Development of quantum sensor technology for synchrotron applications

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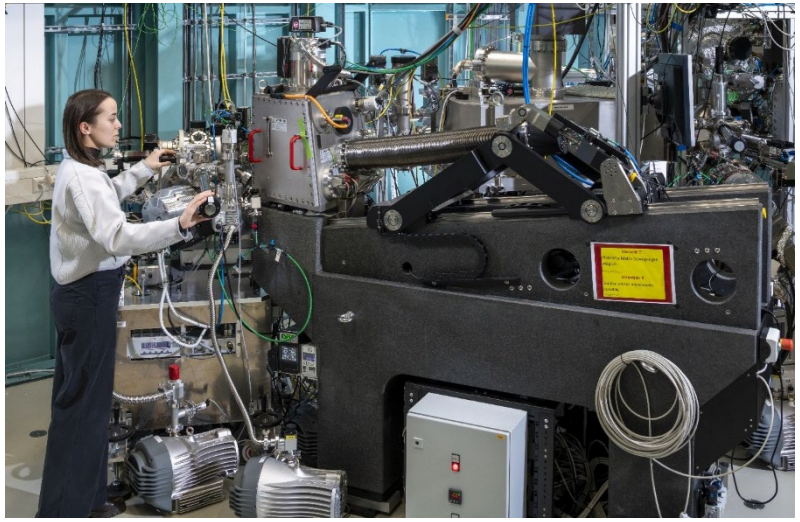
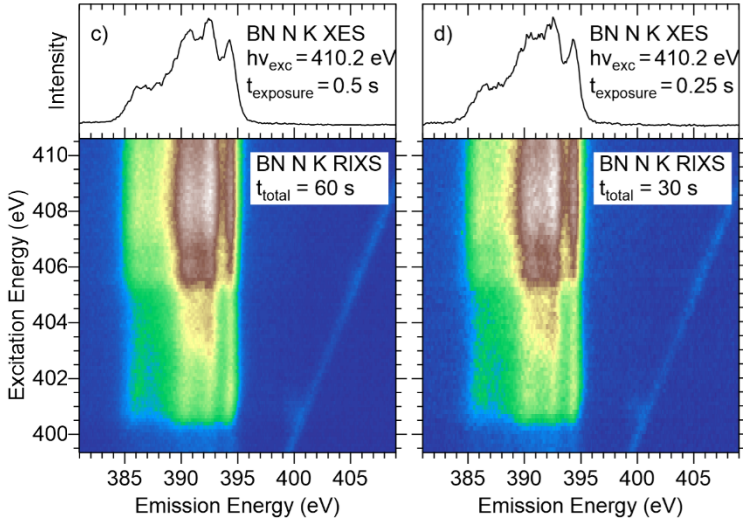
Institute for Chemical Technology and Polymer Chemistry (ITCP), Karlsruhe Institute of Technology (KIT)

Department of Chemistry and Biochemistry, University of Nevada, Las Vegas (UNLV)

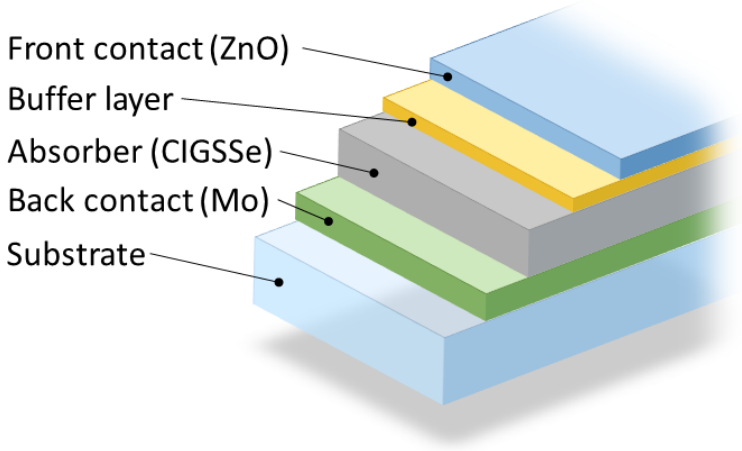


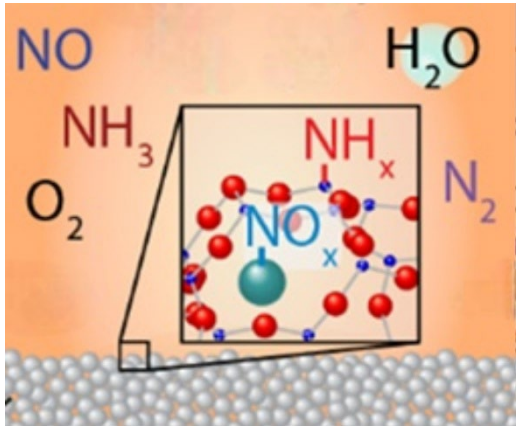


Karlsruher Institut für Technologie

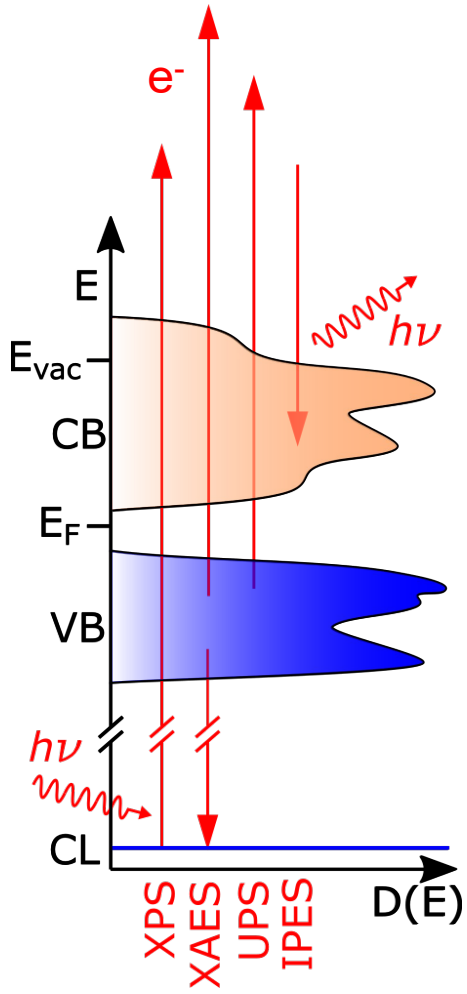
Electronic and chemical structure of applied material systems

