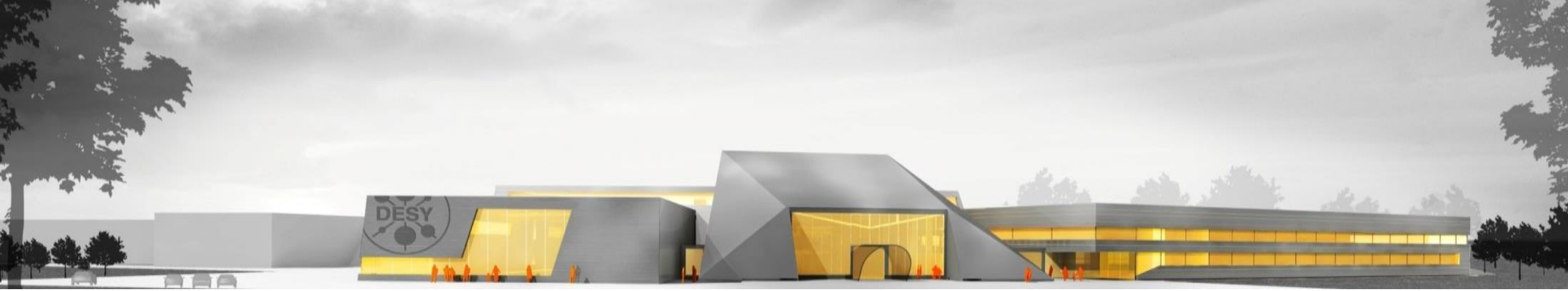
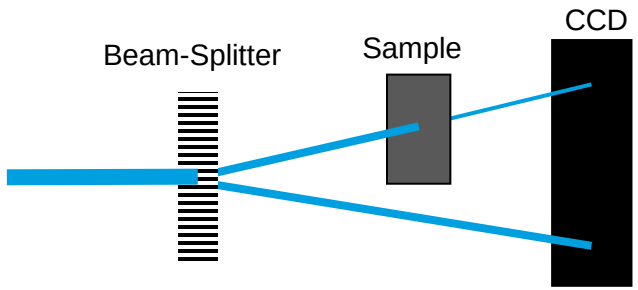


Intensity normalization for X-ray absorption spectroscopy at SASE-FELs using transmission-grating beam-splitters

Realizations of the technique in the soft X-ray regime



Gratings from

PAUL SCHERRER INSTITUT
PSI

Christian David
Benedikt Rösner
Florian Döring

Robin Engel
FLASH

Group of Martin Beye

HELMHOLTZ RESEARCH FOR GRAND CHALLENGES

Universität Hamburg
DER FORSCHUNG | DER LEHRE | DER BILDUNG

PIER
Partnership of Universität Hamburg and DESY

HELMHOLTZ
Young Investigators

DESY.

