Non-linear science capabilities at chemRIXS and qRIXS endstations

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Non-Linear Multidimensional Methodologies for Studying Chemical Sciences

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Outline

1. X-ray beam parameters at NEH2.2.

2. Description of chemRIXS and qRIXS endstations.

3. Options for non-linear experiments.

NEH2.2. beamline

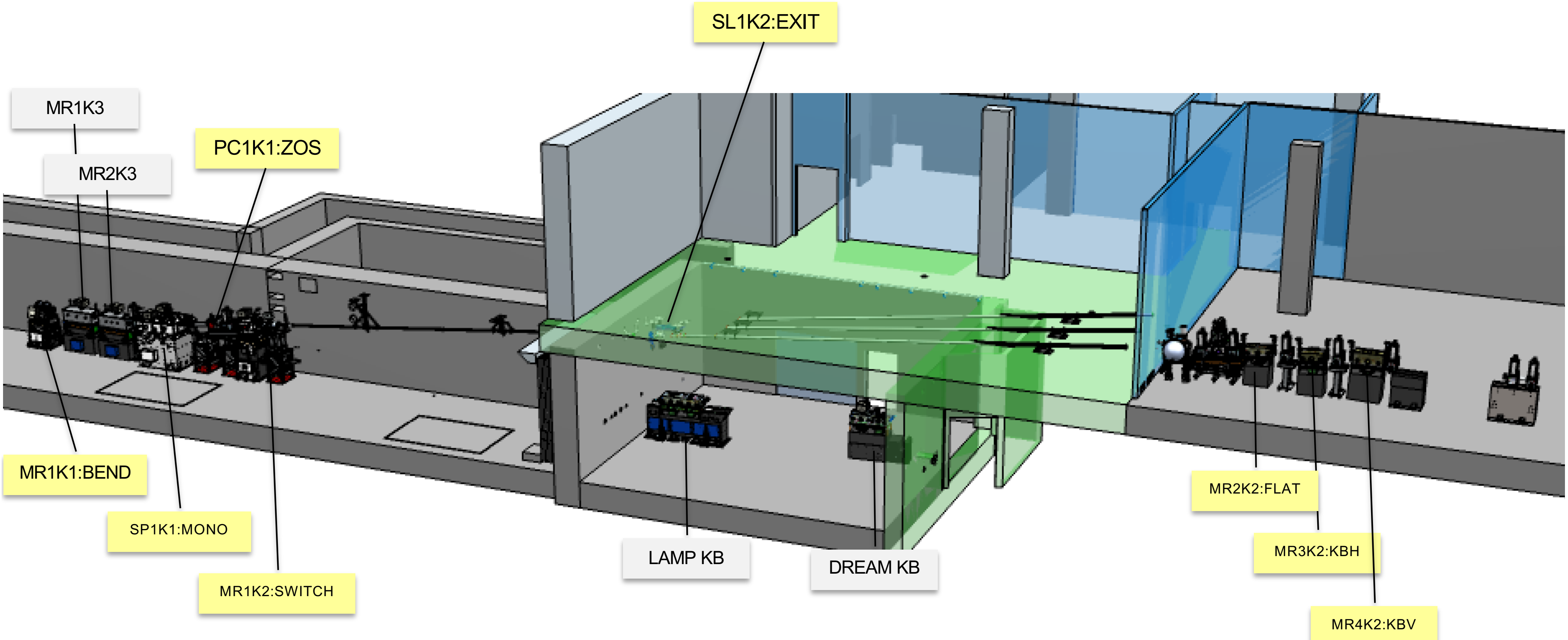
NEH2.2. accepts X-rays from the Soft X-ray Undulator (SXU).
Photon energy range 250 – 1600 eV.

Three beamline configurations:

1. Non-monochromatic (pink beam) mode.
   - No temporal pulse stretching.
   - Suitable for two-color X-ray experiments (co-linear).
2. Low-resolution monochromatic mode (LR).
   - Resolving power ~5000.
   - Temporal pulse stretching <50 fs (transform limit x2).
   - Three gratings for low-, mid- and high-energy (LE, ME, HE).
3. High-resolution monochromatic mode (HR).
   - Resolving power ~50000.
   - Temporal pulse stretching ~500 fs (transform limit x2).
   - Three gratings for low-, mid- and high-energy (LE, ME, HE).

NEH2.2. core capability is (linear) pump-probe experiments.
-> Potential for non-linear experiments, particularly in the non-monochromatic mode.

NEH2.2. beamline optical layout



- 6 mirrors + 1 grating (or mirror) in total.
- Mirror coating B_4C (<1000 eV), optionally Rh for the last 4 mirrors (>1000 eV).
- Horizontal and vertical focusing controlled with two KB mirrors.
- H/V spot size at the experiment can be adjusted between 2 – 1000 μm.

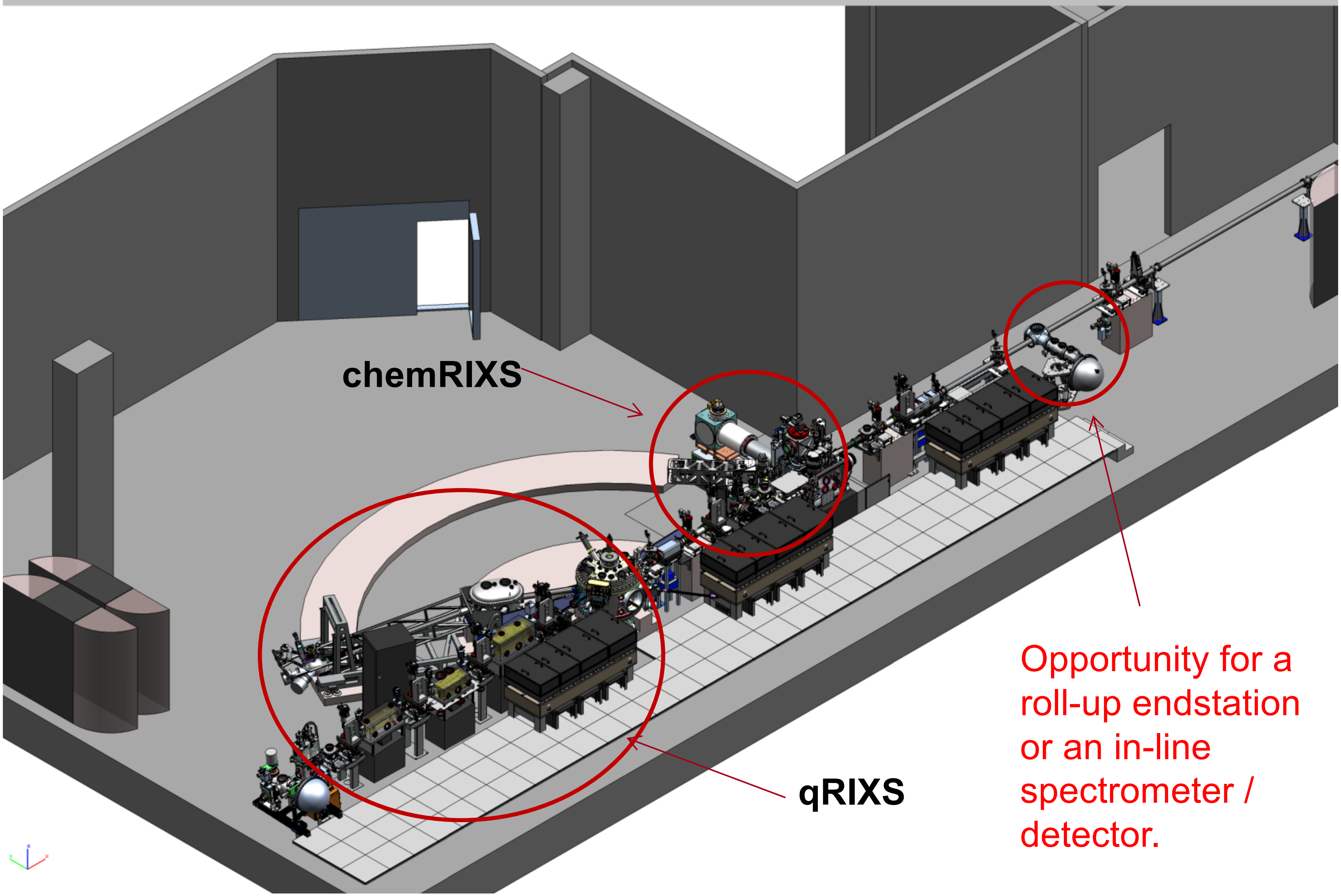
X-ray parameters

Characteristic X-ray fluences at 500 eV with a small spot.

