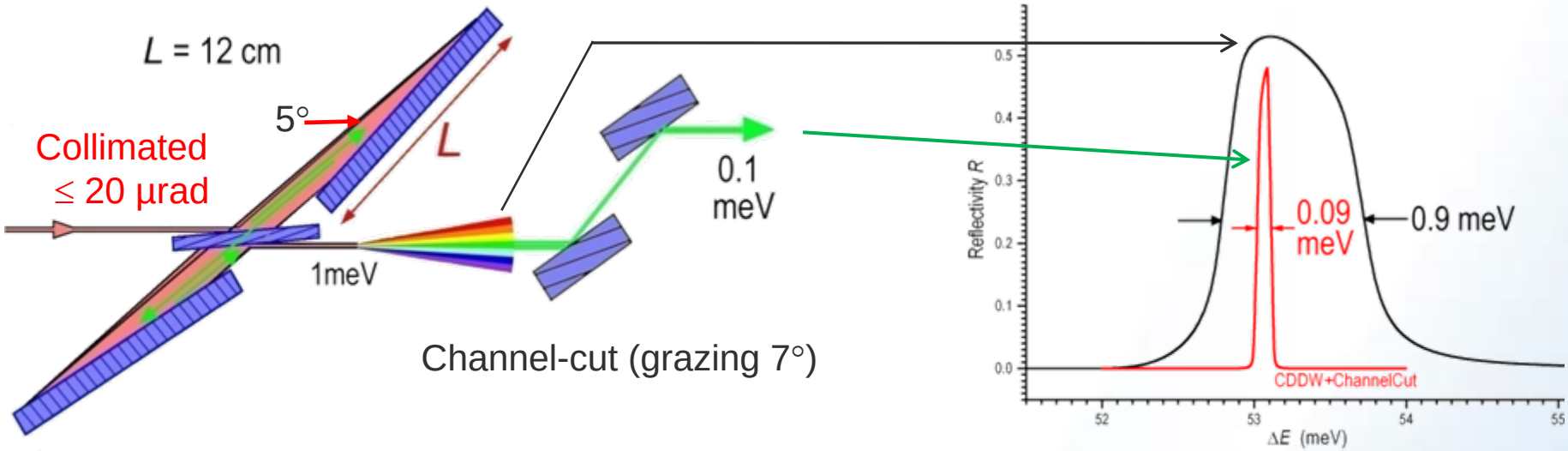


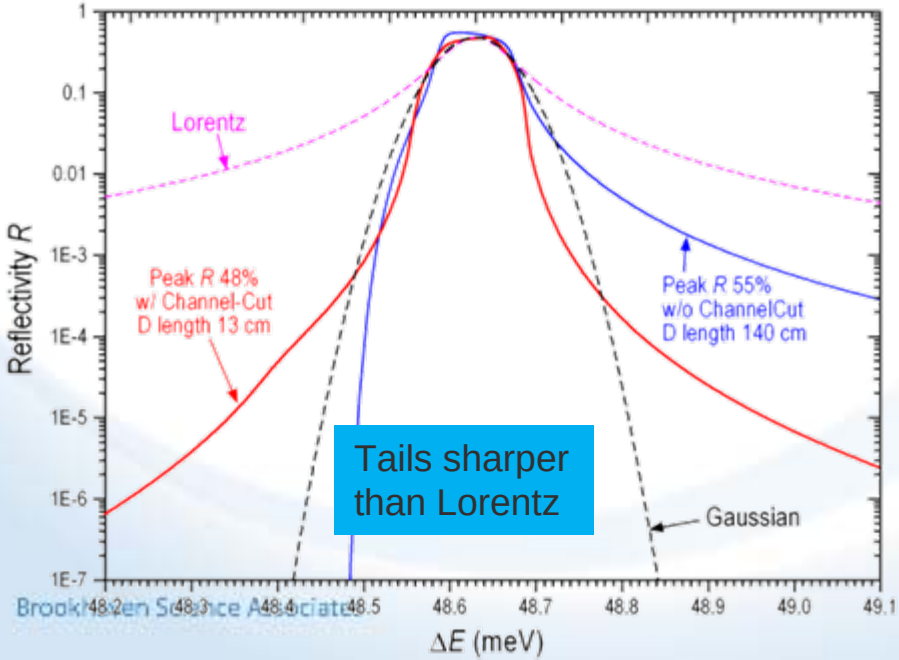
As-cut surface roughness **35.4 nm** by AFM