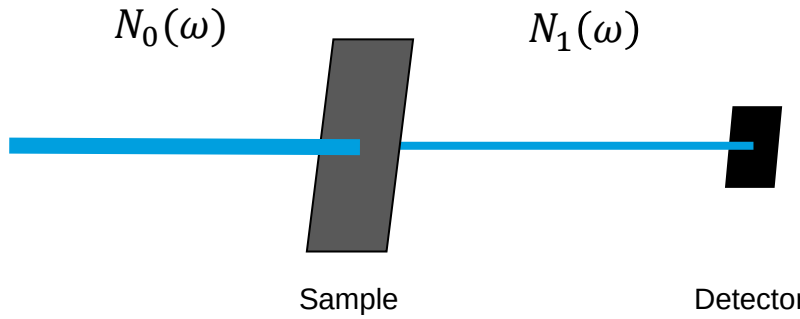
X-Ray Absorption Spectroscopy

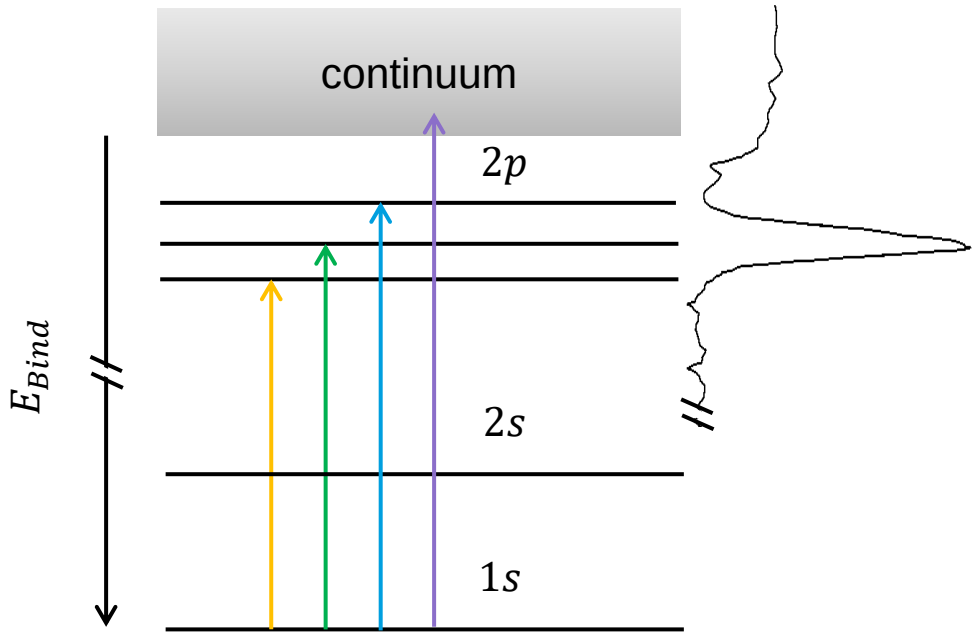
Simple, element-selective and informative!

- Probes **unoccupied local partial** density of electronic states
  - ➢ Crystal fields
  - ➢ Chemical bond length & type
  - ➢ Spin configuration / charge transfer

- VUV/soft X-ray
  - Element specific
  - Sharp core levels
  - Short absorption lengths