	After SXU Photon energy 500 eV				At the experiment Interaction Point Spot size 2 x 2 μm^2			
FEL Mode	Pulse energy (μJ)	Pulse length (fs)	Comment	BL mode	Pulse energy (μJ)	Pulse duration (fs)	Fluence (J/cm^2)	Peak fluence (TW/cm^2)
SASE	2000	50	E/ΔE = 200	Non-mono. LR (RP=5k) HR (RP=50k)	500 15 0.6	50 60 400	12500 375 15	250 6.25 0.0375
Seeded	50	20	E/ΔE = 3500	Non-mono. LR (RP=5k) HR (RP=50k)	12.5 10 4	20 36 400	312.5 250 100	16 7.0 0.25
Single Slotted foil	15	2	Single SASE spike, Spike rate 20%	Non-mono.	4	2	100	50
XLEAP	20	0.5	ΔE = 5 eV	Non-mono.	5	0.5	125	250
SASE Split Undulator	50 (per pulse)	30	Two pulses: Time sep. 0 – 800 fs, Energy sep. up to x2	Non-mono.	12.5	30	312.5	10
XLEAP Split Undulator	15 (per pulse)	0.5	Two pulses: Phase coherent <50 fs time separation	Non-mono.	4	0.5	90	190

FEL parameters taken from the Run 19 update (Oct. 1, 2020). More info: https://lcls.slac.stanford.edu/sites/lcls.slac.stanford.edu/files/LCLS-Parameters-Run-19.pdf

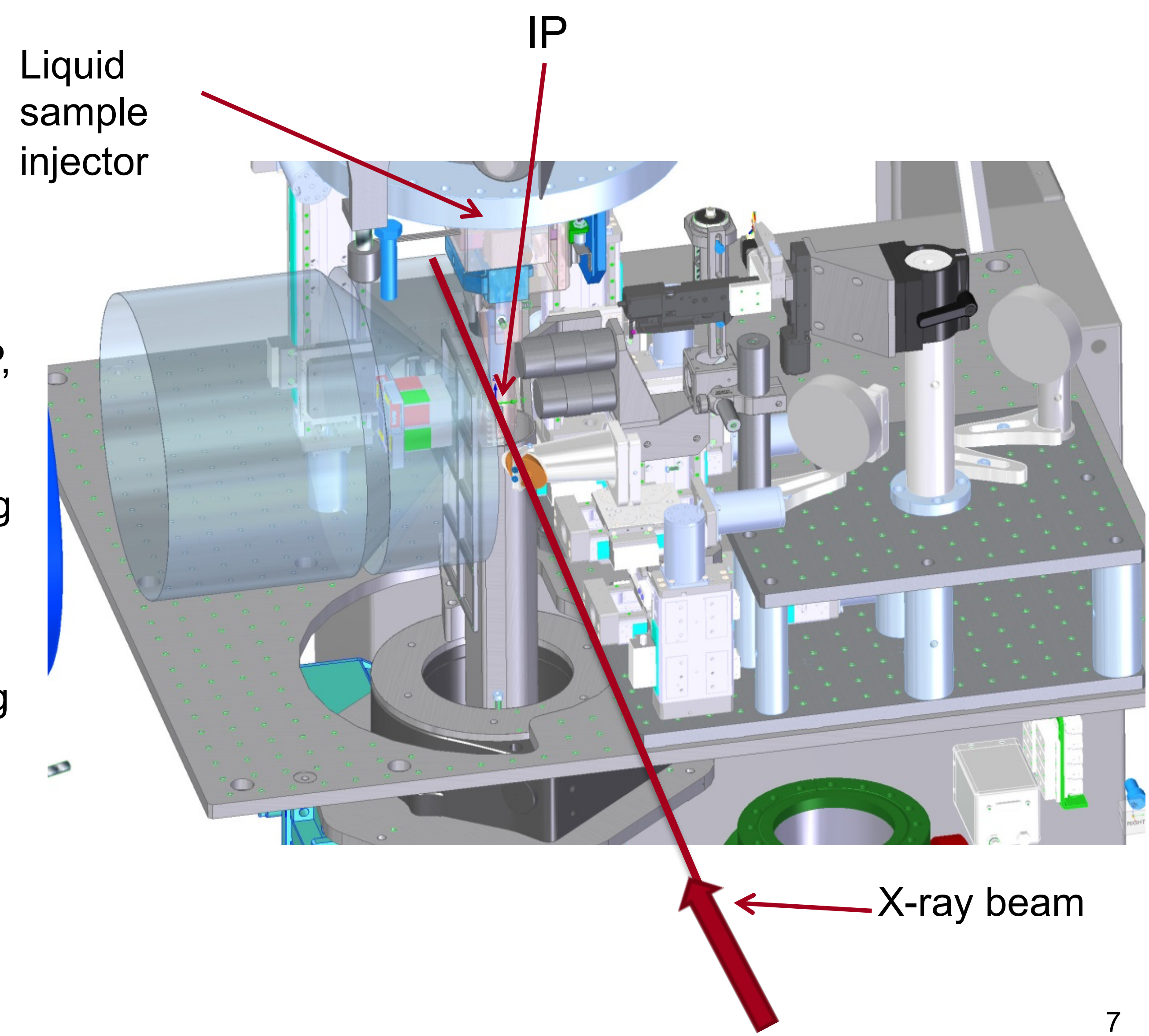
NEH2.2. endstations - hutch layout



chemRIXS endstation

Capabilities:

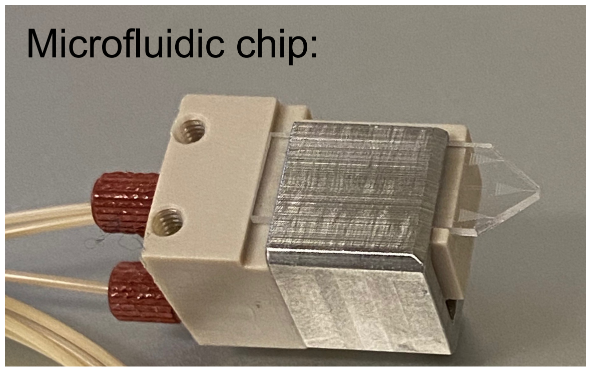
- In-vacuum liquid jets
- Pump laser infrastructure, co-linear incoupling.
- Point detectors: APD and MCP, 1 MHz readout rate
- Downstream CCD for detecting a transmitted beam
- Grating spectrometer (RP~2000). Mounted at 90 deg scattering geometry in the horizontal plane or optionally in-line.
- Sample Viewing: on-axis & perpendicular; infrared illumination

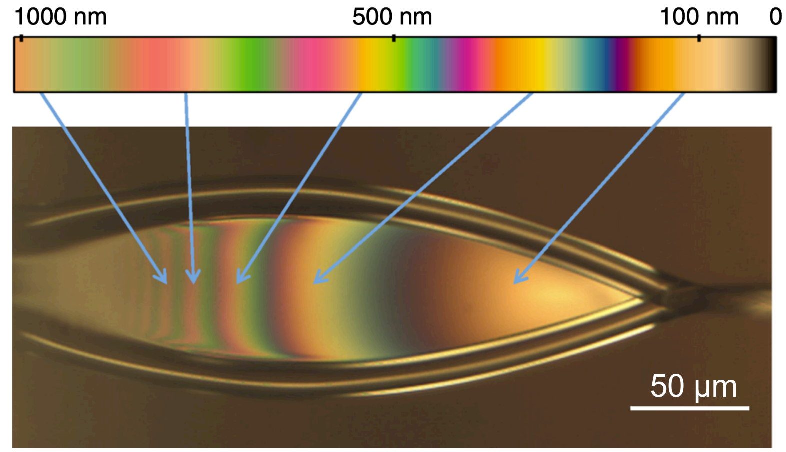


chemRIXS: sheet and cylindrical jets

Gas accelerated sheet jets

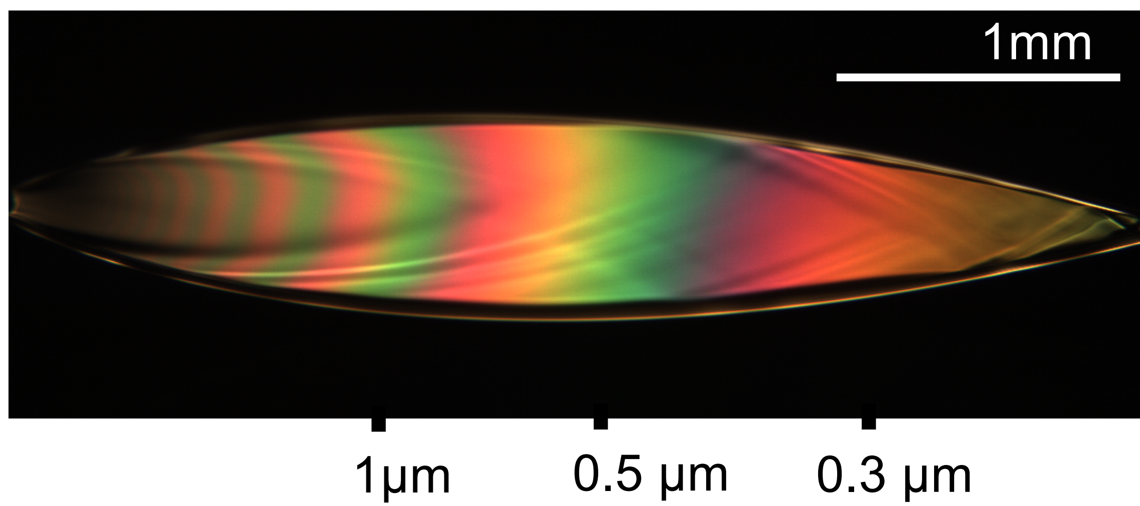
- Thinnest sheets: 0.1 – 1 µm
- 250 µl/min flow rate





Converging nozzle sheet jets (not gas accelerated)