- Charge-carrier transport
- Chemical bonding
- Interaction with light

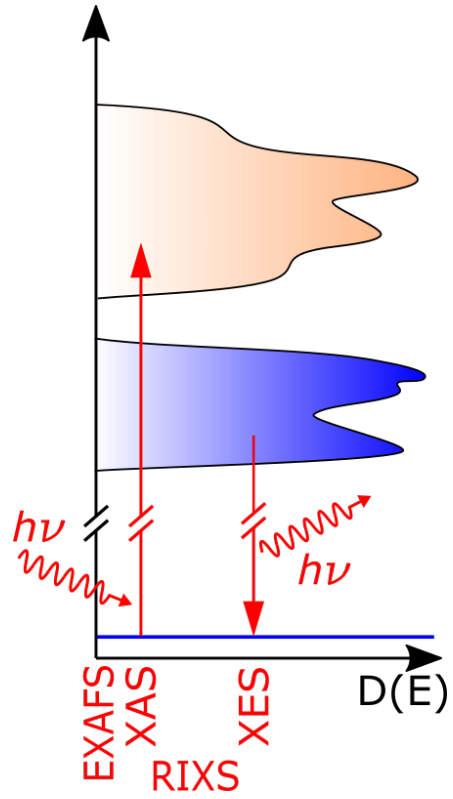




Electron spectroscopies

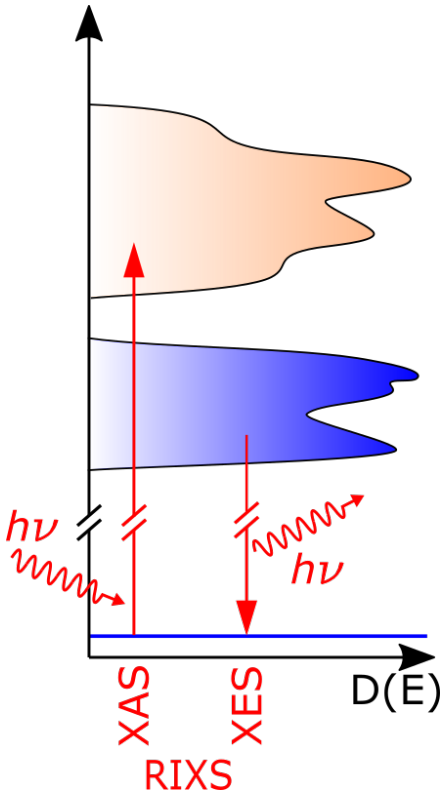


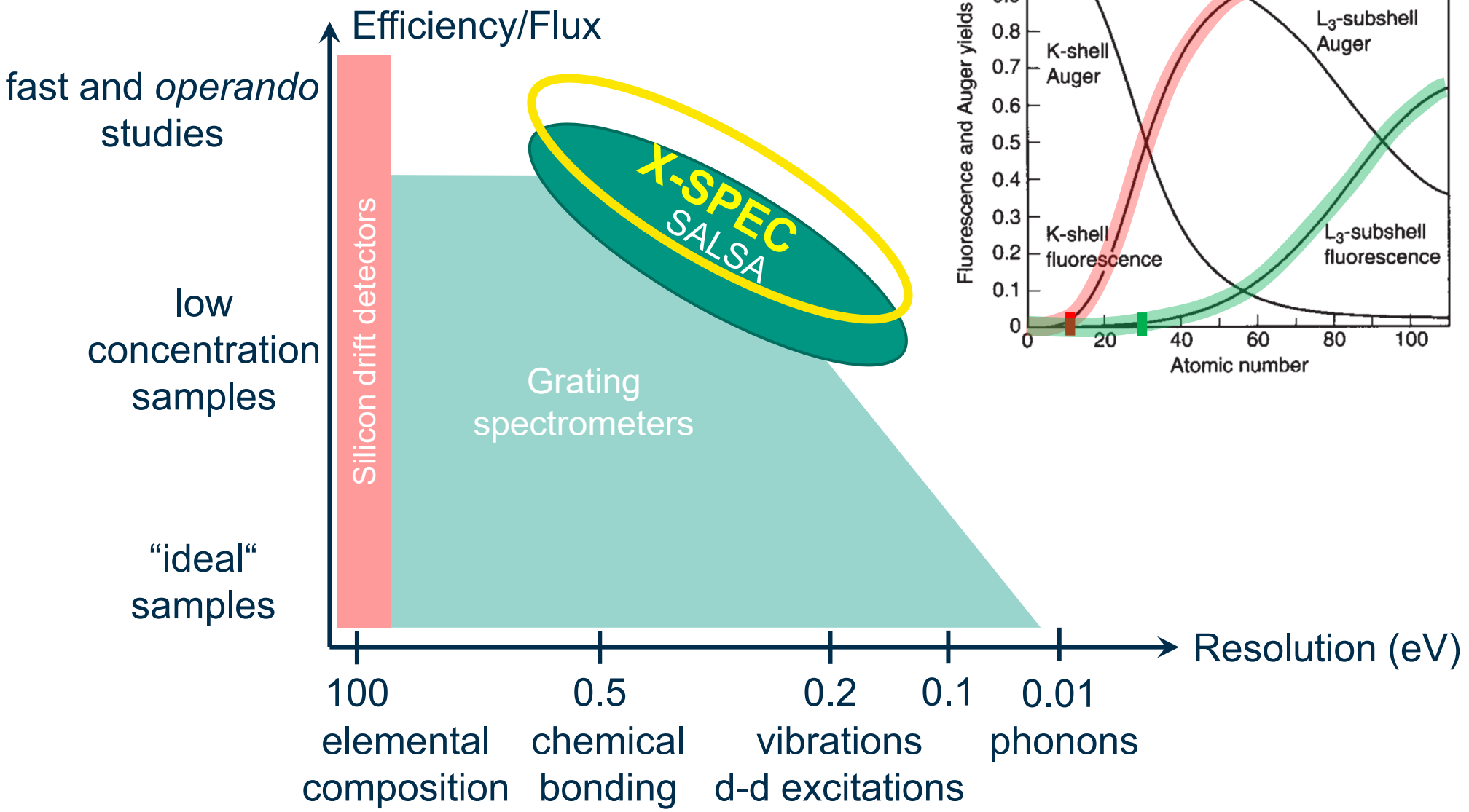
X-ray spectroscopies



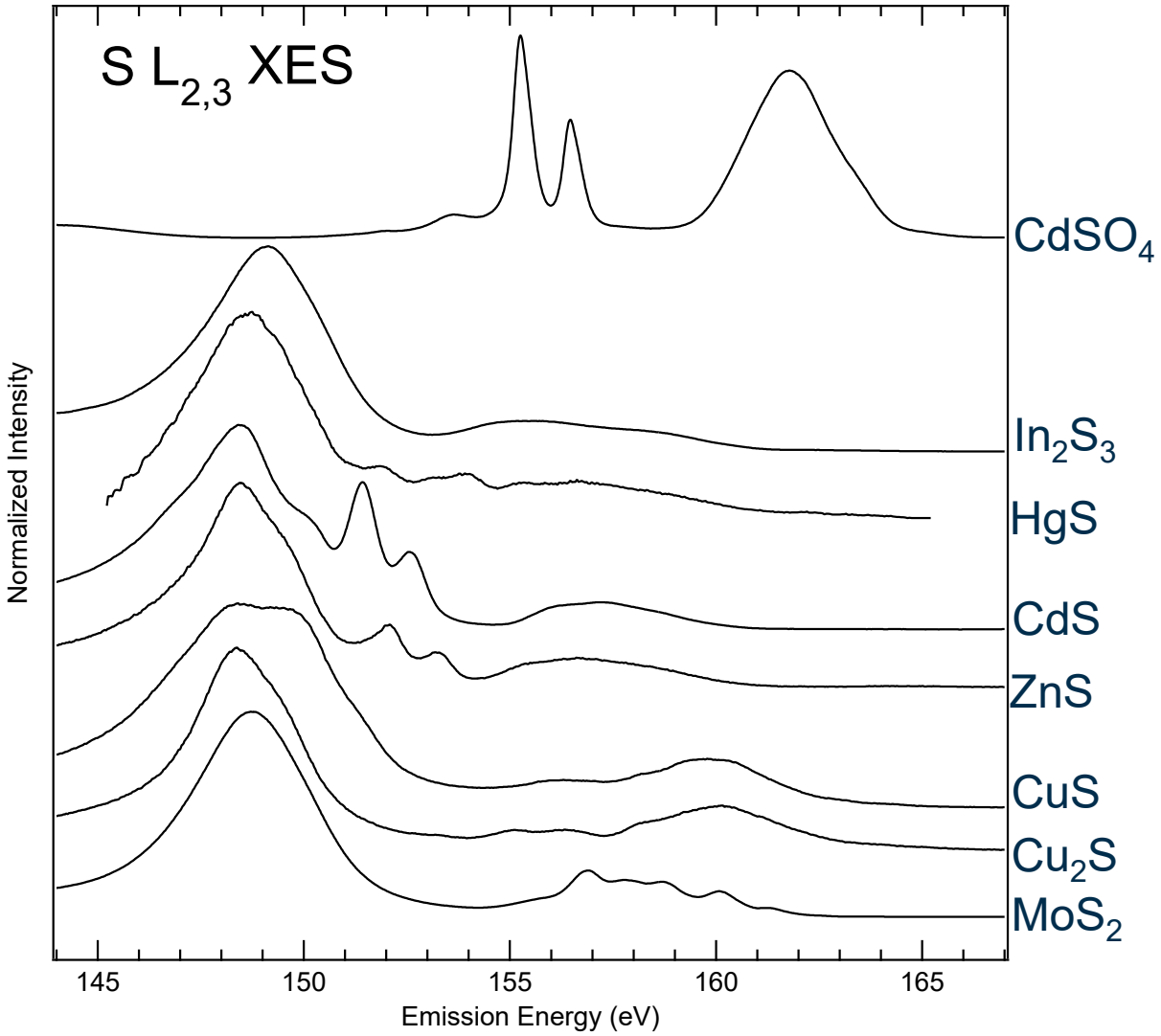
Weinhardt et al., *Advanced Materials* **31**, 1806660 (2019)

What can we learn with x-ray spectroscopy?

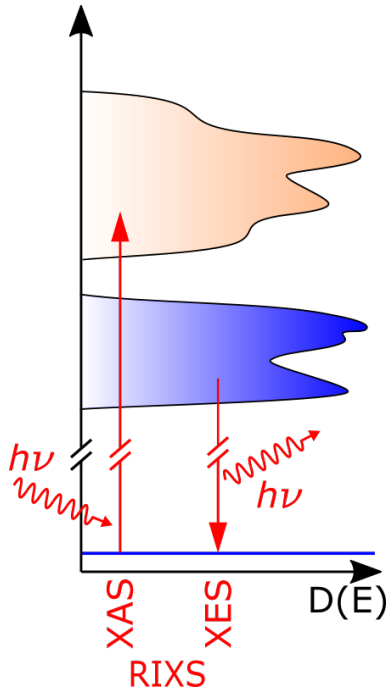




Soft x-ray emission spectroscopy (XES) is sensitive to the local chemical environment – example: sulfur



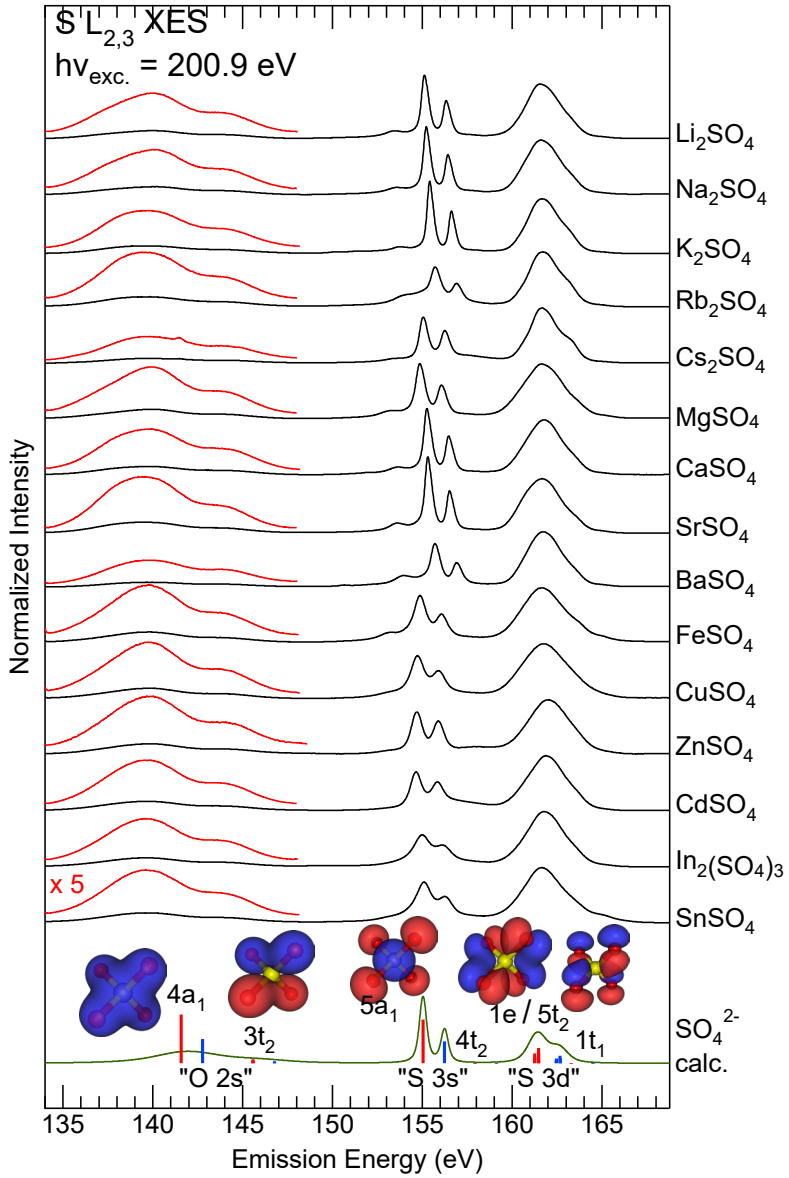
- XES probes the local partial density of states
  "Valence band structure from the viewpoint of a selected core level (atom)"
- Extremely sensitive to local bonding environment

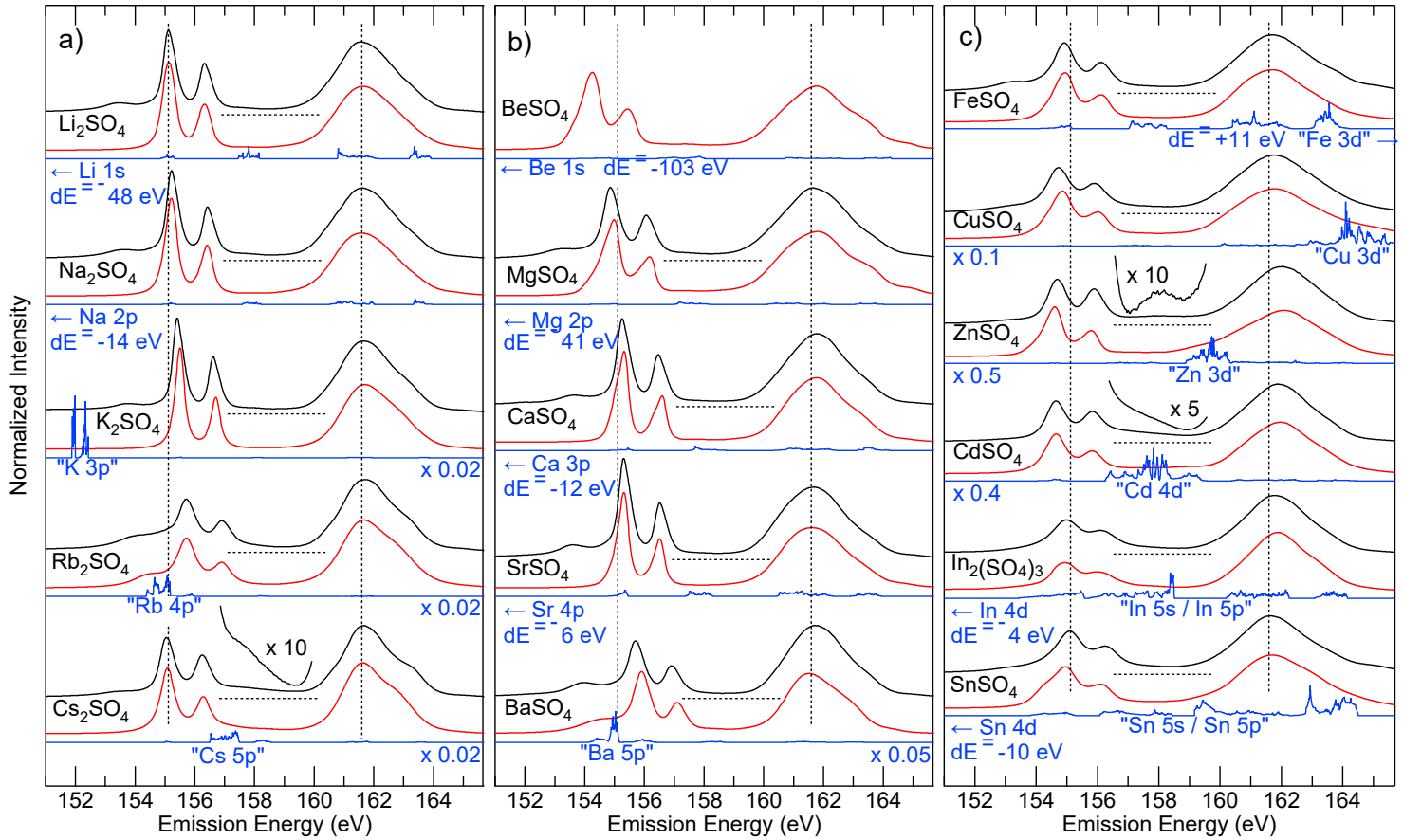


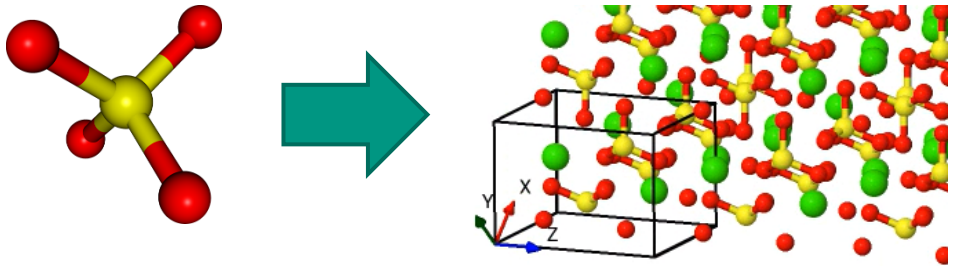
Phys. Stat. Sol. **187**, 13 (2001) (Cu_2S and CuS)
Appl. Phys. Lett. **83**, 2360 (2003) (HgS and ZnS)
J. Chem. Phys. **124**, 074705 (2006) (MoS_2)
Phys. Rev. B **75**, 165207 (2007), *Phys. Rev. B* **79**, 165305 (2009) (CdS)
Appl. Phys. Lett. **97**, 074101 (2010) (In_2S_3)
J. Phys. Chem. C **119**, 10412 (2015) (In_2S_3)
ACS Appl. Mater. Interf. **8**, 33256 (2016) (In_2S_3 and ZnS)
Anal. Chem. **23**, 8300 (2021) ($CdSO_4$)
Sol. RRL **7**, 2201091 (2023) (ZnS)
Prog. Photovolt.: Res. Appl. **32**, 904 (2024) (In_2S_3 and ZnS)
Phys. Chem. Chem. Phys. **26**, 26389 (2024) (ZnS, CdS, HgS, In_2S_3)
Adv. Mater. Interf. ***12***, 2401002 (2025) (CdS, $CdSO_4$)
…