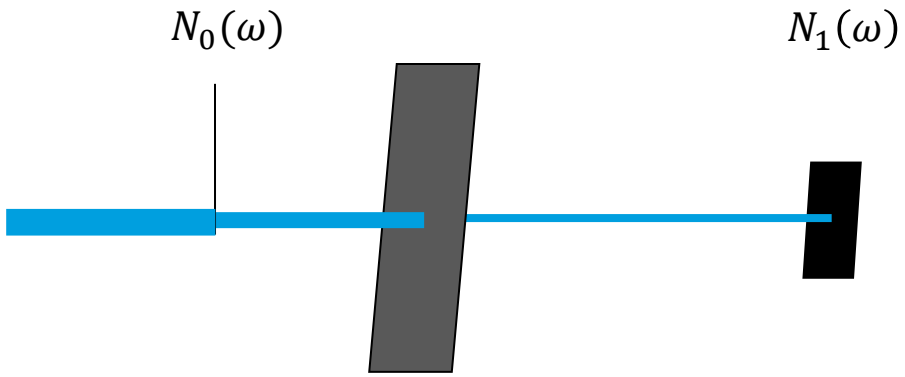
XAS is simple but informative

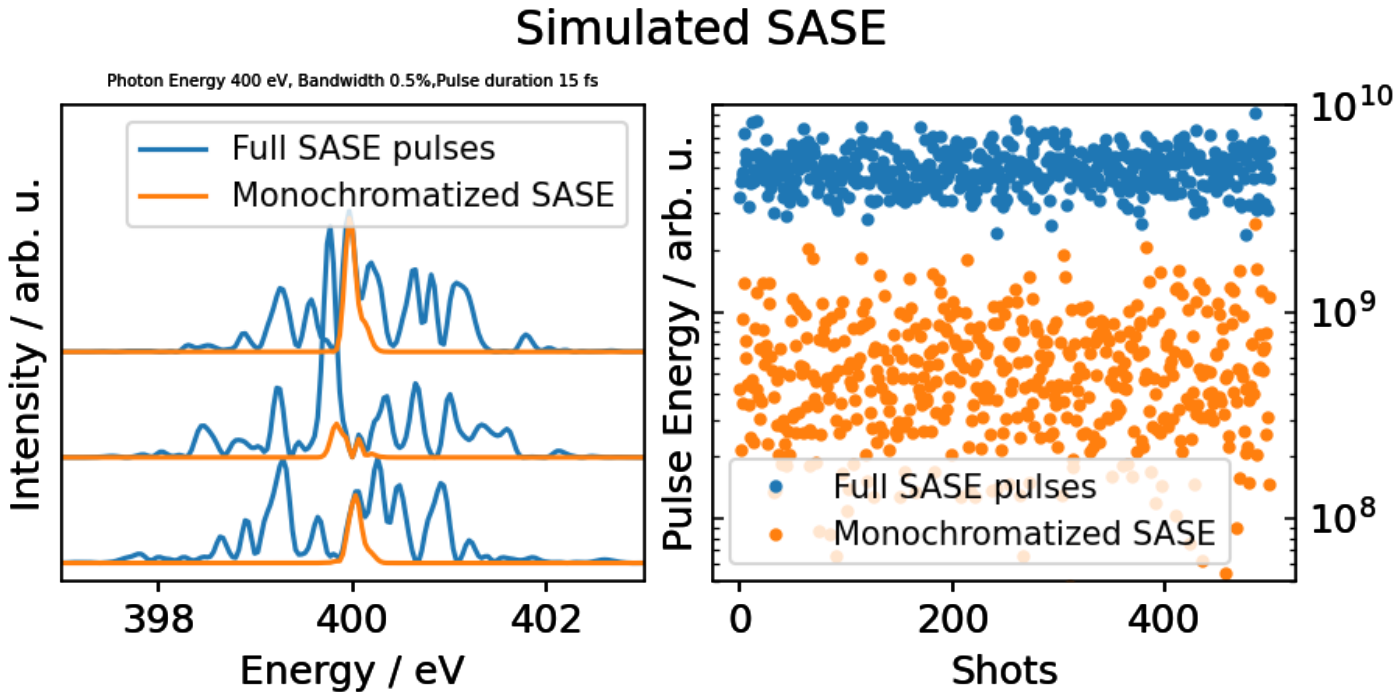




The Problem at SASE-FELs

Pulse intensity fluctuations





Simple if incident flux ("I_0") is constant
Otherwise → Normalization