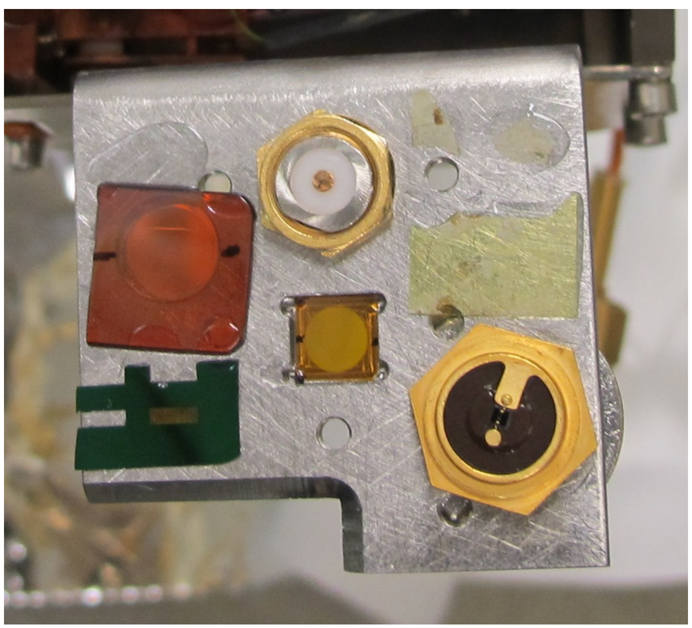
- Thickness: 0.2 – 2 µm
- 2 to 4 ml/min

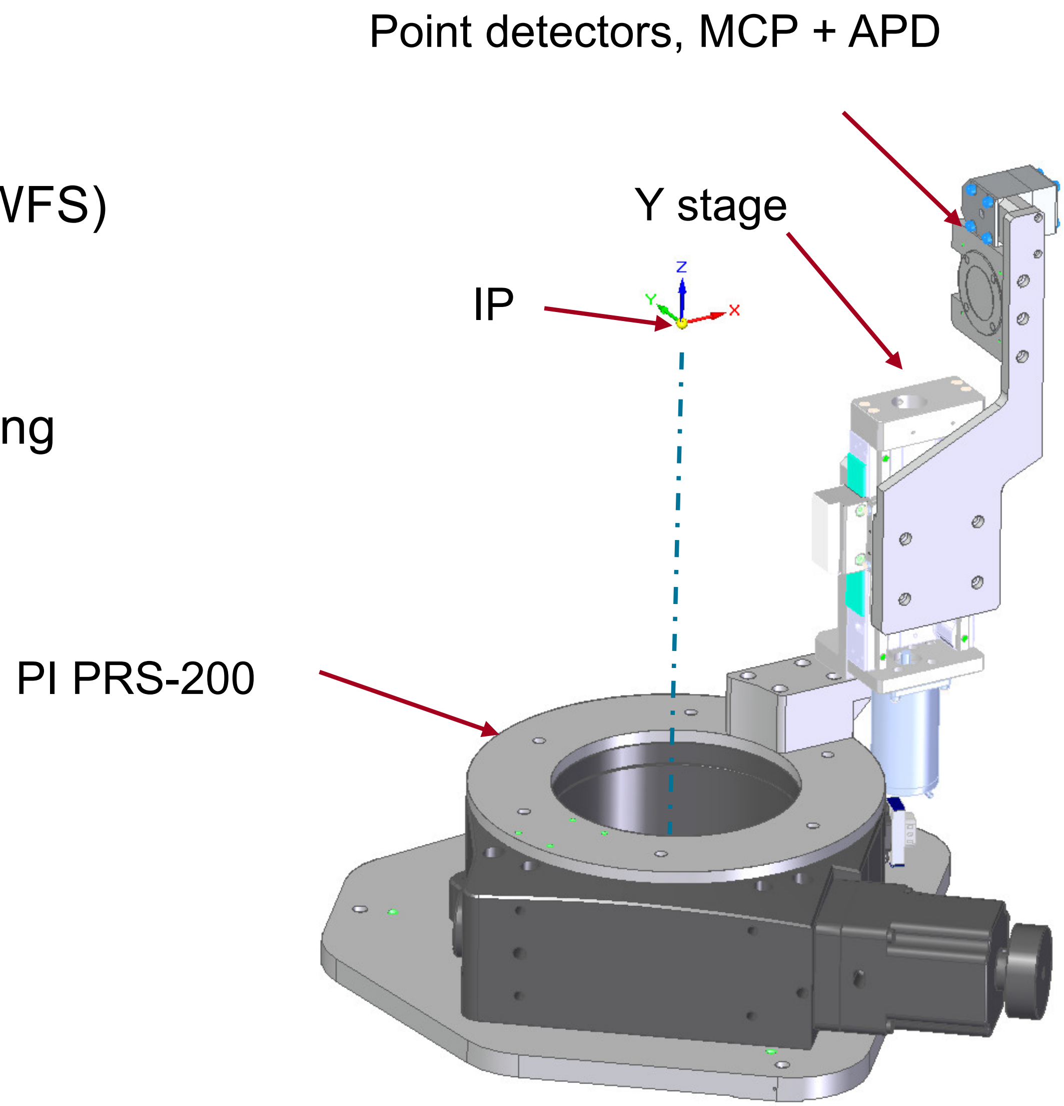


LCLS SED website:
https://lcls.slac.stanford.edu/sed

chemRIXS: Overlap Diagnostics and Point Detectors

- YAG for x-ray visualization
- Frosted YAG for laser
- Pinhole for wavefront sensor (WFS)
- Timing Crystals
- Knife edge blades
- Fast photodiode for coarse timing

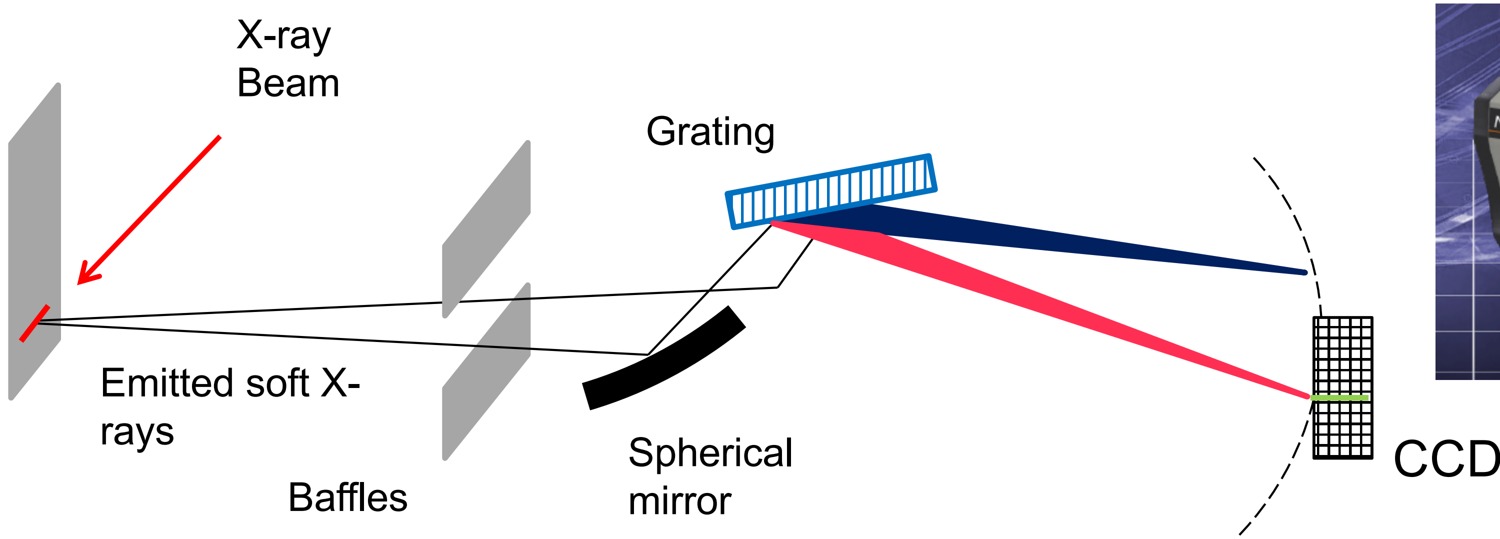


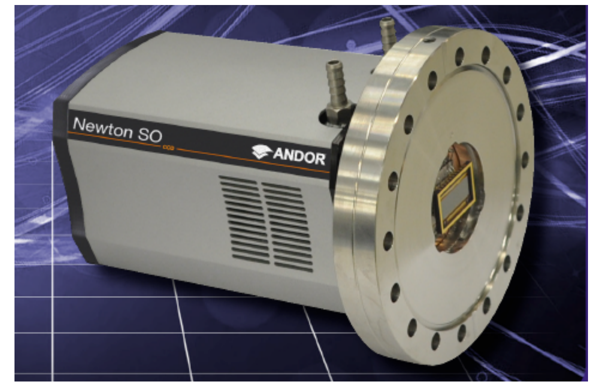


chemRIXS: X-ray emission VLS spectrometer

1200 l/mm VLS

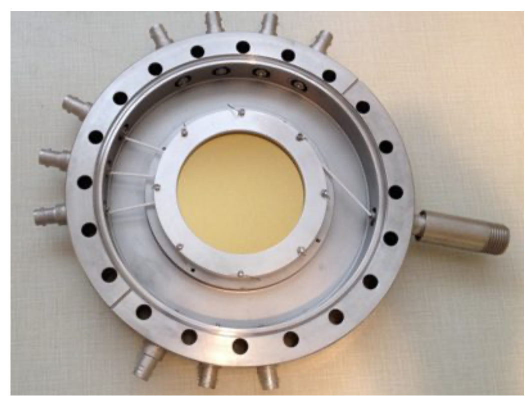
Resolving power ~2000





Standard configuration:
Mounted at 90 deg scattering geometry in the horizontal plane for fluorescence detection.