Cation speciation in sulfates

Experiment — Calculation

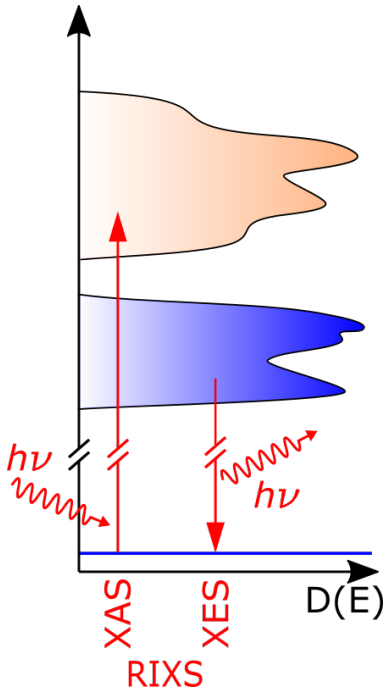


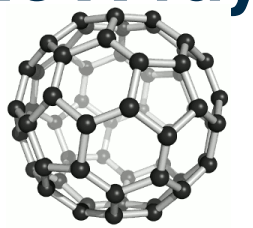




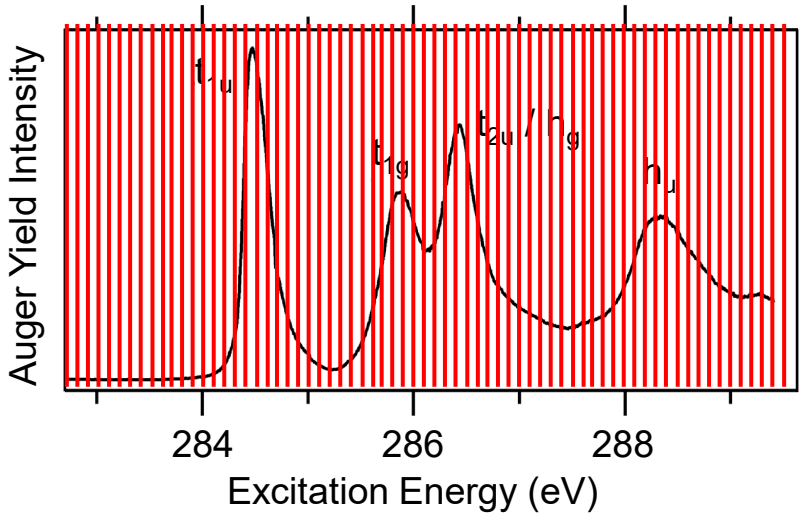
Weinhardt et al.,
Analytical Chemistry **93**, 8300 (2021)

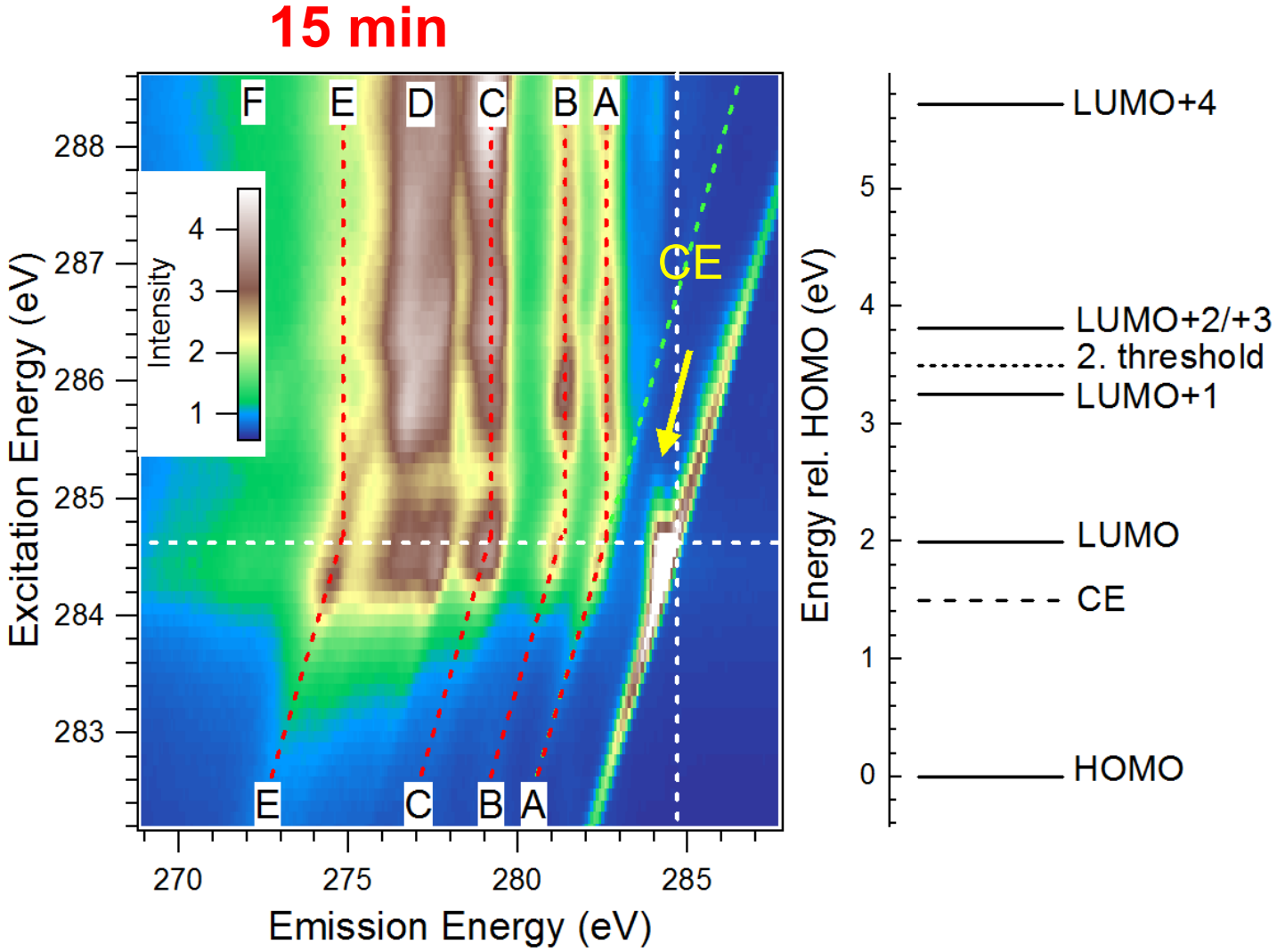
Resonant Inelastic X-ray Scattering (RIXS) map of C_{60}





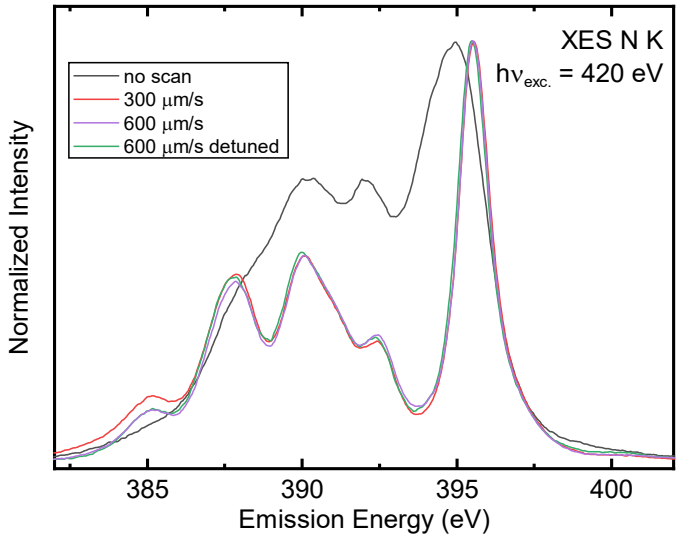
- Before SALSA: Measu
  spectra at a few selecte
  energies
- With SALSA: Closely s
  absorption scan with a
  spectrum at every exci

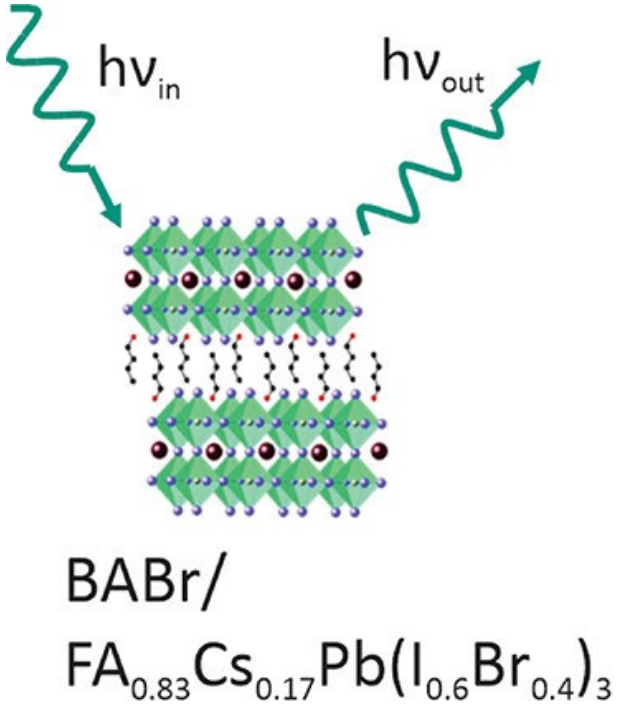


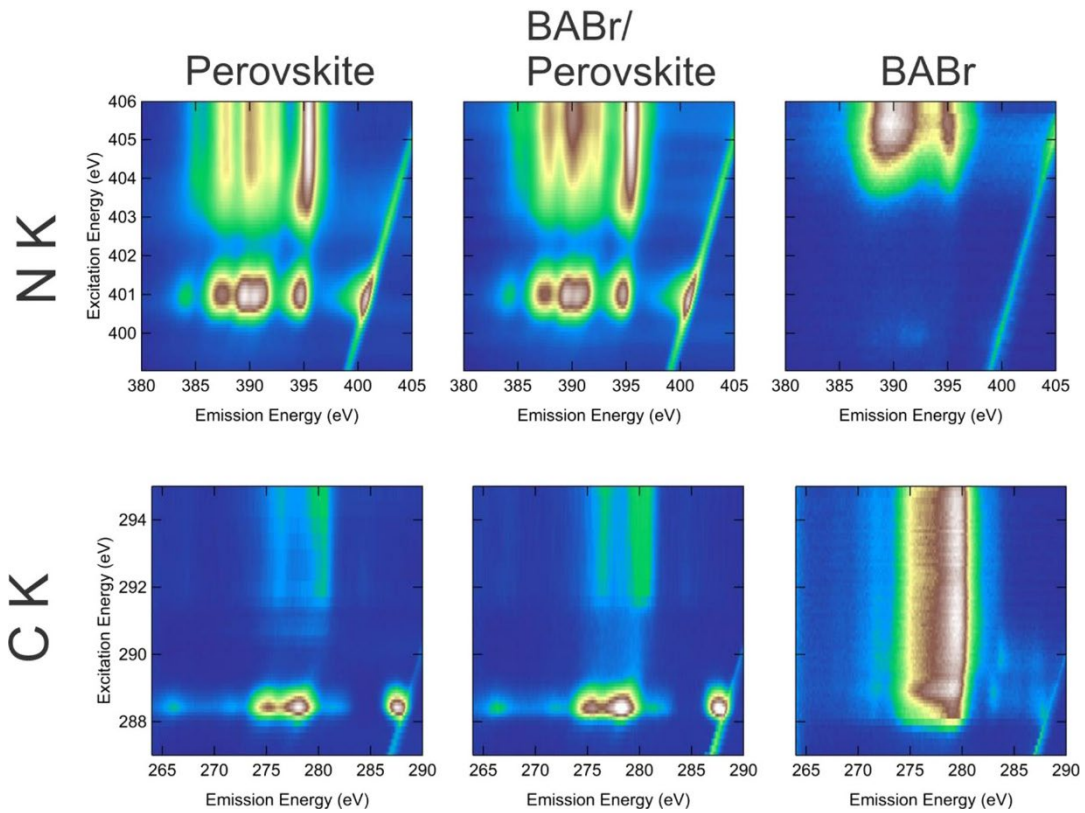


Weinhardt et al., JCP **135**, 104705 (2011)