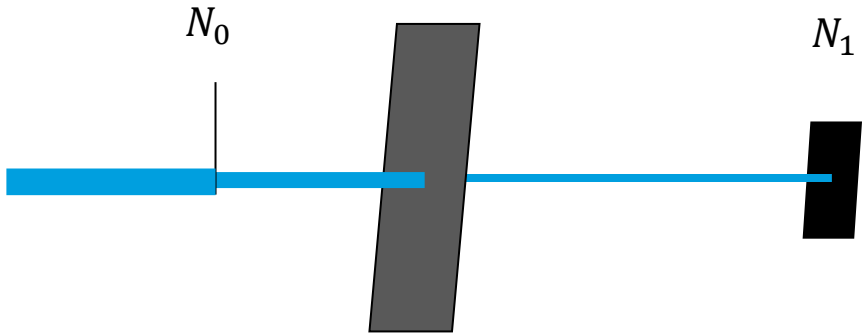
Intensity Normalization

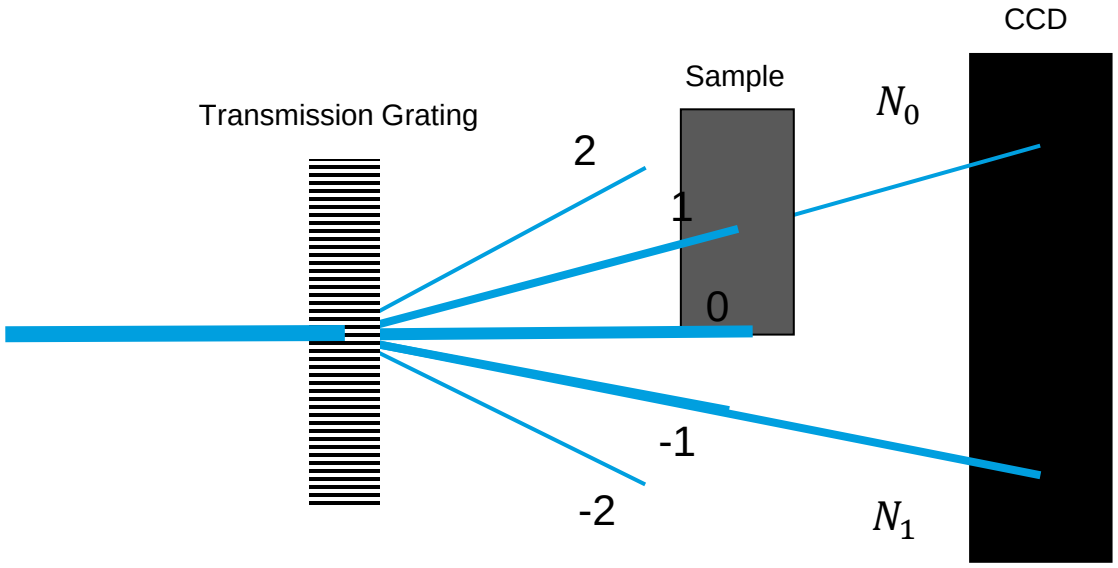
Making the best use of photons

- Measuring intensity means drawing a sample of the number of photons passing by
- Best Signal to Noise Ratio is the ***shot noise limit***:

$$SNR = \frac{N_{h\nu}}{\sqrt{N_{h\nu}}} = \sqrt{N_{h\nu}}$$

- Ideal case:
  - 50% of photons measure I_0
  - 50% of photons measure I_1



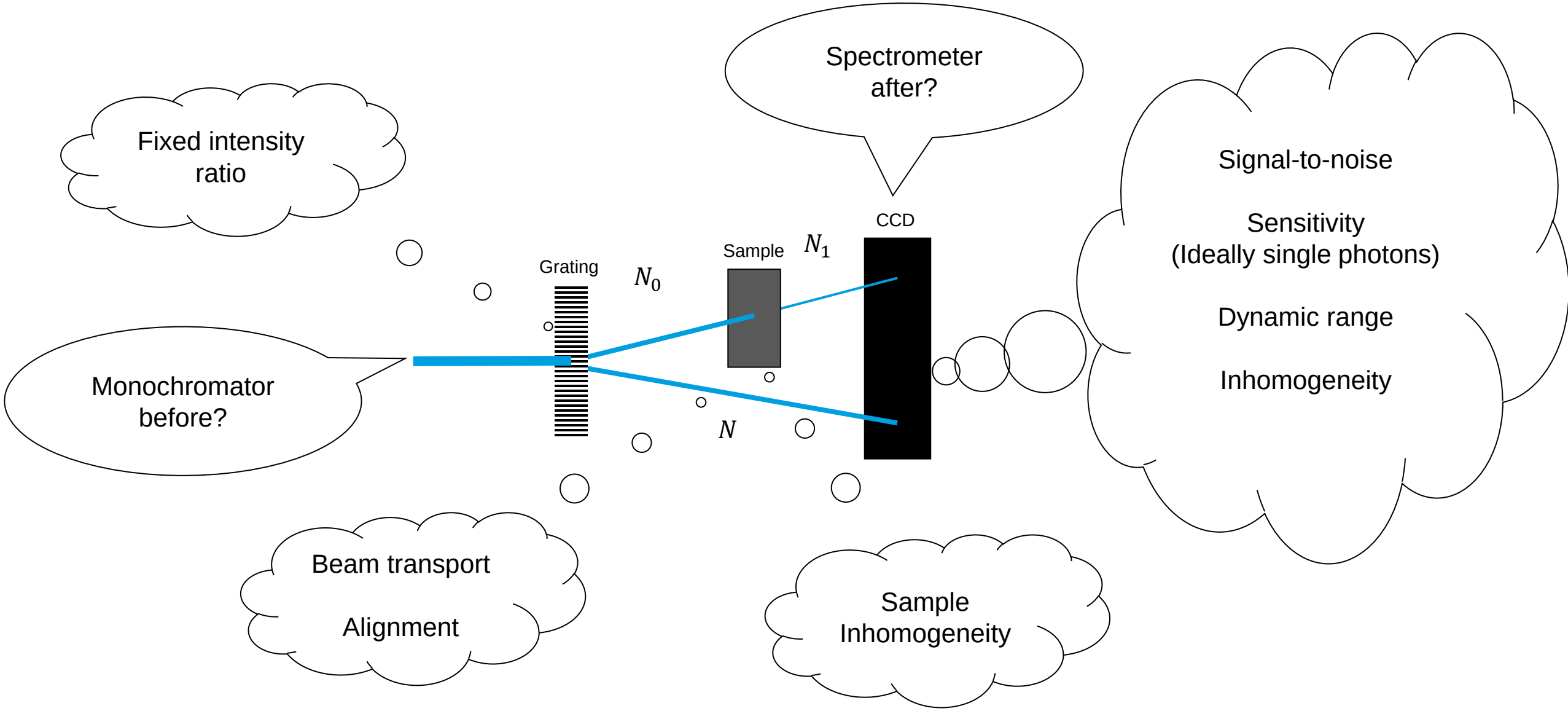


- Split the beam in two identical copies!

  Measured with the same Detector (CCD)

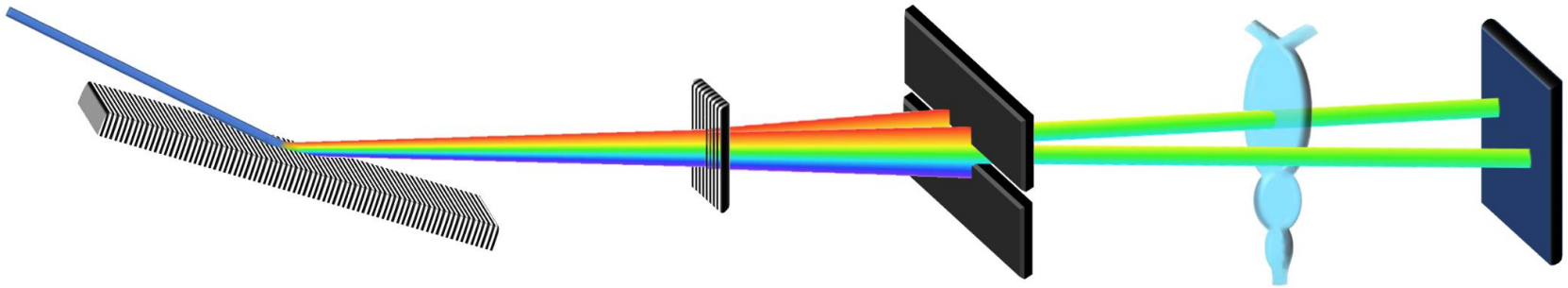
  "Split-Beam Normalization"

Challenges for a XAS-normalization setup



#1: Monochromatic separate beams FLASH I – PG2

Science case:
Explore the **electronic response** of **metallo-organic** molecules to **optical excitation**.



People Involved

Many thanks to:

Martin Beye
Piter Miedema

Siarhei Dziarzhytski
Günter Brenner

Marie Kristin Czwalinna

Maria Ekimova
Carlo Kleine
Jan Ludwig
Erik T. J. Nibbering

Nils Huse
Miguel Ochmann
Benjamin Grimm-Lebsanft
Michael Rübhausen
Melissa Teubner
Rory Ma