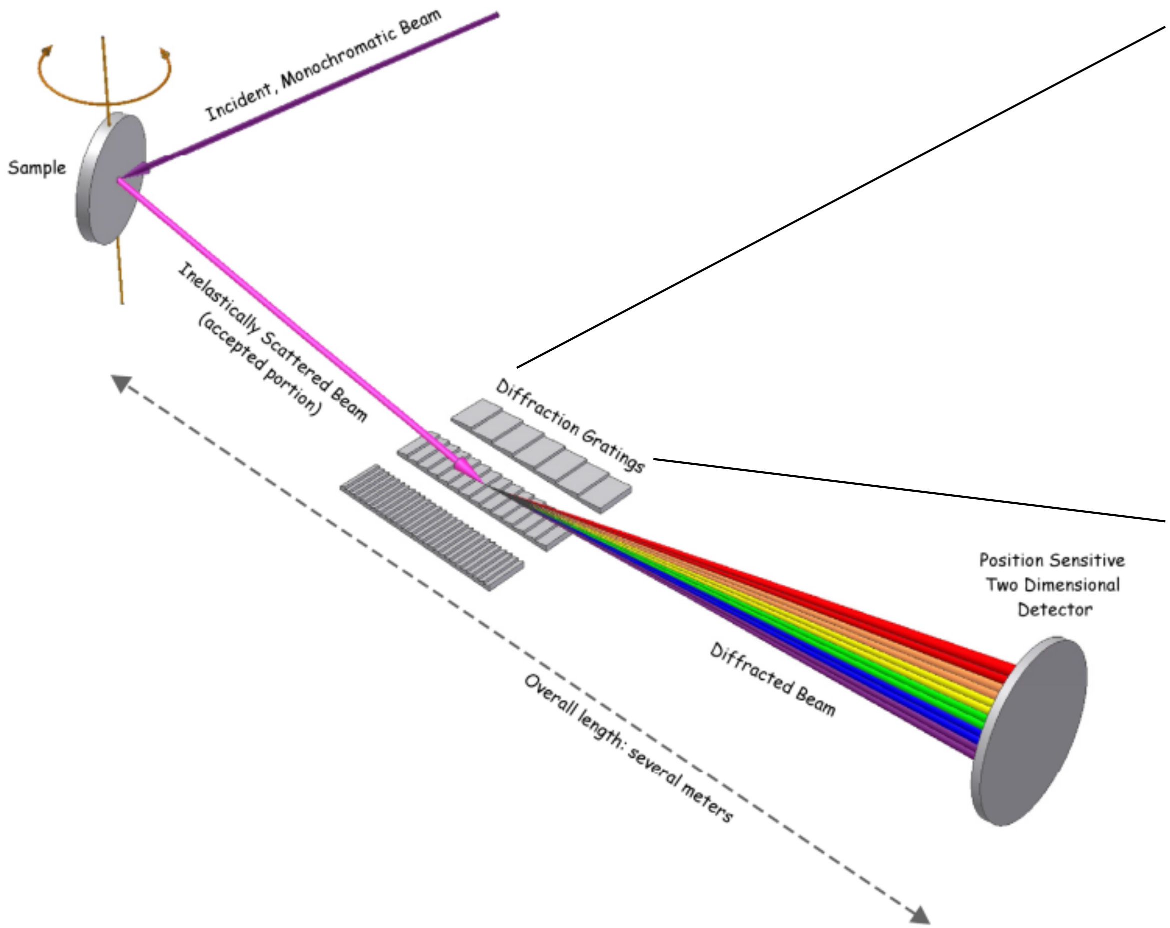
In-line option:
Can be mounted downstream of the endstation at 0 deg scattering geometry (in-line) in the horizontal plane.



Alternative detector:
Large MCP +
Phosphor screen +
optical camera

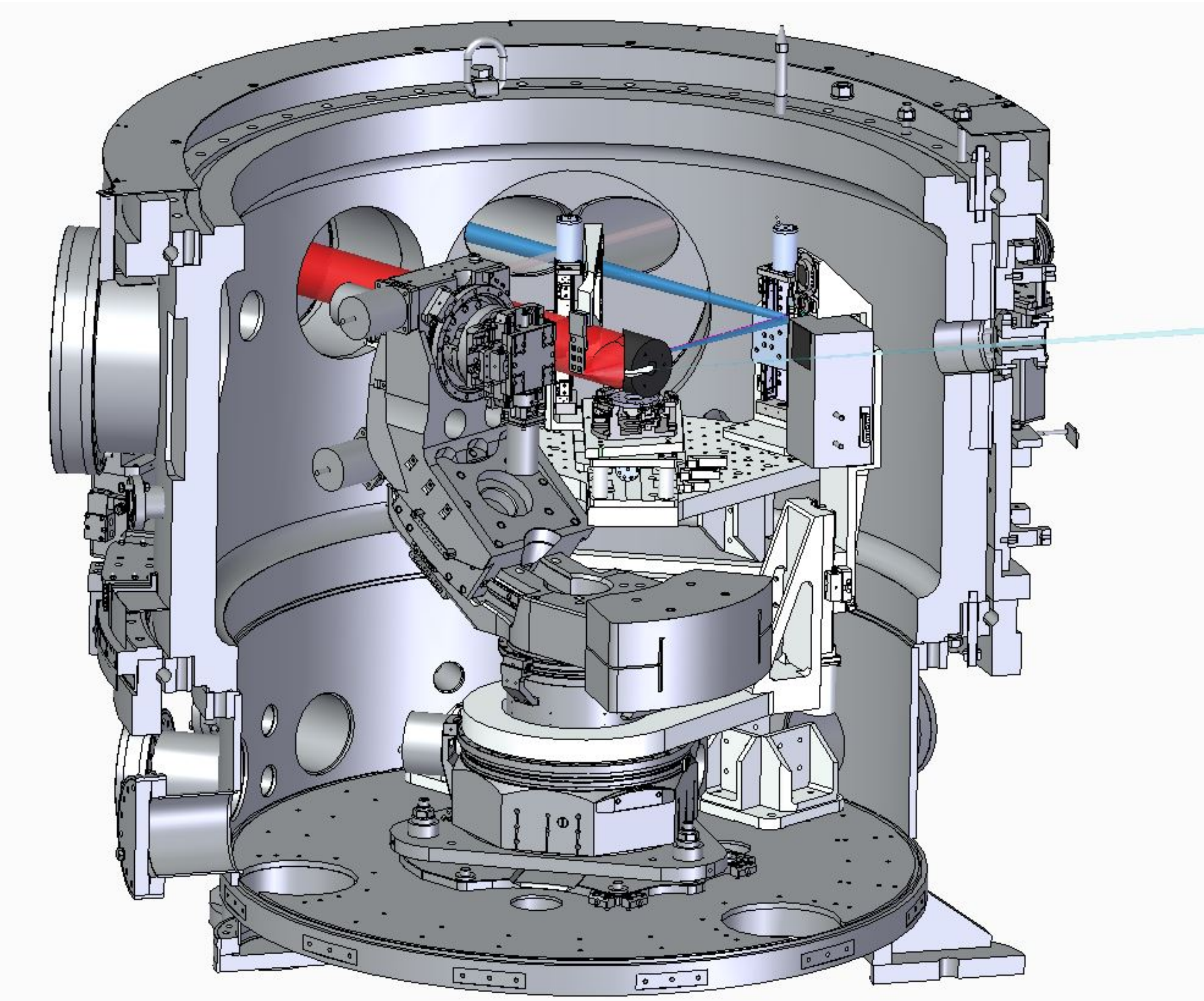
Larger acceptance,
but RP ~500

qRIXS endstation



- Sample positioning and cooling
- Characterization: X-ray Absorption Spectroscopy, Resonant Elastic X-ray Scattering
- Spectrometer arm (grating & detector) on a movable arm
- Laser in-coupling for a wide range of wavelengths
- Diagnostics

qRIXS: sample chamber



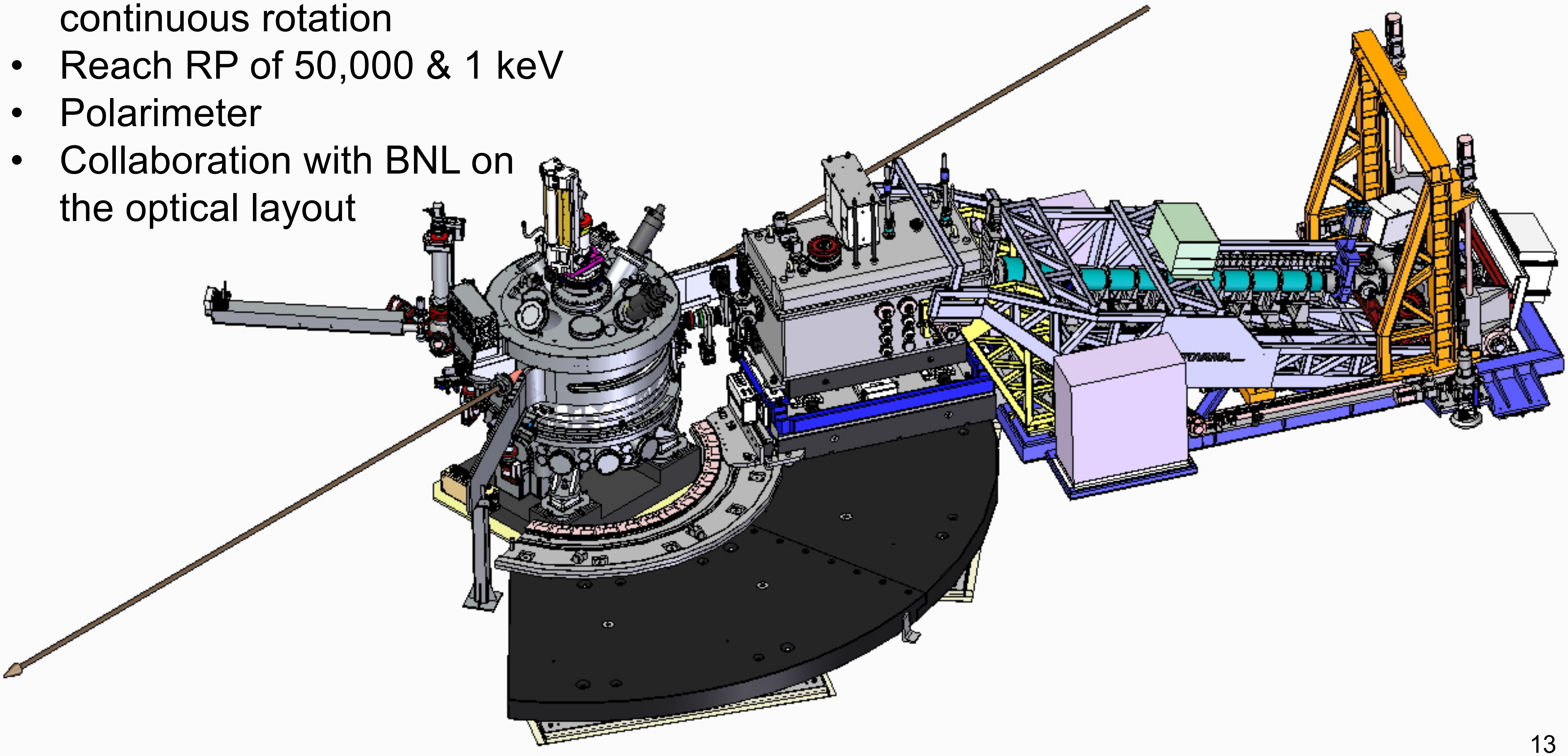
Capabilities:

- Sample motion: 6 degrees of freedom: in-vacuum diffractometer
- UHV (<3 x 10^{-9} Torr)
- Cryostat: 20 K
- Long-wavelength laser in-coupling
- In-vacuum detectors for peak finding and XAS in fluorescence yield mode

qRIXS: spectrometer arm

Capabilities:

- Scattering angle 40 – 150 deg: continuous rotation
- Reach RP of 50,000 & 1 keV
- Polarimeter
- Collaboration with BNL on the optical layout



Non-linear science options

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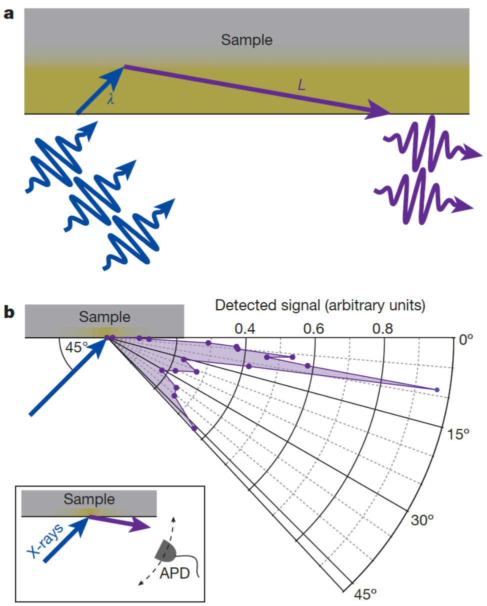
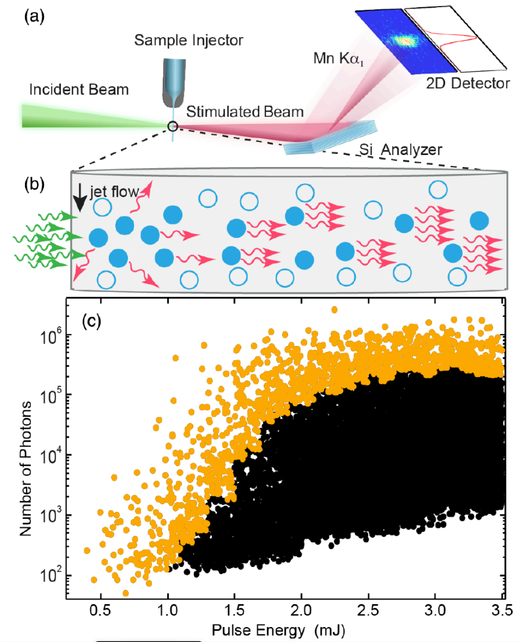
1. Stimulated XES/RIXS (amplified spontaneous emission)



- Soft X-rays
- Solid sample

Beye et al., *Nature* **501**, 191 (2013)



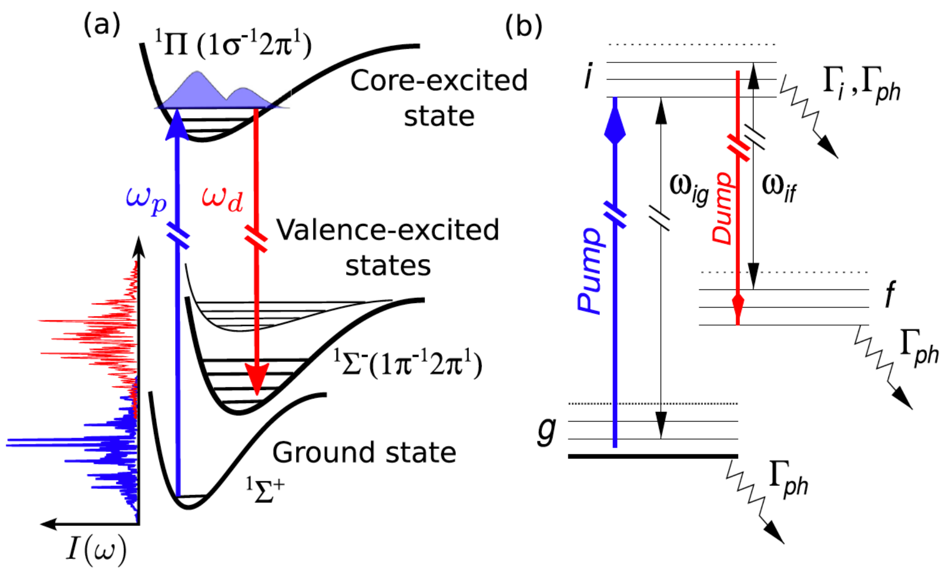
- Hard X-rays
- Liquid sample

Kroll et al., *PRL* **120**, 133203 (2018)

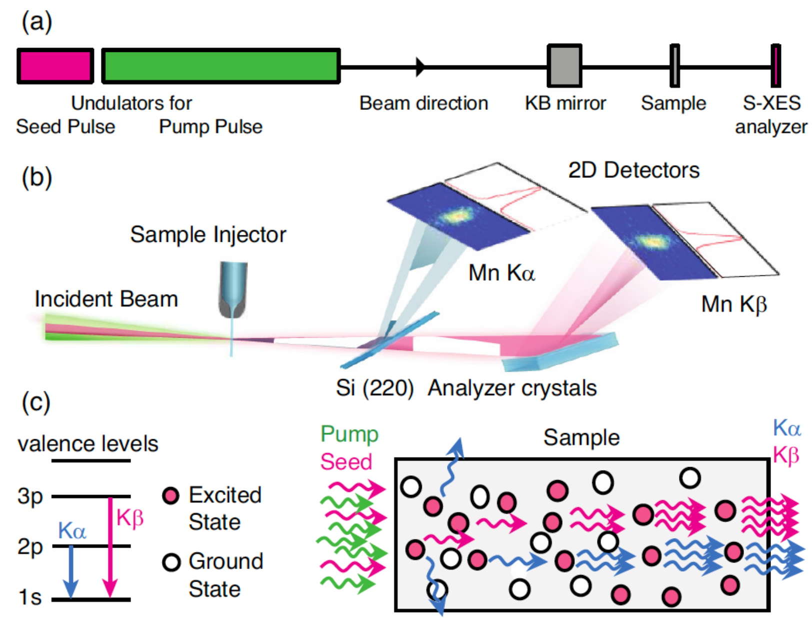
- Amplification of low-yield fluorescence signals (superfluorescence).
- Suppression of non-radiative decay channels and electronic damage.
- Demonstrated chemical sensitivity.

Non-linear science options

2. Seeded (pump-dump) stimulated XES/RIXS



Kimberg and Rohringer, *Struct. Dyn.* **3**, 034101 (2016)