2D/3D perovskite heterostructures for thin-film solar cells







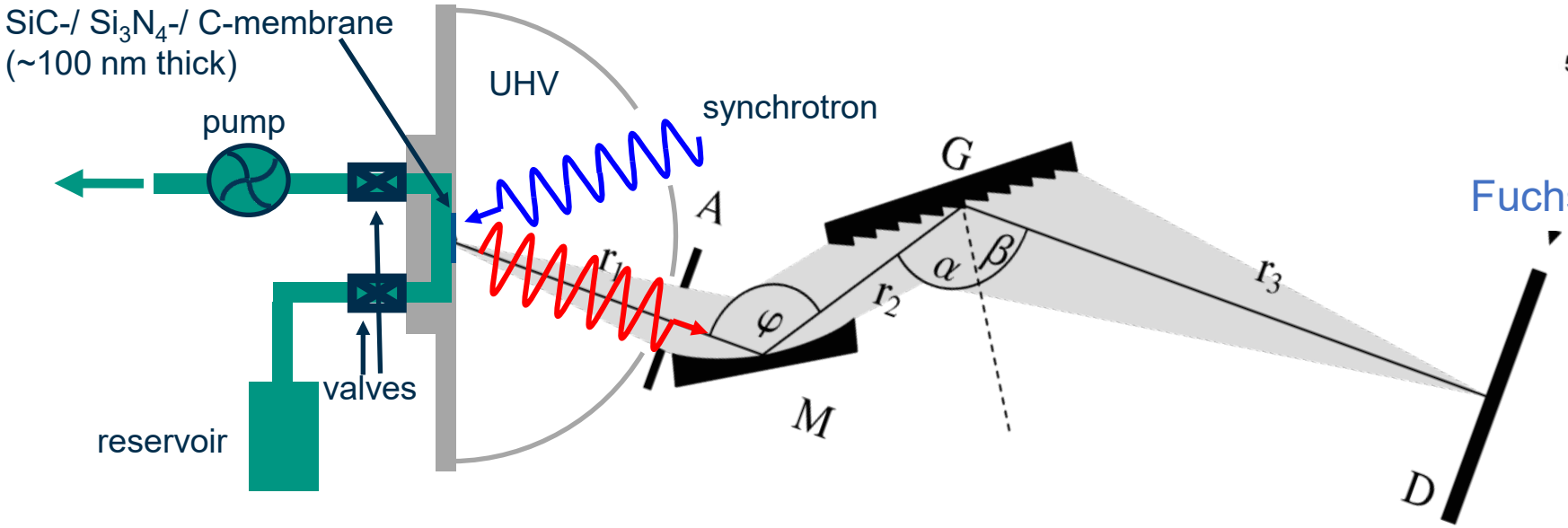
- Detailed chemical picture
- Organic materials sensitive to x-ray irradiation
- dose must be kept low
- here by continuous moving sample under beam
  → exposure of a given point: 50 ms

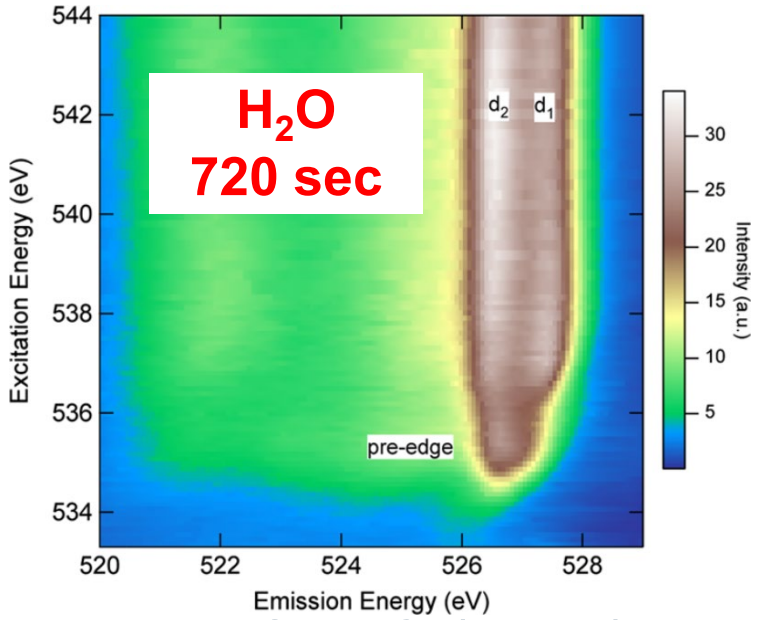
Hauschild et al.,
ACS Appl. Mater. Interfaces **13**, 53202 (2021)

In situ / operando experiments

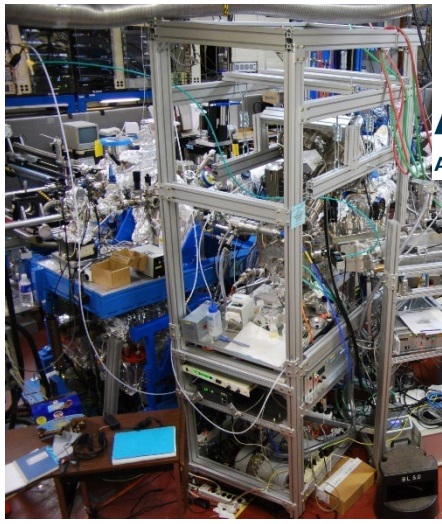
Gas or liquid continuously pumped through probing volume

Separated from UHV by a thin membrane (Si_3N_4, SiC, C)





Fuchs et al., *PRL* **100**, 249802 (2008)



ALS ADVANCED LIGHT SOURCE

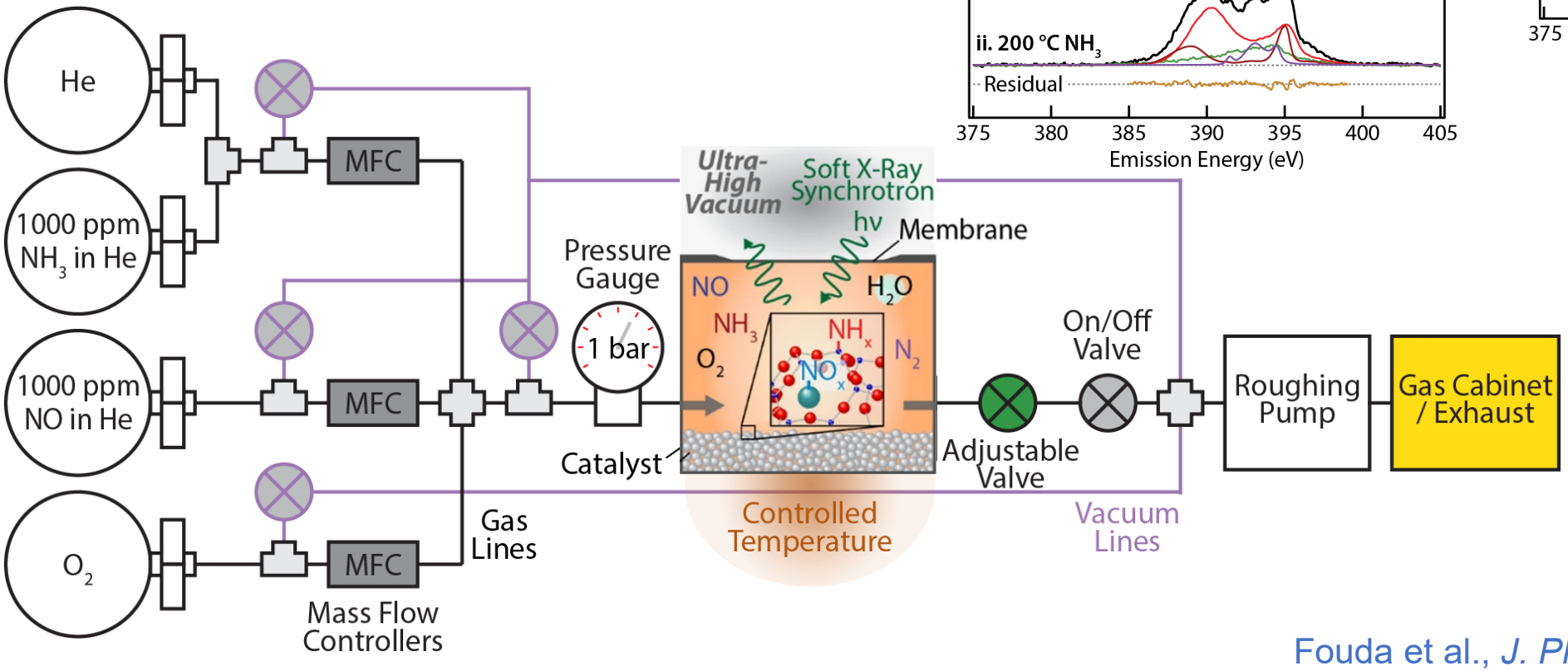
SALSA (2008)

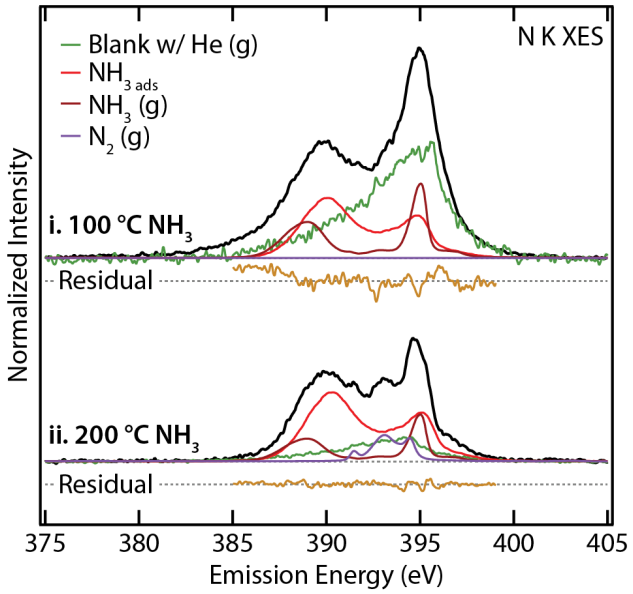
Fuchs et al., *Nucl. Instrum. Methods Phys. Res.* **585**, 172 (2008)
Blum et al., *Rev. Sci. Instr.* **80**, 123102 (2009)
Weinhardt et al., *J. Electr. Spectr. Rel. Phenom.* **188**, 111 (2013)
Benkert et al., *Rev. Sci. Instr.* **85**, 015119 (2014)
Weinhardt et al., J. Synchrotron Rad. **28**, 609 (2021)