CDDW for 1 meV + Channel Cut

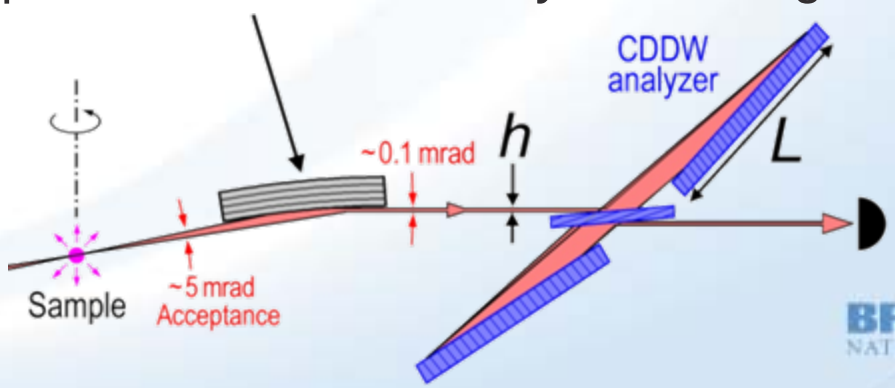




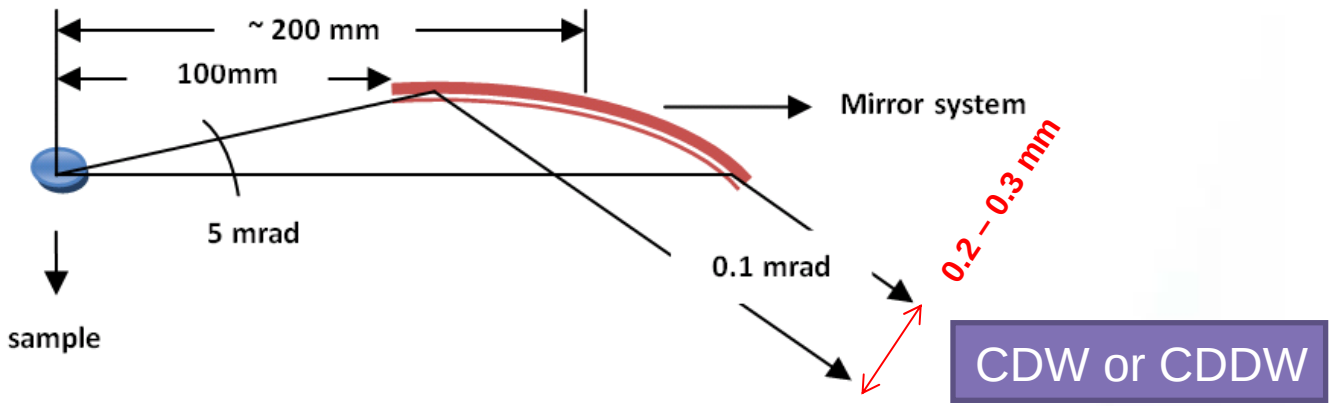
- → No need for long D crystal
- → Switchable between 0.1 and 1 meV
- → Require incident beam collimated to
  ≤ 20 µrad
  no problem for mono
  analyzer needs high-precision mirror.
- → CC causes ~20% efficiency loss

Other Major Technical Challenges

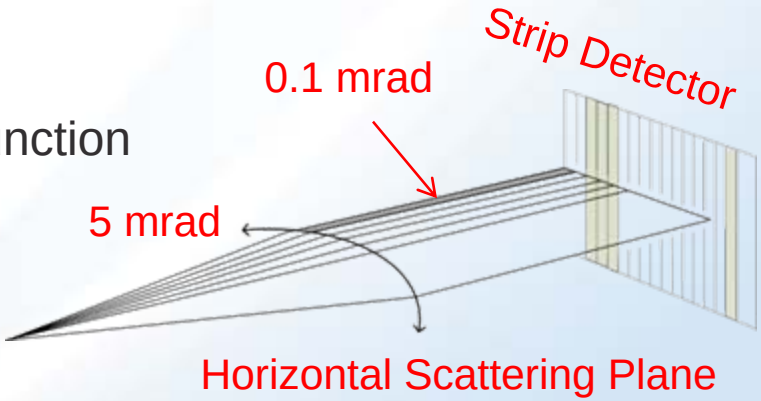
- Lattice homogeneity of the crystal: $\Delta d/d = \Delta E/E \sim 10^{-8}$ (0.1 meV/10 keV)
- Temperature uniformity and stability: $\Delta T \leq 4$ mK
  ($\Delta d/d = \alpha \Delta T$ and thermal coefficient of Si: $\alpha = 2.56 \times 10^{-6}$ K^{-1})
- Surface quality
  slope errors (straightness of surface): < 10 μrad,
  crystal bending (due to mounting or gravity sag): < 0.2 μrad,
  surface roughness (causes diffused scattering): < 2 nm
- Crystal angular stability: < 0.2 μrad
  as a result of energy tuning by angular rotation, independent of resolution 0.1 – 1 meV,
- CDW/CDDW analyzer requires 50~100:1 multilayer focusing mirrors for 5~10 mrad acceptance