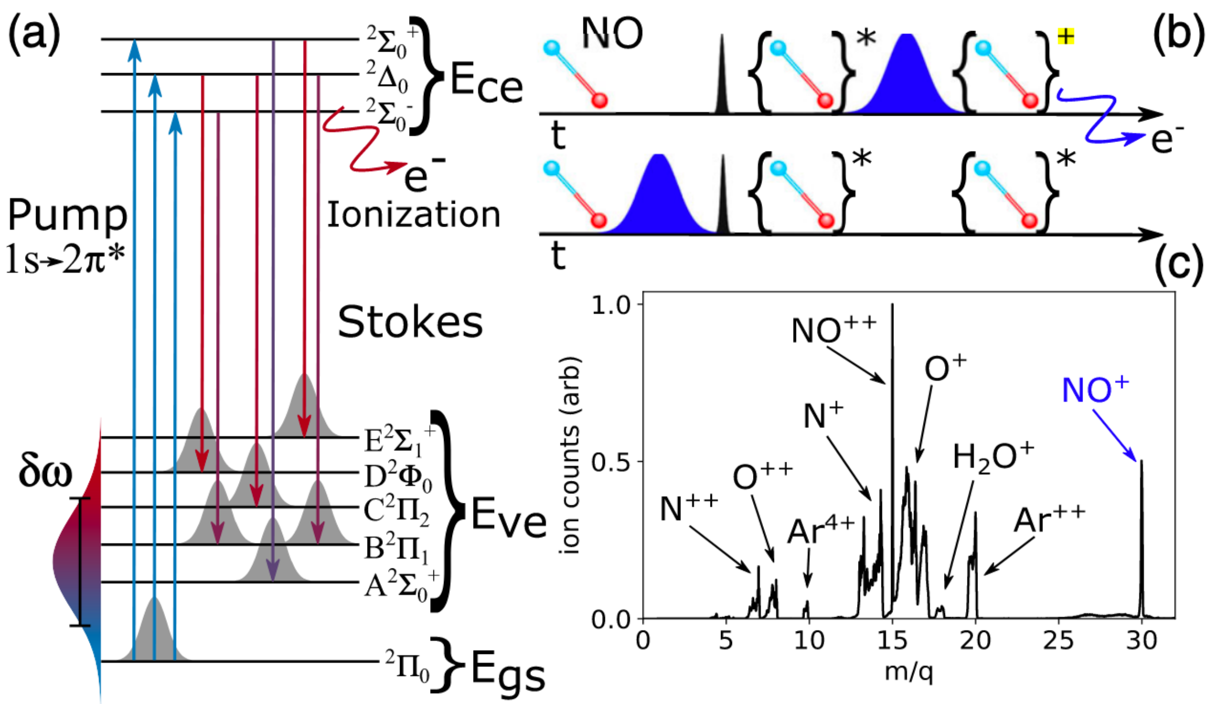
- Hard X-rays
- Liquid sample
- LCLS split undulator method

Kroll et al., *PRL* **125**, 037404 (2020)

- Stimulation of "forbidden" or low cross-section core-transitions that have low yield in spontanaous XES/RIXS.

Non-linear science options

3. Impulsive X-ray Raman pump / laser probe or X-ray probe



- Soft X-rays
- Gas phase
- Coherent broad-band (eVs) X-ray pump pulses
- LCLS XLEAP method

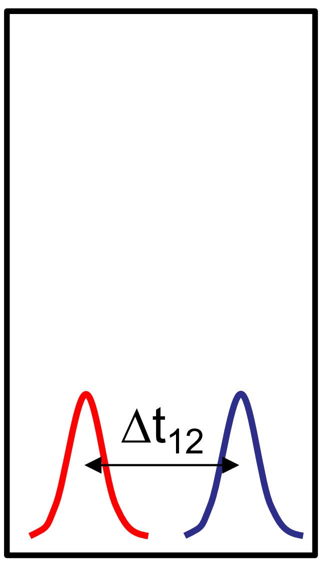
O'Neil et al., *PRL* **125**, 073203 (2020)

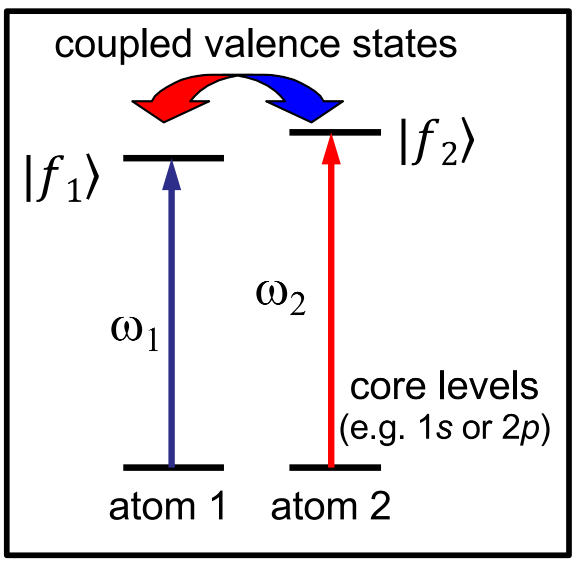
- Generation of localized valence electronic wavepacket allows probing of electronic dynamics and coupled nuclear dynamics.

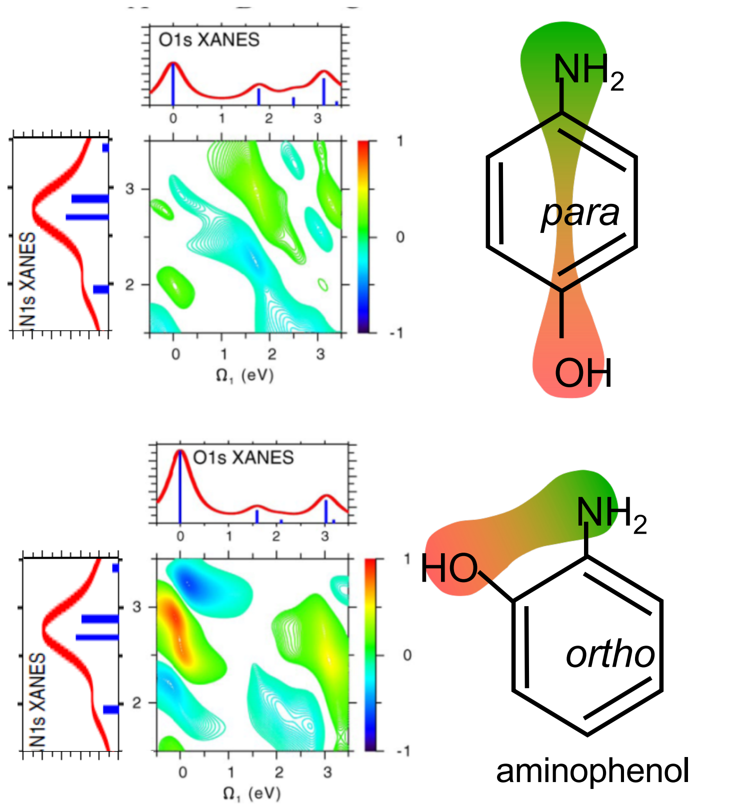
Non-linear science options

4. 2D coherent core-hole correlation spectroscopy

- Correlated valence electronic states spanning different atomic sites
- τ_{pulse} < core-hole lifetime
- Δt_{12} < core-hole lifetime (few fs)
- Two-color ultrashort pulses (resonant with different atomic/chemical species)
- Energy separation (few 100s eV)
- FWM two colors are coupled by correlated valence states







Schweigert and Mukamel, *PRL* **99**, 163001 (2007)

Non-linear science options

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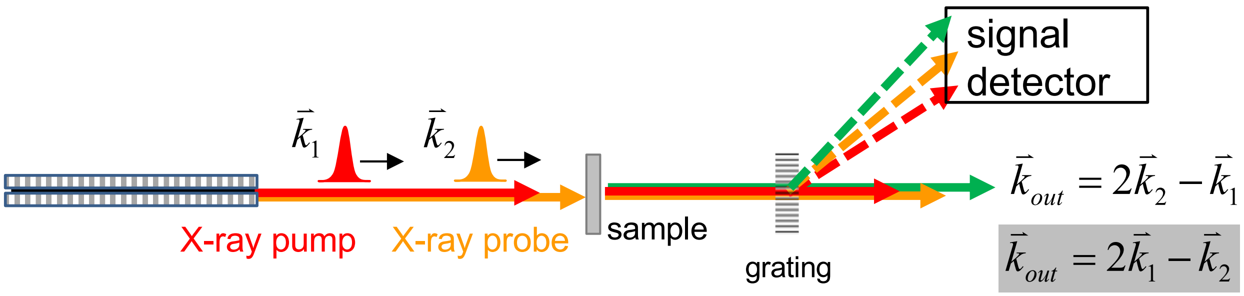
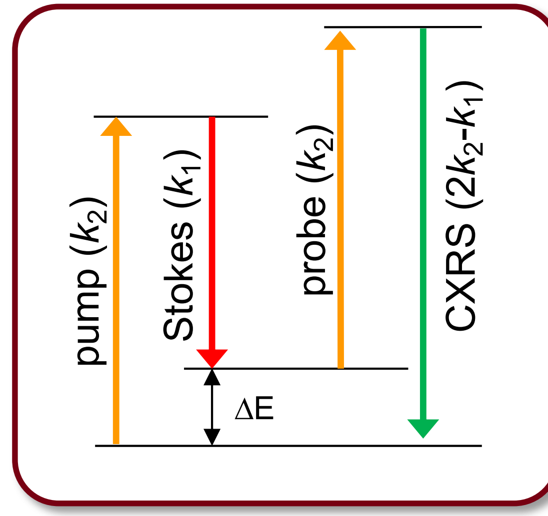
5. Co-linear two-color coherent X-ray Raman spectroscopy