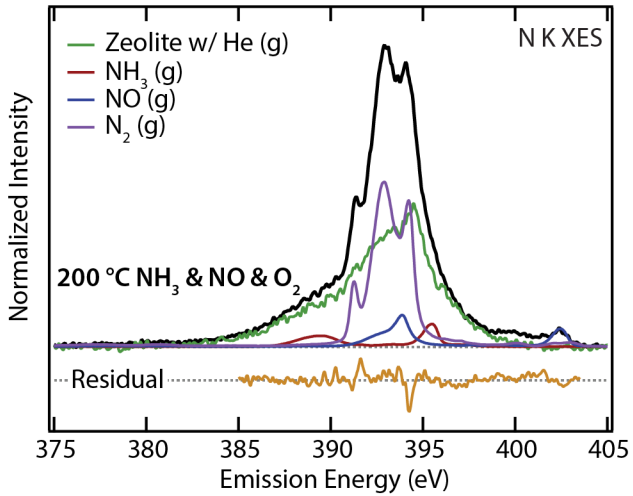
Operando study of catalytical processes

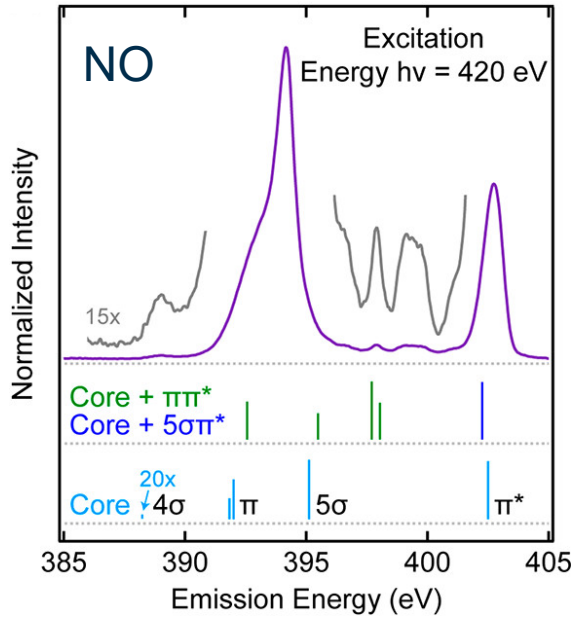
- Direct study of reactant, intermediate, and product species

Seitz et al., *J. Phys. Chem. C* ***126***, *20998 (2022)*





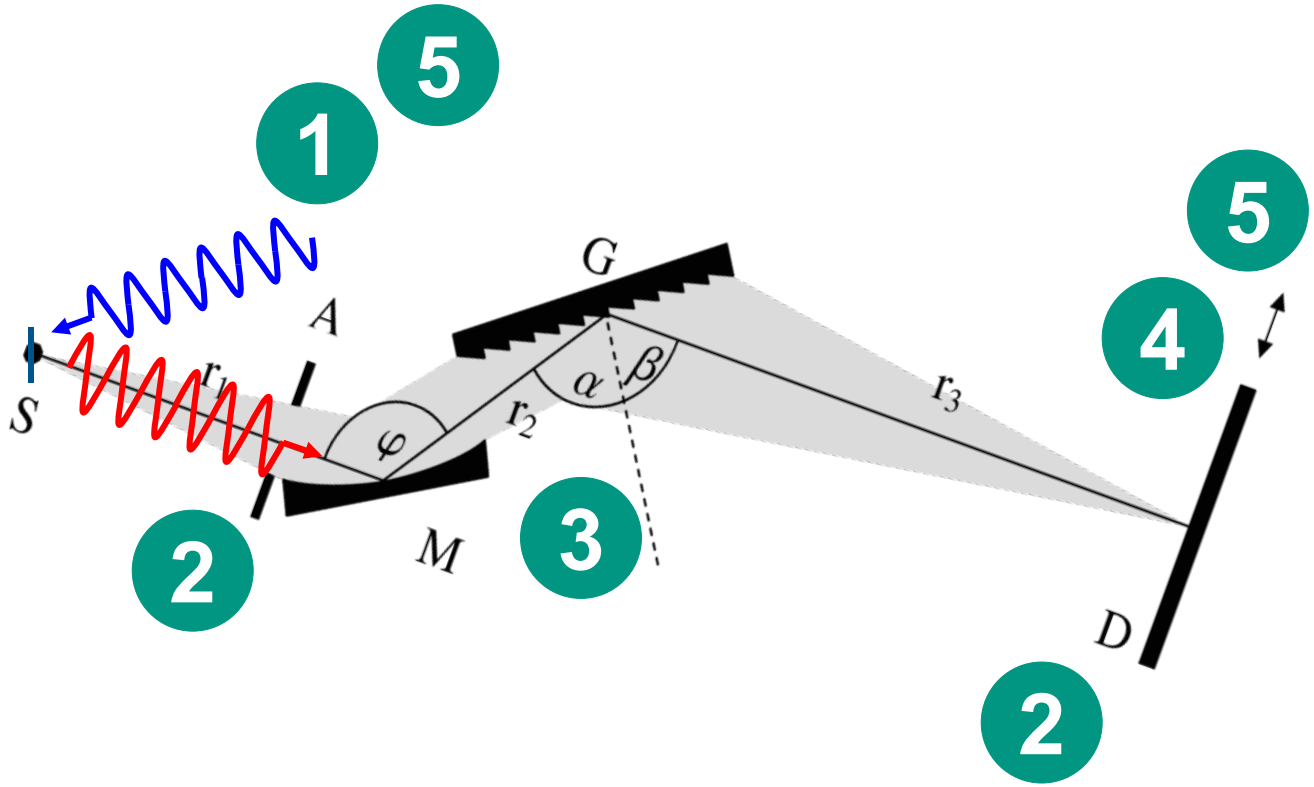




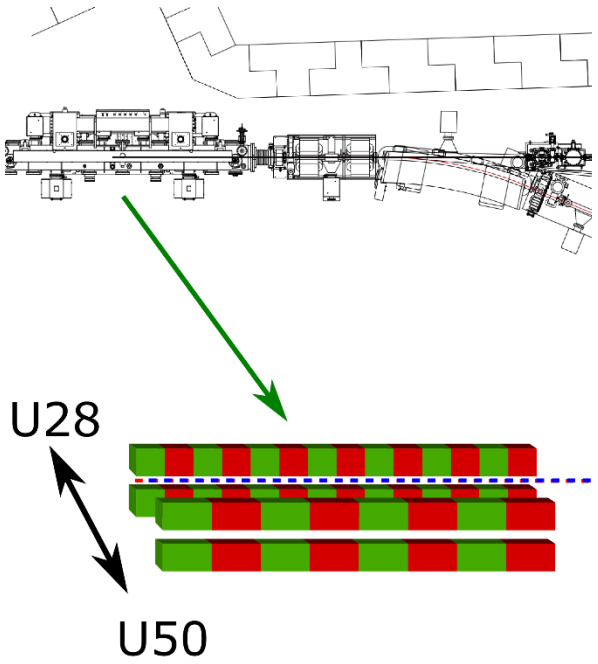
Fouda et al., *J. Phys. Chem. Lett.* **11**,7476 (2020)

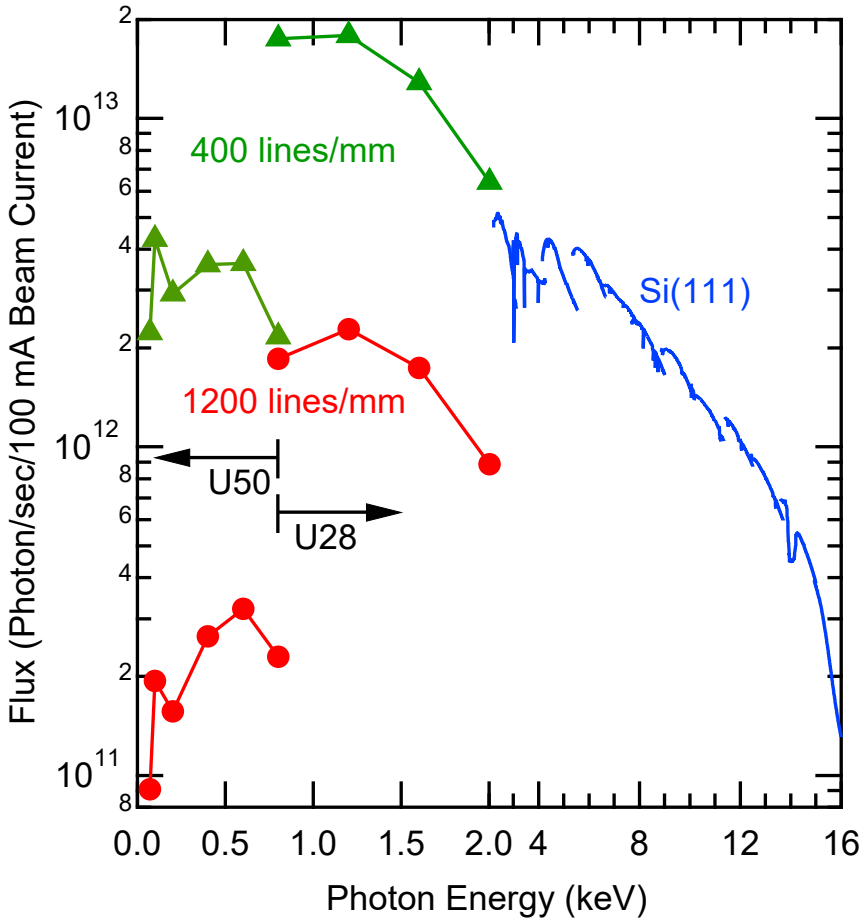
High efficiency soft x-ray spectrometer

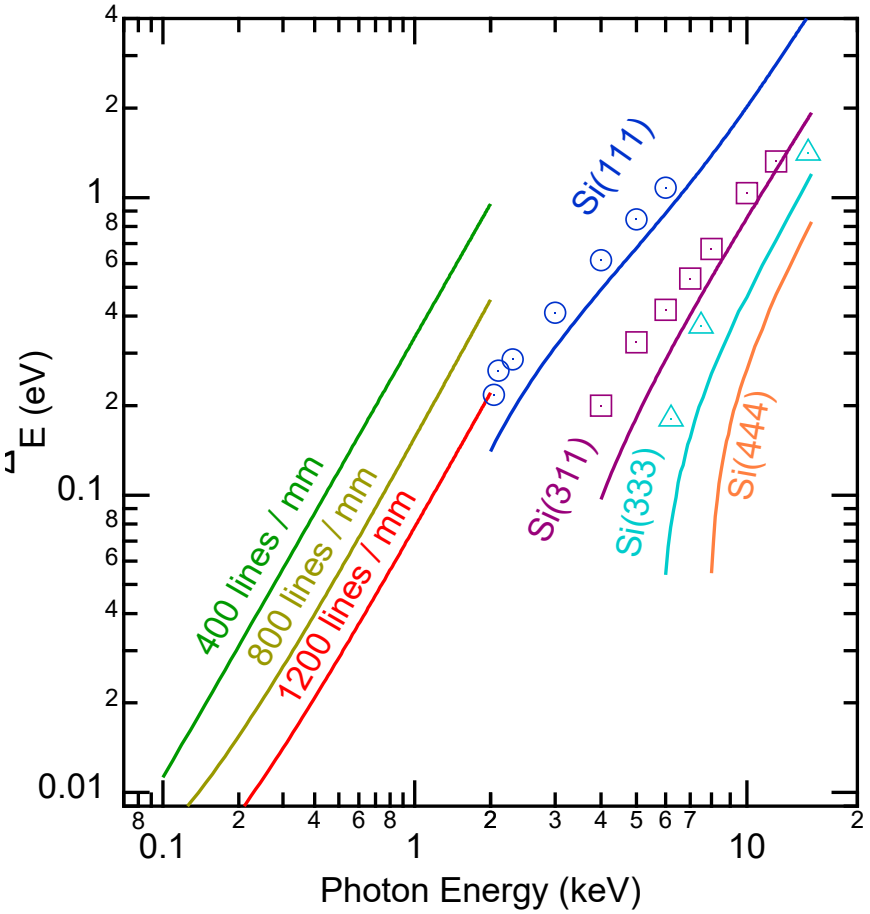
1. Beamline flux
2. Solid angle
3. Efficiency of optics
4. Detector efficiency
5. Dead times