Laterally Graded Multi-layer Mirror



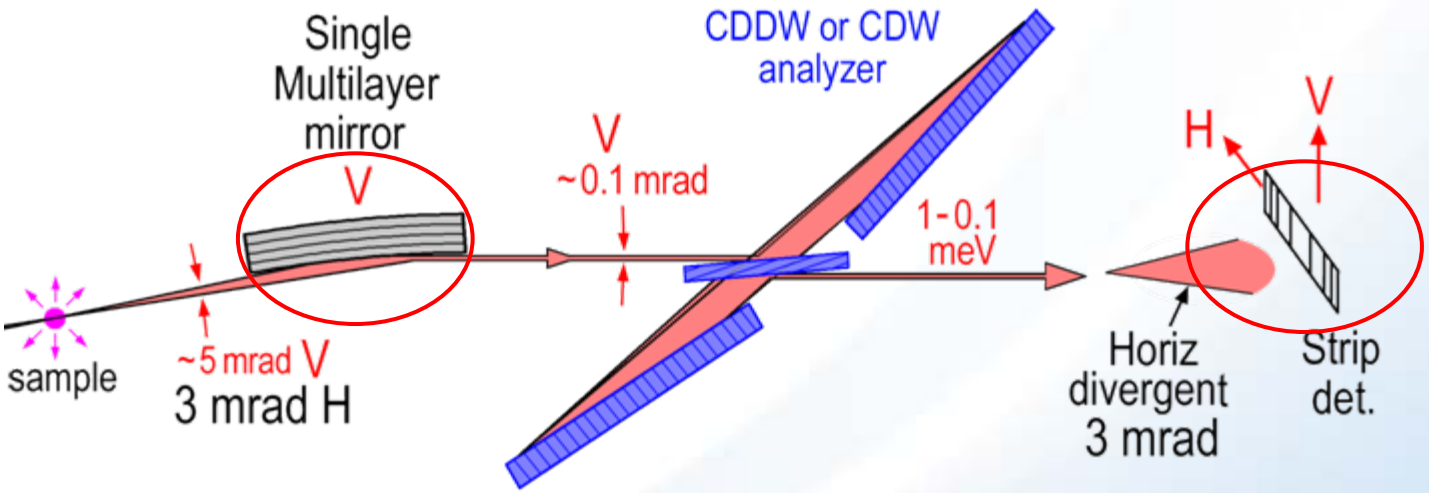
- Design Objectives:
  - → Angular acceptance: 5 mrad x 5 mrad
  - → Best Q resolution: 0.01 nm^{-1}, corresponding to ~0.1 mrad in horizontal fan at 9.1 keV or 20 µm transverse width at 200 mm from source.
    Possible solution: use area detector in conjunction with a horizontally collimating mirror



Extremely high horizontal collimation required!