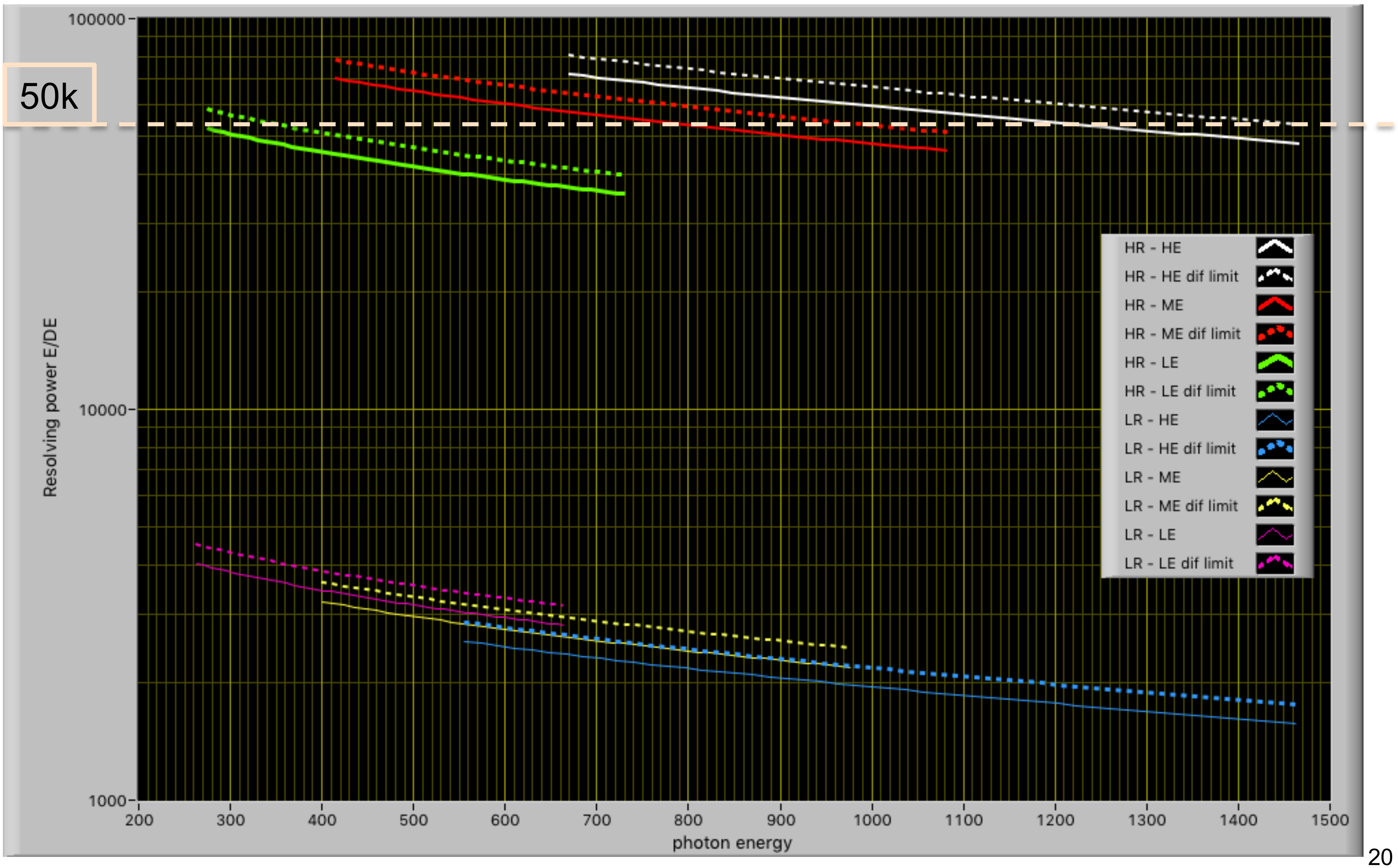
$$\vec{k}_{out} = 2\vec{k}_2 - \vec{k}_1 \qquad P^{(3)} = \chi^{(3)} E_1 E_2 E_2$$

- Choose k_1 and k_2 with an energy difference corresponding to a valence excitation.
- Scattering wavevector between the sites a and b: $q = 2\pi/(R_a - R_b) = k_2 - k_1 = k_{out} - k_2$
- Directly probes the coupling (transfer) of the valence excitation between the sites a and b

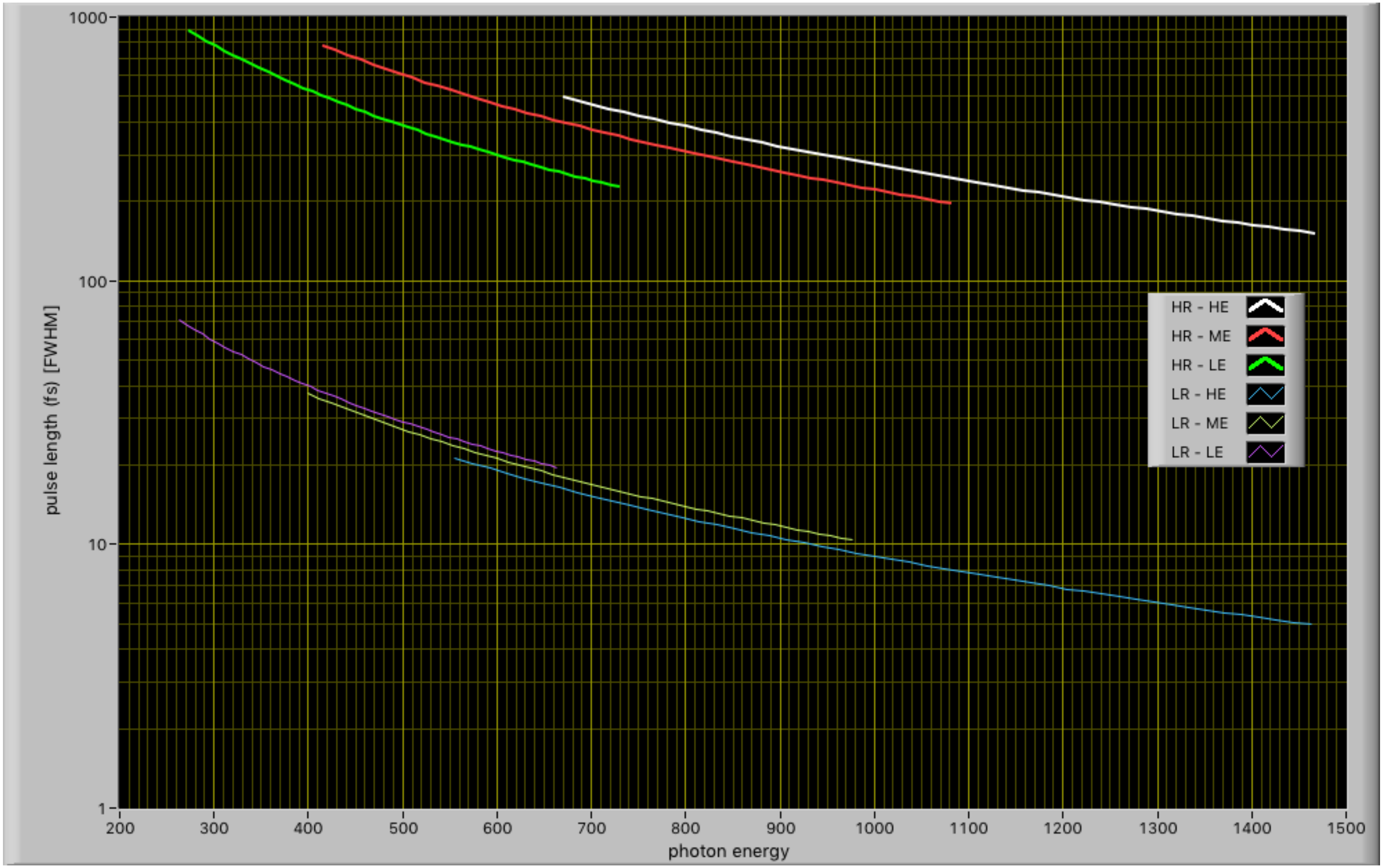
Tanaka and Mukamel, *PRL* **89**, 043001 (2002)