X-SPEC @ KIT Light Source



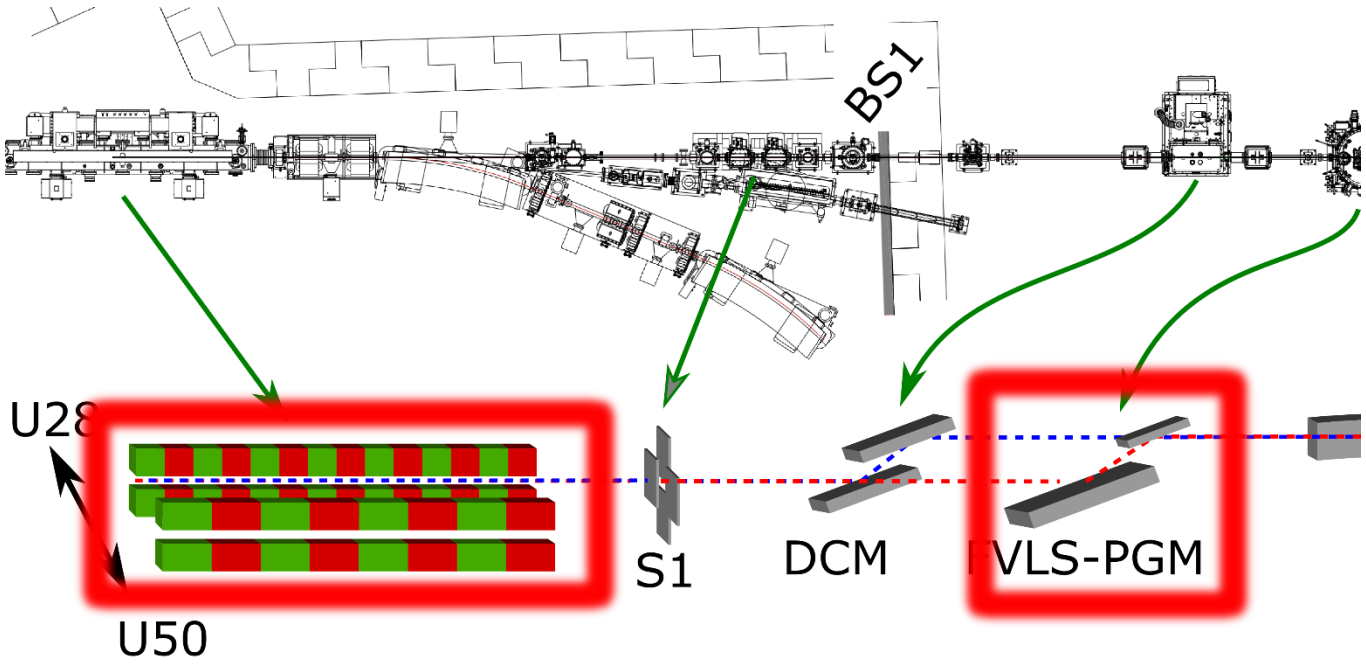


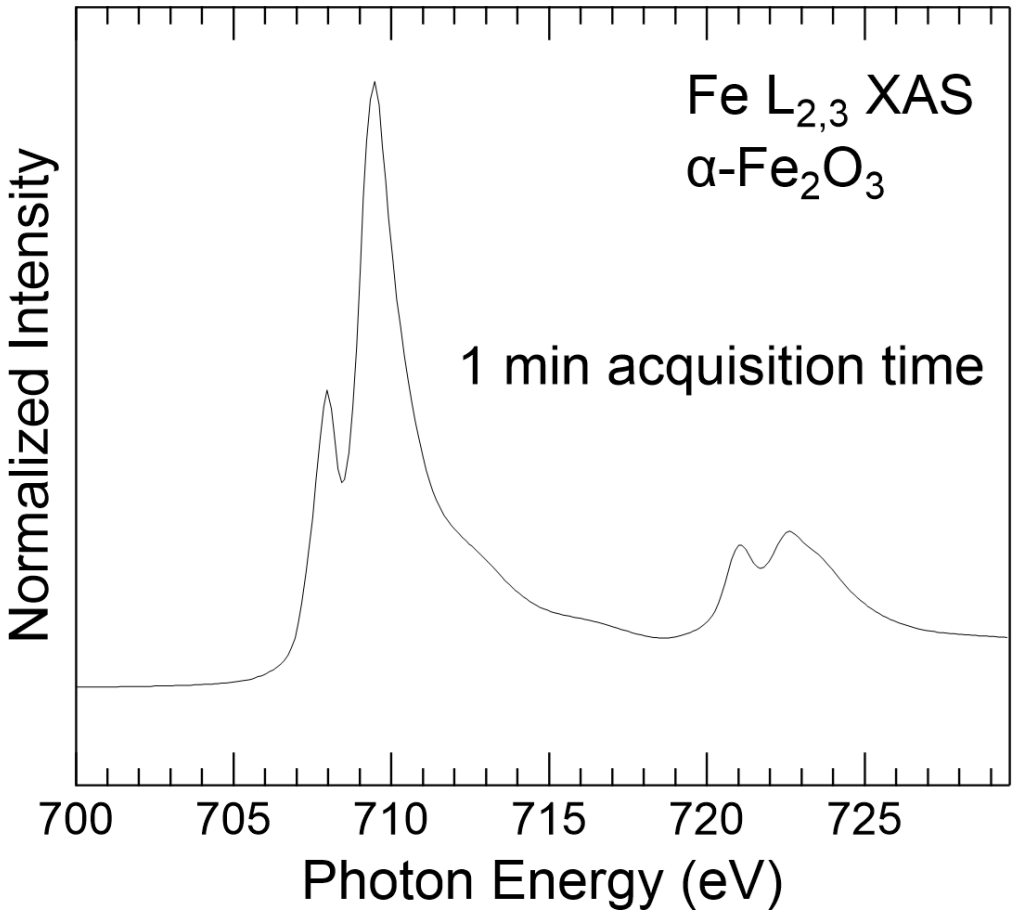


UHV and *in situ* / *operando* environments
XES, XAS, RIXS, EXAFS, PES, HAXPES

Weinhardt et al., J. Synchrotron Rad. **28**, 609 (2021)

X-SPEC @ KIT Light Source

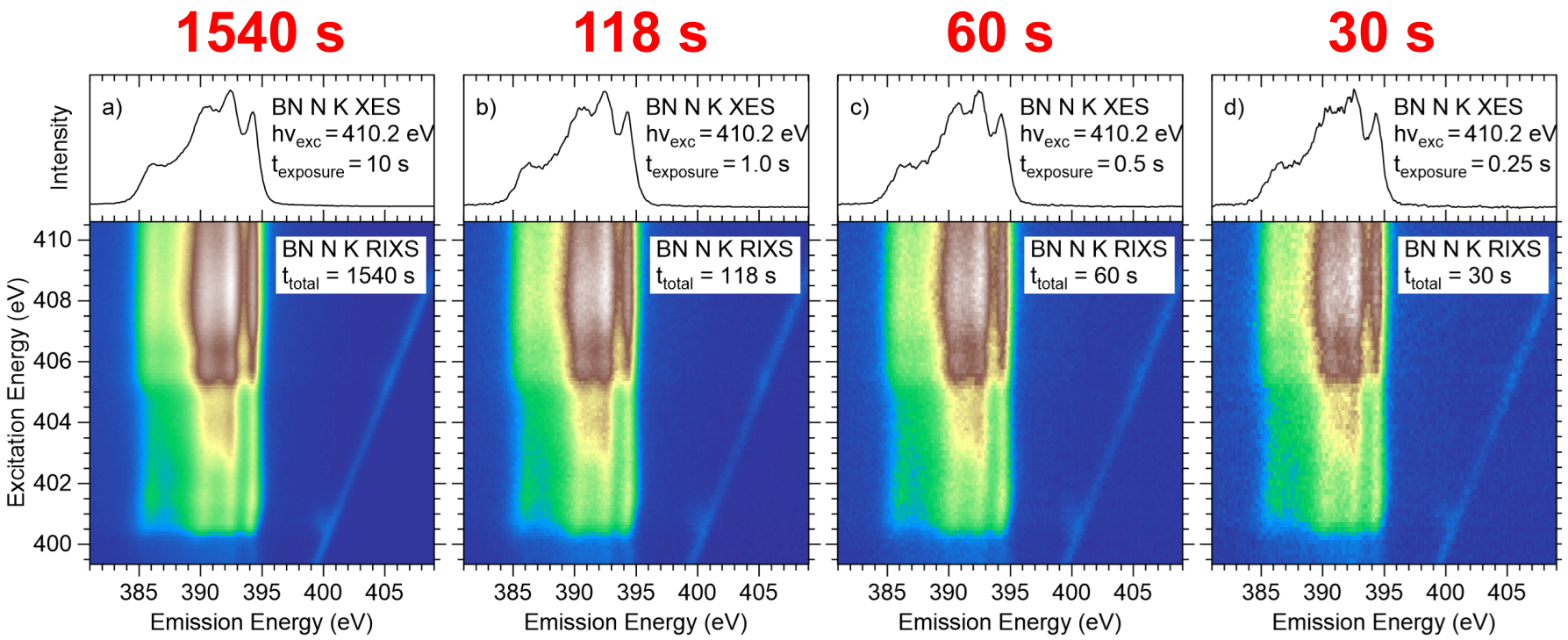


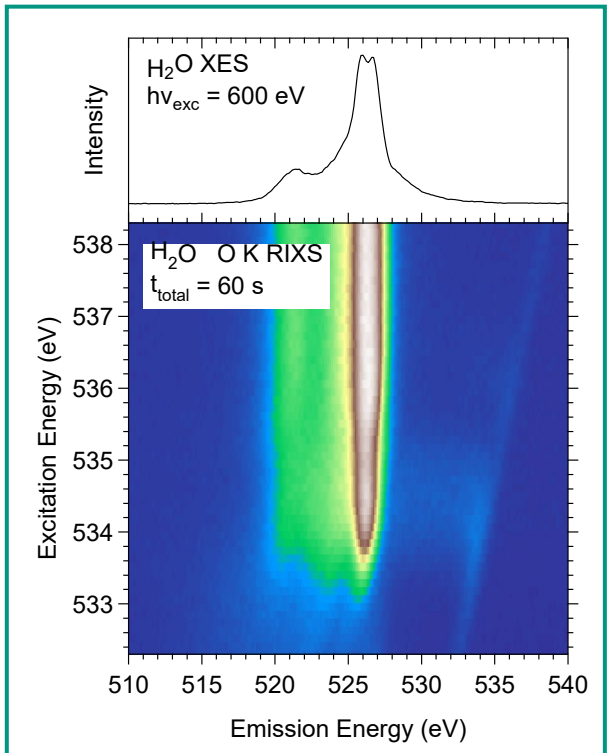


- Coordinated continuous movement of monochromator and undulator
- Fast CMOS sensor

Weinhardt et al., J. Synchrotron Rad. **28**, 609 (2021)

Rapid RIXS maps (rRIXS) at X-SPEC (KIT Light Source)