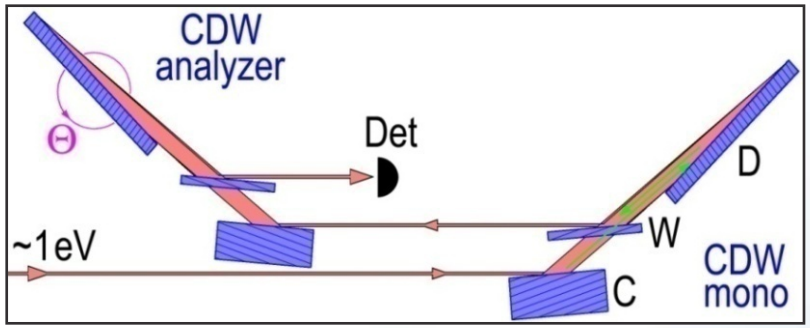
A Simplified Mirror Configuration

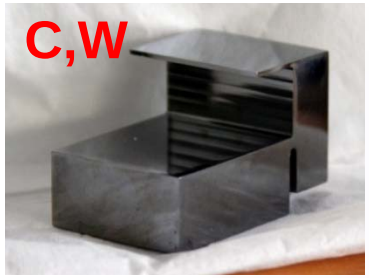
- A single mirror focusing / collimating only for the vertical direction
  - horizontal natural angular acceptance: ~ 3mrad.
  - energy dispersion: $E(\varphi) = E_0(1 - \varphi^2/2)$
- Multiple mirrors + multiple CDW/CDDW analyzers in the horizontal to increase efficiency

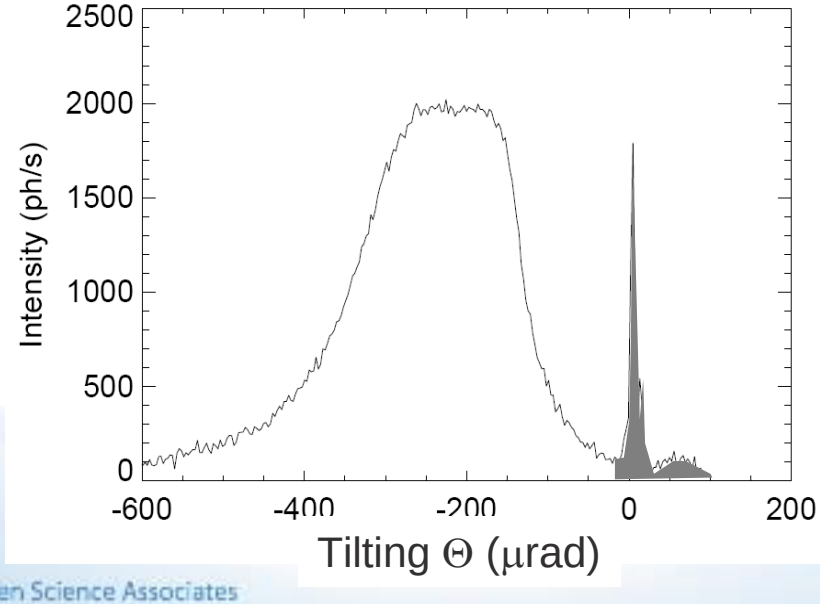


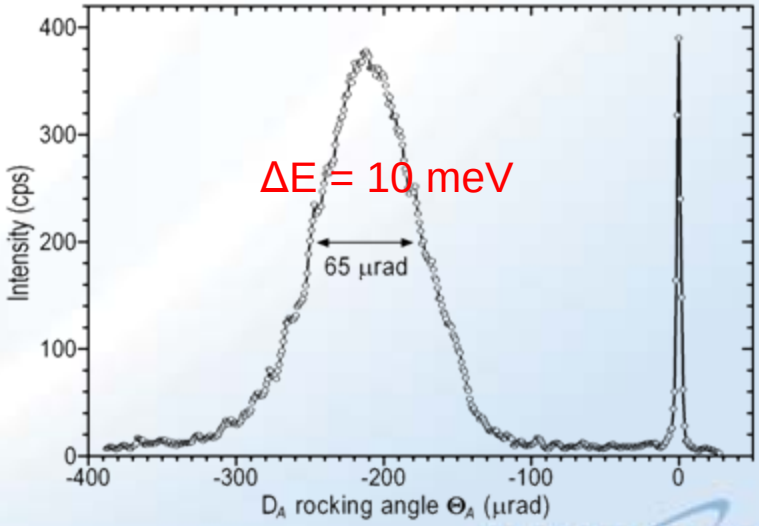
First CDW-CDW Test at NSLS

- **Proof of Principles:**
  - → Collimation effect of C crystal
  - → Enhanced Bormann transmission of W crystal
  - → Most importantly, the dispersion effect of the D crystal
  - → ΔE = 10 meV achieved, possible causes being analyzed