SUPPLEMENTARY SLIDES

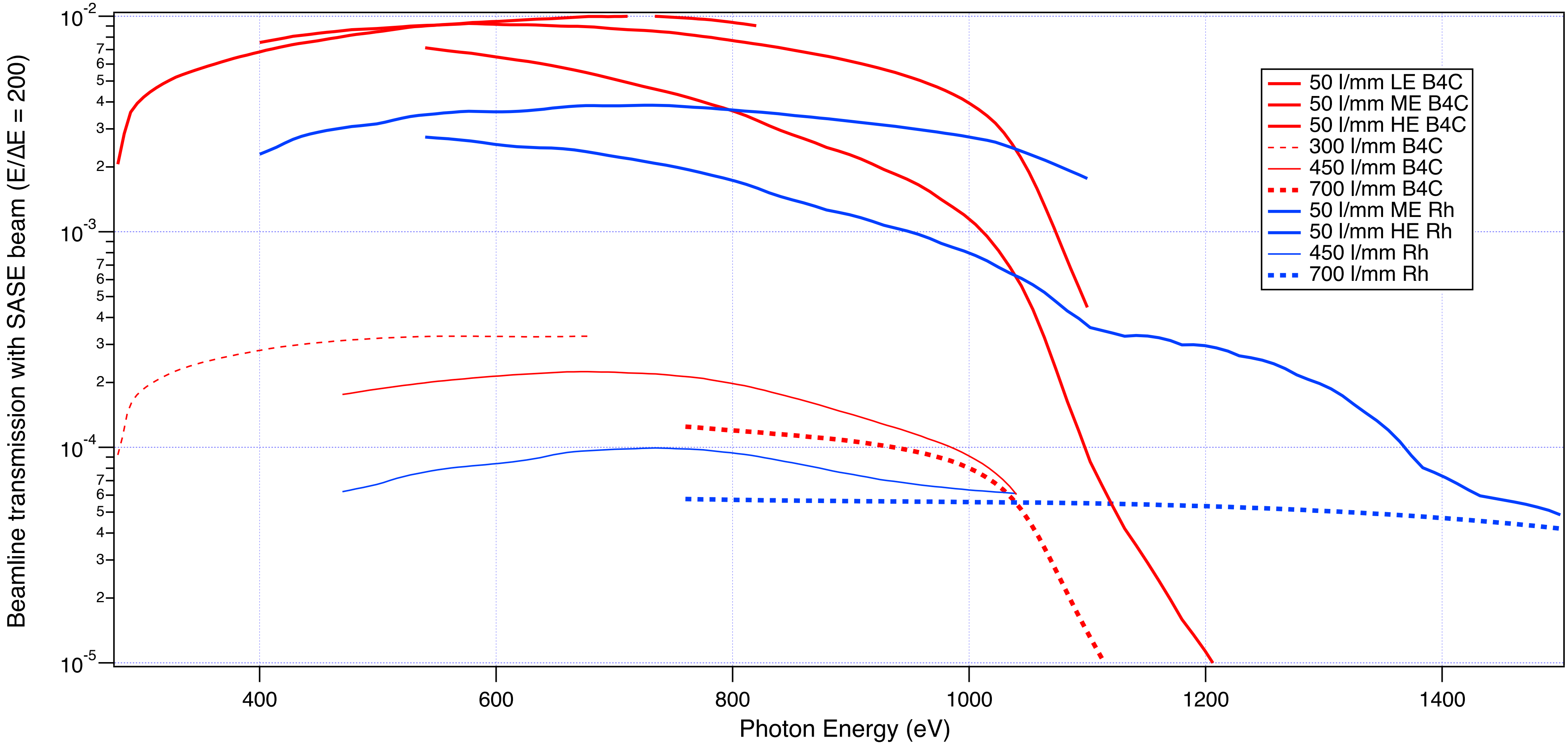
Resolving Power



Pulse length

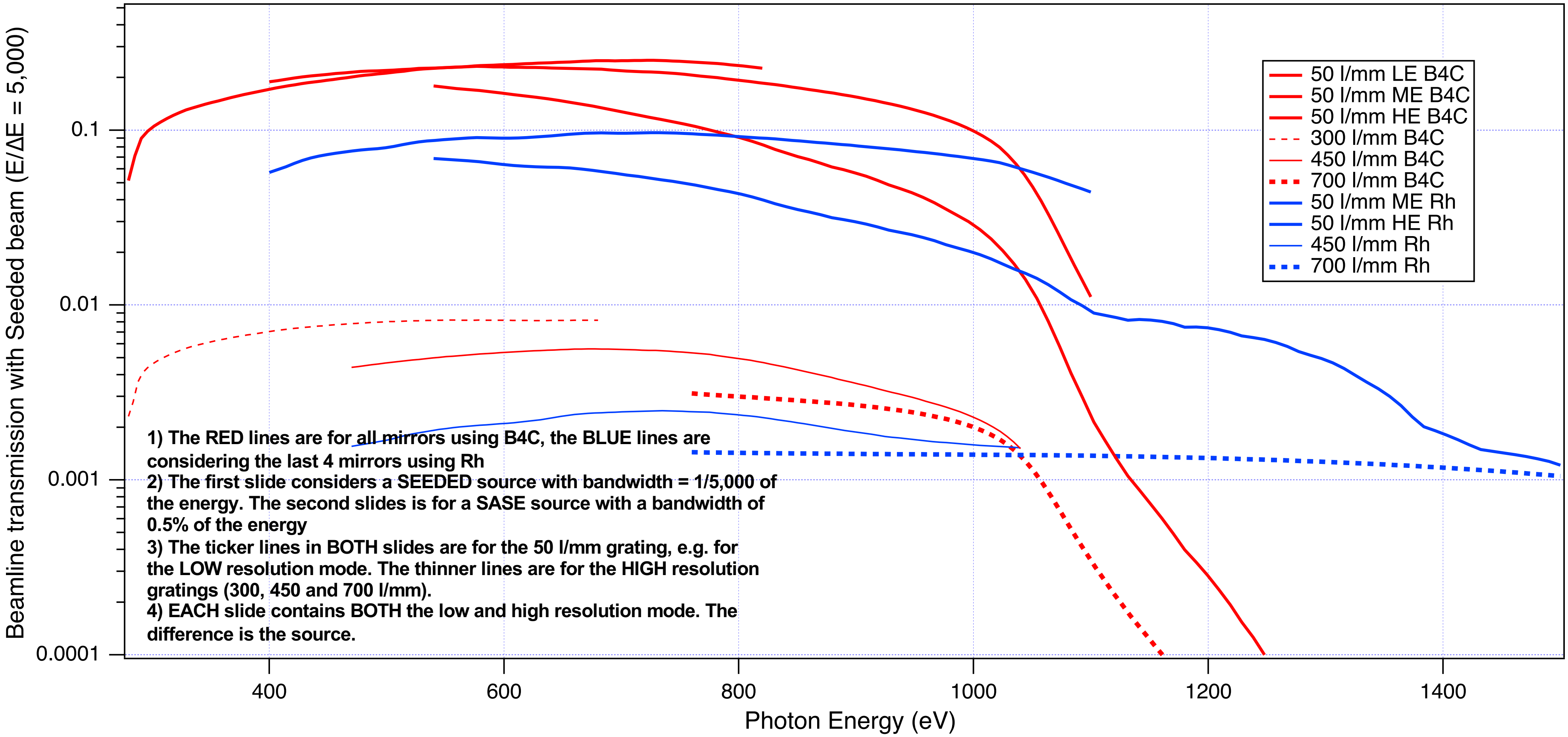


Beamline Transmission – SASE mode



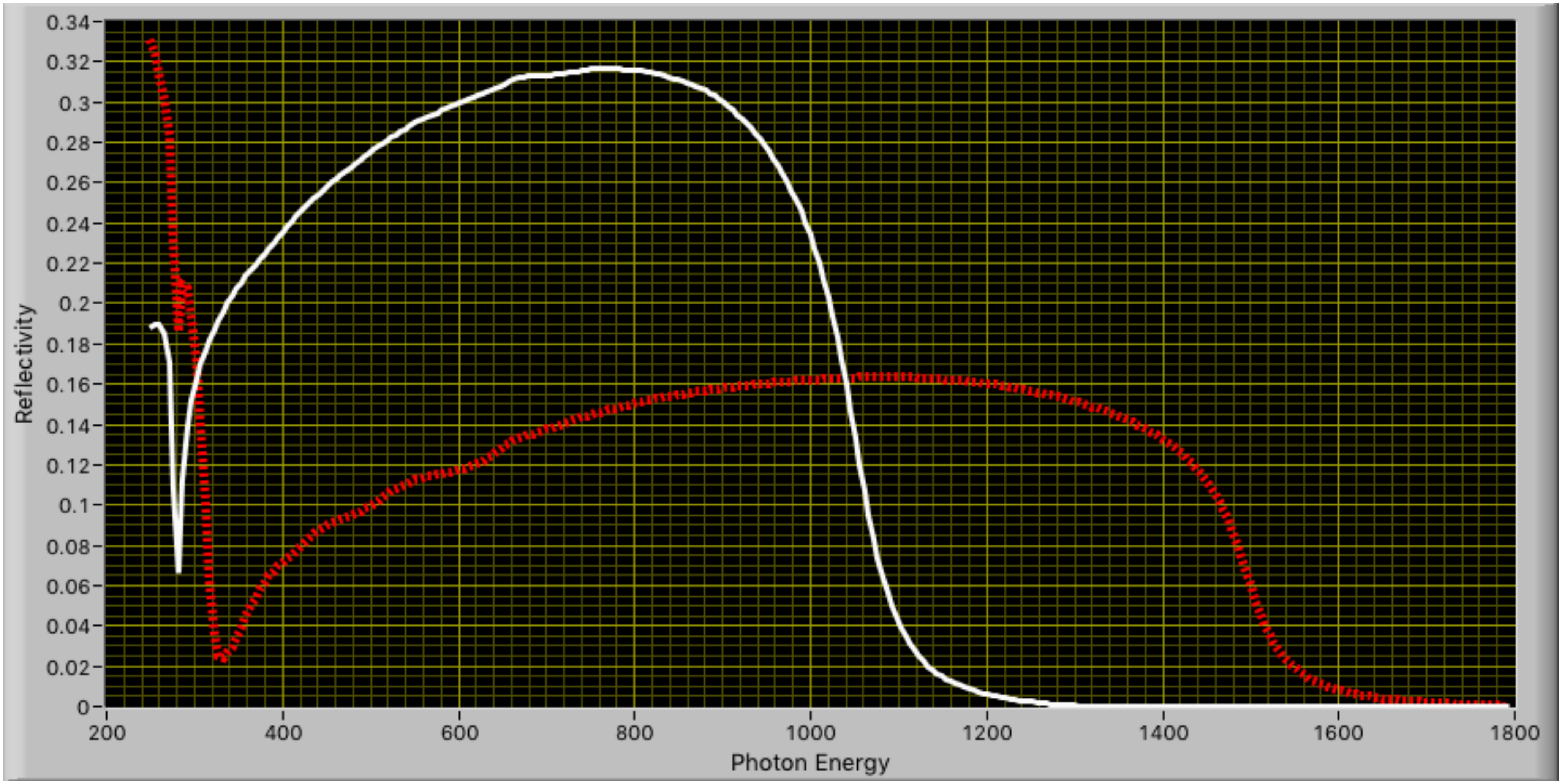
Including grating efficiency and source bandwidth (E/ΔE @ source = 200).
To have the power/pulse energy at qRIXS or chemRIXS, multiply the source power/energy by the numbers in this chart.
Resolving power corresponding to the data on slide 20.

Beamline Transmission – Seeded mode



Including grating efficiency and source bandwidth (E/ΔE @ source = 5,000).
To have the power/pulse energy at qRIXS or chemRIXS, multiply the source power/energy by the numbers in this chart.
Resolving power corresponding to the data on slide 20.

Zero order transmission (After M4K2)



White: All mirrors using B_4C coating

Red: M1K1, M2K1 and M3K1 using B4C; M1K2, M2K2, M3K2 and M4K2 using Rh

Geometrical Spot in the experimental chamber