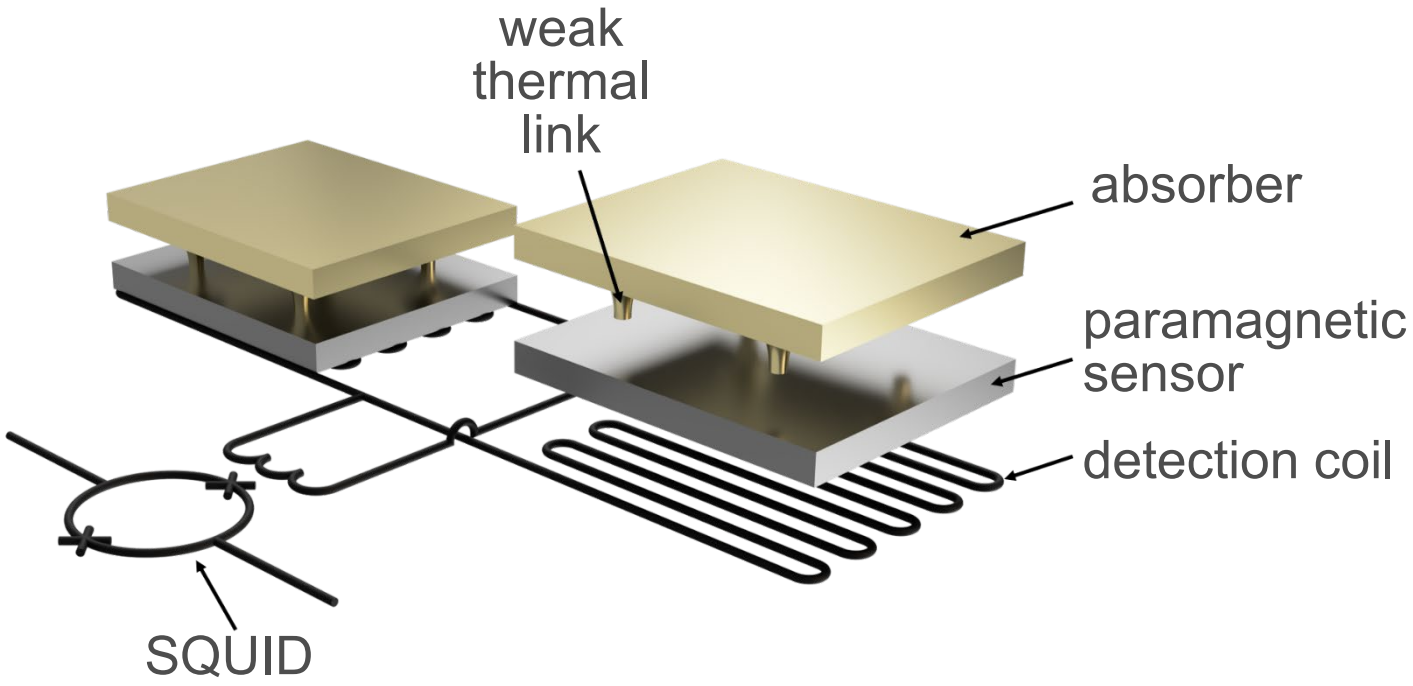


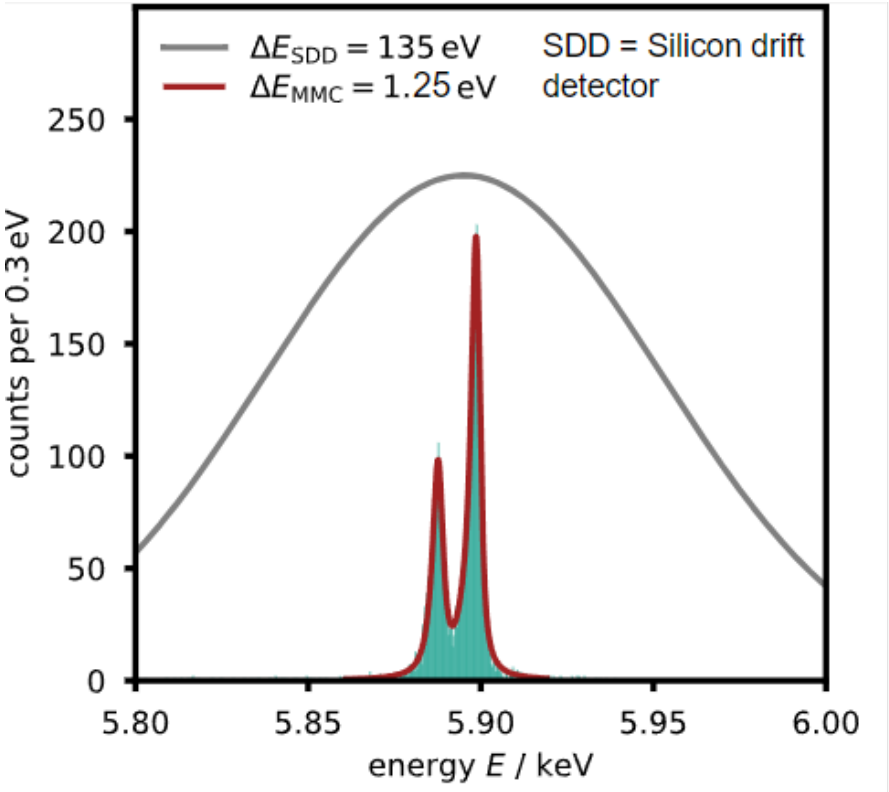


vs. 720 s in
Fuchs et al., *PRL* **100**, 249802 (2008)

Weinhardt et al. J. Synchrotron Rad. **31**, 1481 (2024)

Quantum x-ray sensors

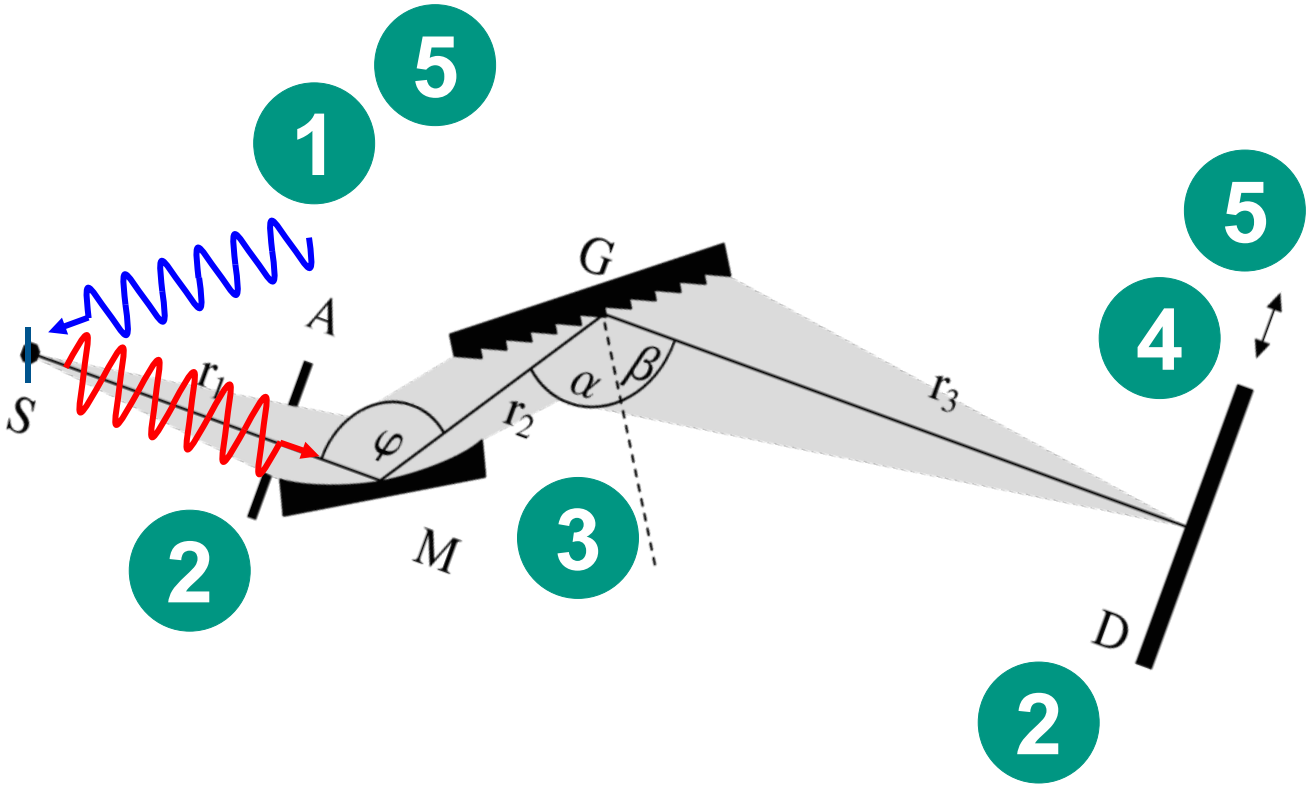




Krantz et al., Appl. Phys. Lett. 124, 032601 (2024)

High efficiency soft x-ray spectrometer

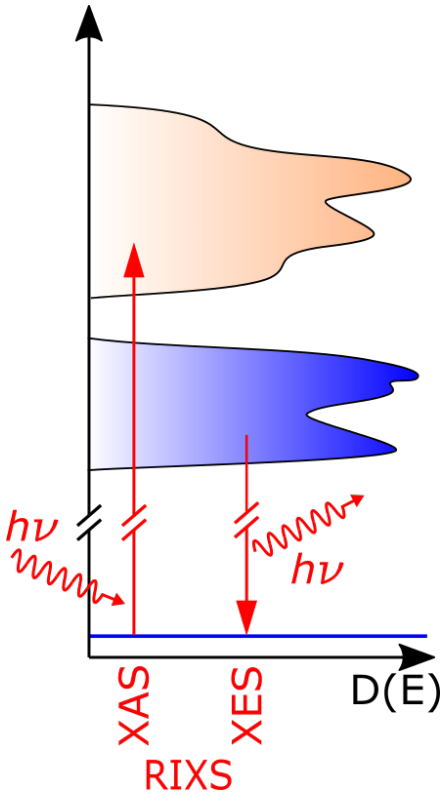
1. Beamline flux
2. Solid angle
3. Efficiency of optics
4. Detector efficiency
5. Dead times

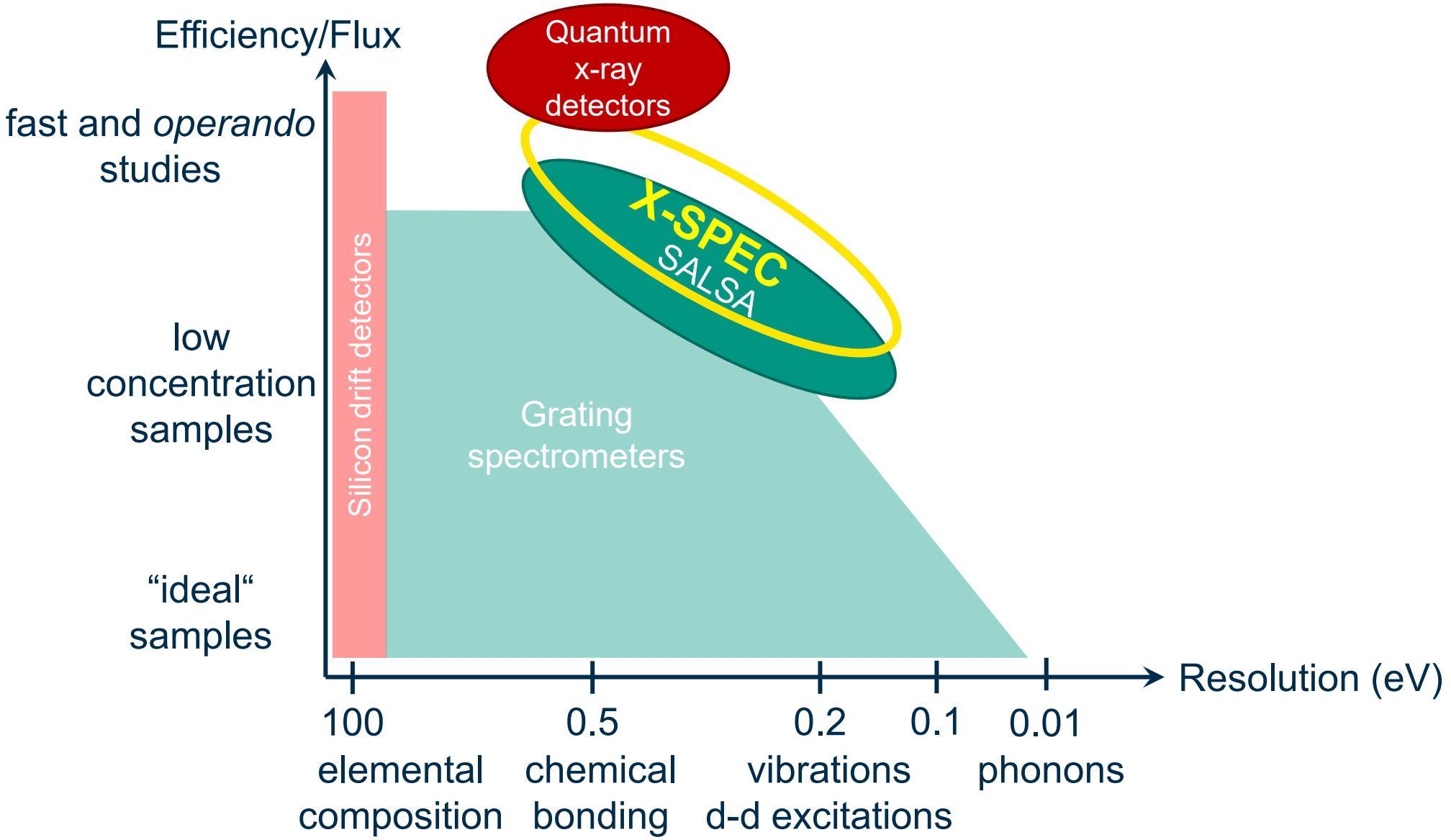


High efficiency soft x-ray spectrometer

	Grating spectrometer (X-SPEC @ KIT Light Source)	Quantum sensor
1 Beamline flux		
2 Solid angle	0.0017 – 0.0045 % of ½ sphere	Much larger!
3 Efficiency of optics	2 – 25 %	**??**
4 Detector efficiency	> 90 %	> 90 %
5 Dead times		