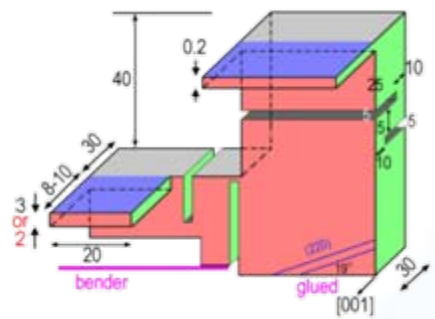


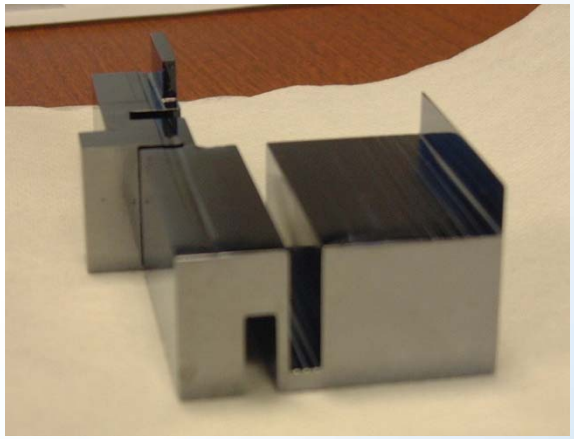


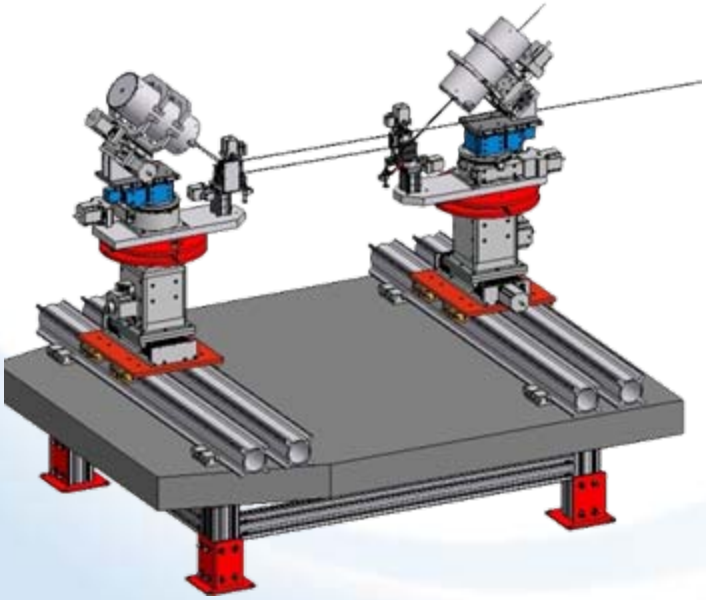


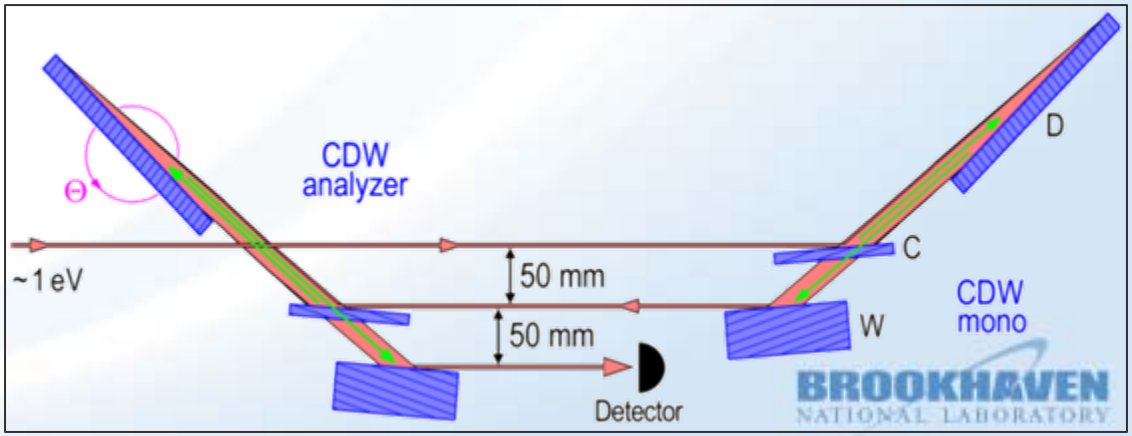
New CDW-CDW Test (in progress)

- Verify resolution function and sharp tail **with new optical path**
- Aim for a more controlled study of the optics to understand conditions needed to achieve the targeted energy resolution
  - → Temperature control oven for D crystal
  - → New C/W crystal to minimize strain
  - → New control stages
  - → Dedicated R&D beamline



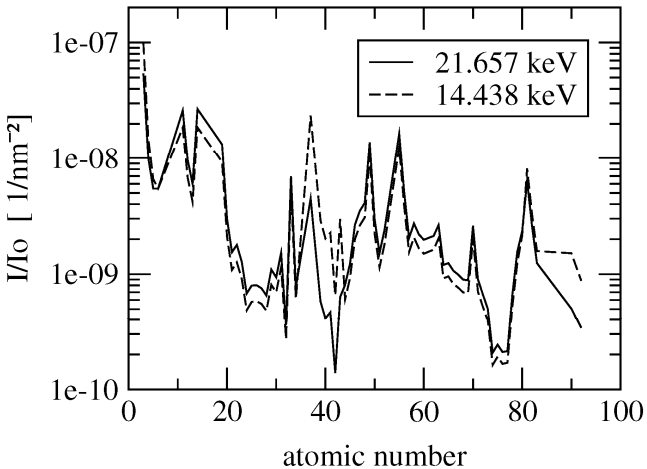






Count Rate Estimates (9.1 vs 21.7 keV)

- Given the same q resolution, I/I_0 is proportional to analyzer solid angle and sample absorption length (volume):



$$\frac{I}{I_0} = \frac{r_e^2 \hbar^2 c^2 (\delta q)^2}{2eE_i^2} Z^2 \rho l_{abs} \frac{k_B T}{M\upsilon^2}$$

$$\frac{(\delta q)^2}{E_i^2} = \left(\frac{2}{\hbar}\delta\vartheta\right)^2 \rightarrow \boxed{\frac{I}{I_0} \propto (\delta\vartheta)^2 l_{abs}}$$

H. Sinn, J. Phys. Cond. Mat. 13, 7525 (2001)

Actual count rate obtained on ID28 at ESRF for four sample systems

Sample	l_{abs} (µm) (9.1 keV)	l_{abs} (µm) (21.7keV)	Thickness (µm)	Solid Angle mrad^2	Int. @ ΔE (Ph/S @ meV)	Count Rate (cnts/sec)
v-SiO_2	190.2	2454.7	~400	~30	4×10^9 @ 1.6	~0.2 @21.7 keV
H_2O (HP)	1468	17165	~700	~30	4×10^9 @ 1.6	~1.0 @21.7 keV
La_2CuO_4	7.13	75.3	~140	~30	2.7×10^{10} @ 3.3	~0.5 @ 17.8 keV
Solid O_2	-	-	~30	~30	1.0×10^{10} @ 3.3	~1.5 @ 17.8 keV

(Data courtesy of M. Krisch)

Count Rate Estimates (preliminary)

Estimated count rate for NSLS-II instrument at 1 meV resolution

Sample	I_{abs} Ratio Gain	Solid Angle Gain	Intensity Gain @ 1meV	Count Rate (cnts/sec)
v-SiO_2	~ 0.5	~0.85	~6.4	~0.5
H_2O (HP)	~ 1	~0.85	~6.4	**~5.4**
La_2CuO_4	~ 0.1	~0.85	~2	~0.08
Solid O_2	~1	~0.85	~5.3	**~6.8**

Estimated CR = Mea. CR x I_{abs} Gain x SA Gain x Int. Gain

- Based on actual count rates obtained at ID28 of ESRF
- Assumptions:

→ Sample volume determined by environment (eg., HP DAC) or by absorption

→ New optics scheme delivers flux of 1.6×10^{10} ph/sec at 1 meV resolution (assuming 10% overall efficiency)

→ New analyzer scheme is as efficient as state-of-the-art backscattering analyzers

Better performance for low-Z materials!

Key Scientific Applications

- Dynamics of liquids and disordered systems
  - The frequency and wavelength dependence of sound absorption in glasses
- Vibrational dynamics under high pressure and temperature
  - Advantages of x-rays (small beam size, high penetration)
- Phonon and phonon-damping in bio-materials
  - Phonon-assisted molecule transport across lipid bilayers

→ *Extra: Similar schemes may provide sub-meV resolution x-rays over 6-10 keV for hard x-ray photoemission*