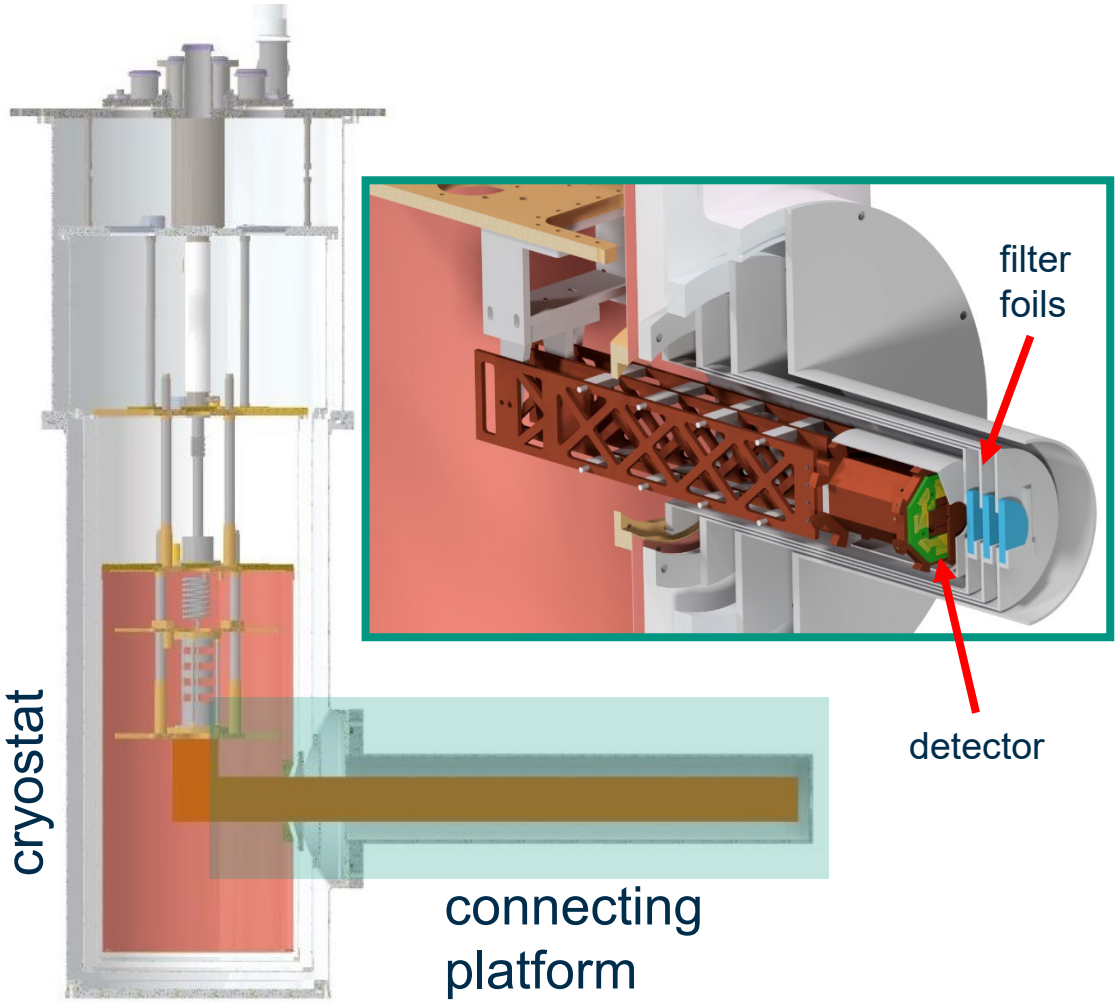
Is this the best we can do?





Quantum x-ray sensors

Poster Mary Blankenship



Work items

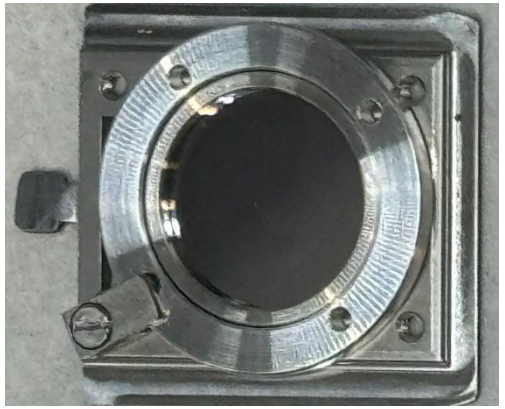
- Develop suitable filters to block IR radiation but transmit (soft) x-rays
- We need many pixels for high count rates!
- Filter “unwanted” spectral components?

Ultra-thin Filter Foil Candidates

Commercially available ultra-thin foils for filtering IR radiation:

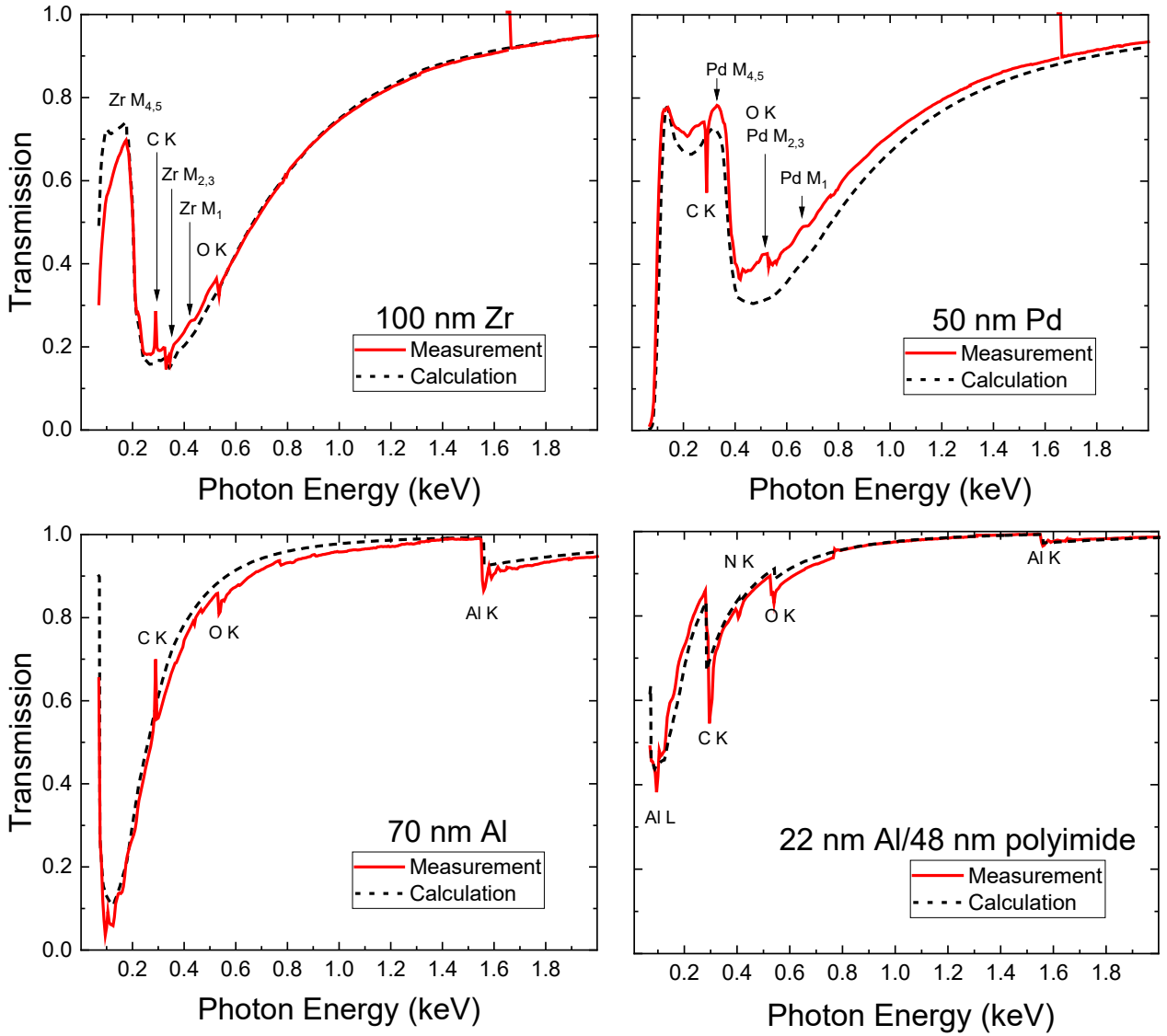
- 50 nm Pd, 70 nm Al, 100 nm Zr
- 21.5 nm Al/47.6 nm polyimide

→ Fragility of these foils makes measurements challenging!



unbroken foil example

K. Bitsch et al., Master Thesis (2024)



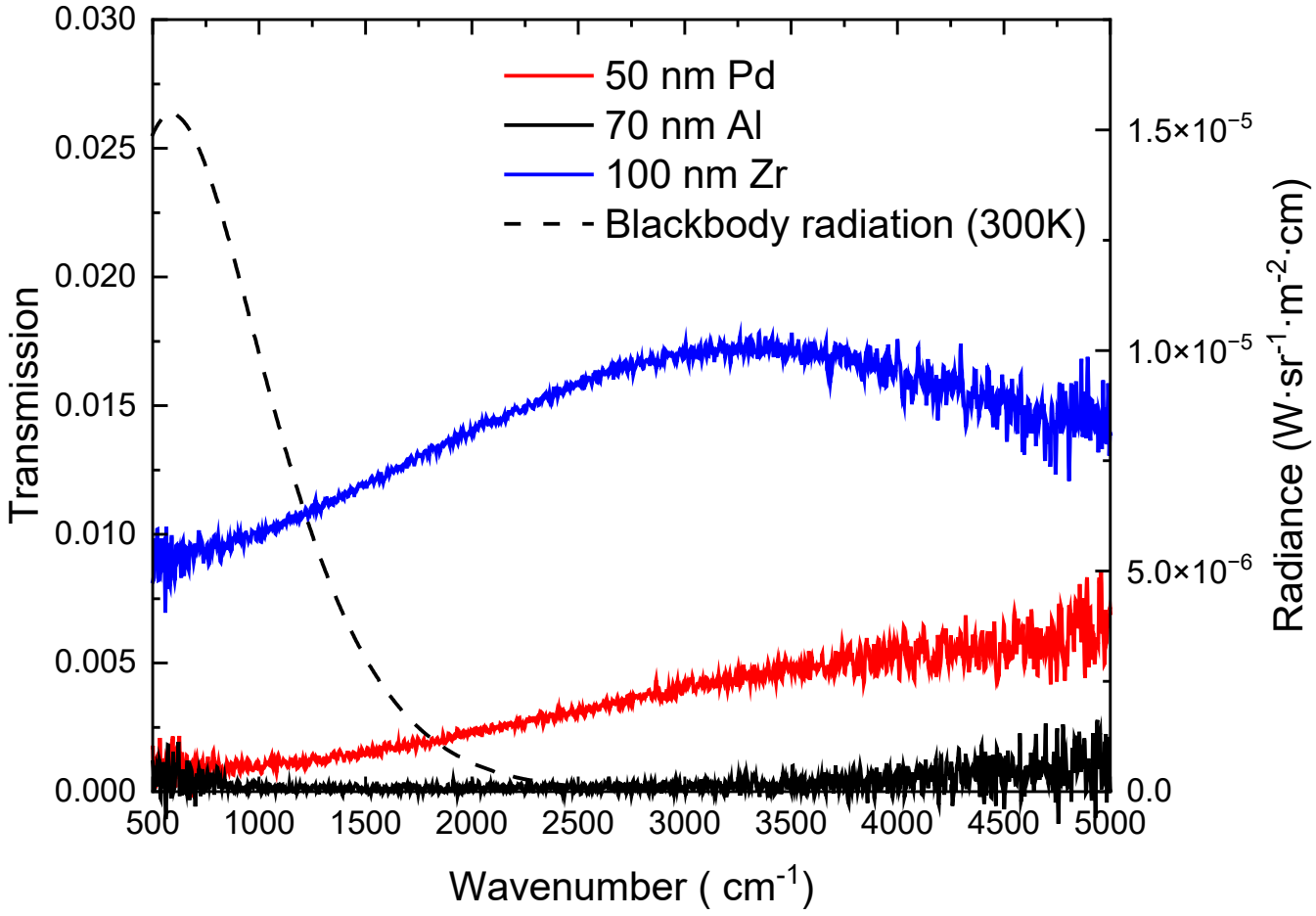
IR Radiation Transmission

Cryostat base temperatures reach as low as 7 mK, sensor array cooled to temperatures of 17 mK

M. Krantz et al., Appl. Phys. Lett. 124, 032601 (2024)

Detector resolution is noticeably reduced even at such low transmission

- Multiple filters are necessary to block enough of the IR radiation



K. Bitsch et al., Master Thesis (2024)

Summary

- Soft X-ray Spectroscopies are very powerful techniques for electronic and chemical structure studies
- Material research needs high-transmission x-ray spectrometers
- Quantum detectors promise the next big leap forward