Christian David
Benedikt Rösner

Sonja Herres-Pawlis

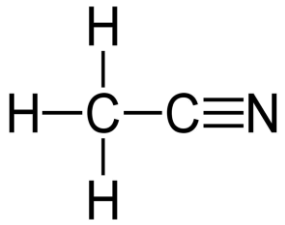
Tae Kyu Kim

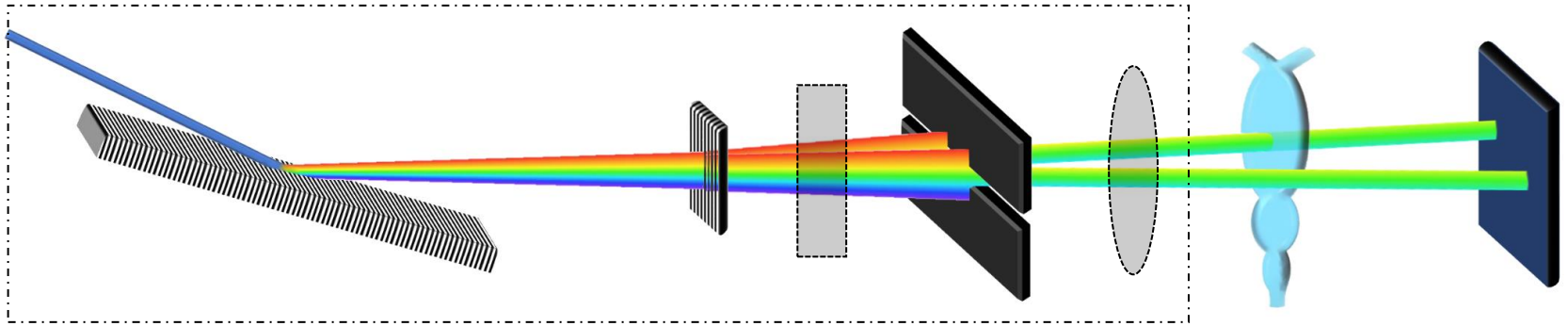
Setup

Monochromatic individually steered beams

- Thin-film liquid jet target
- Grating between monochromator grating and exit slit
- Beams steered individually by SDU-unit mirrors
- Both beams focused by beamline KB-optics

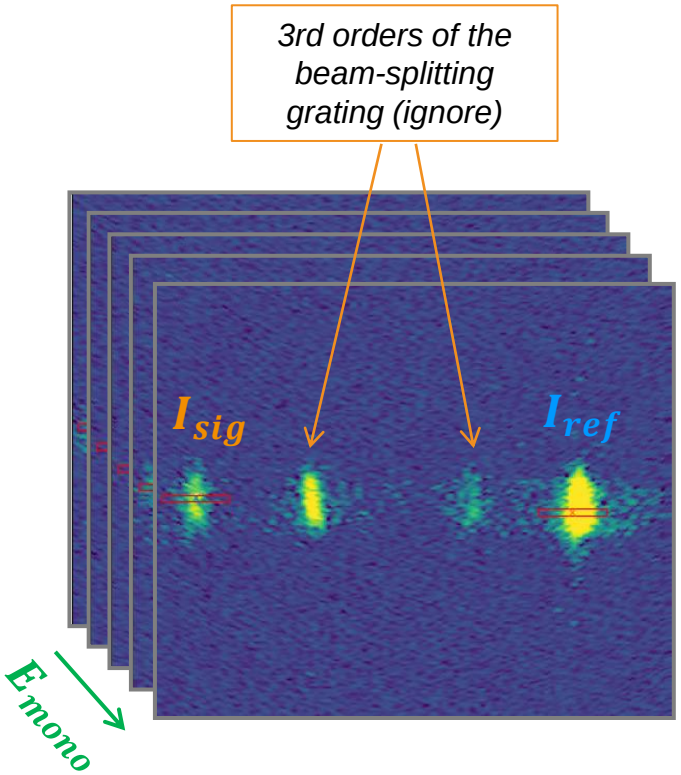
Test sample:
Acetonitrile
1M solution in ethanol

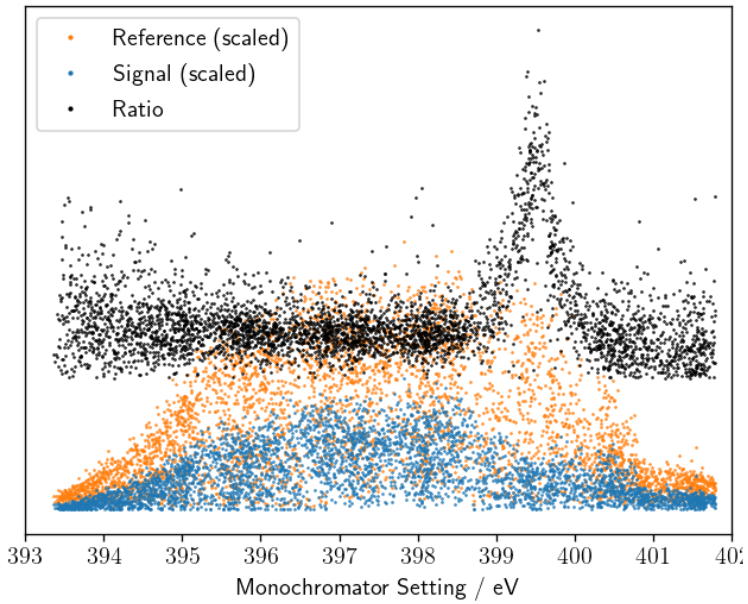


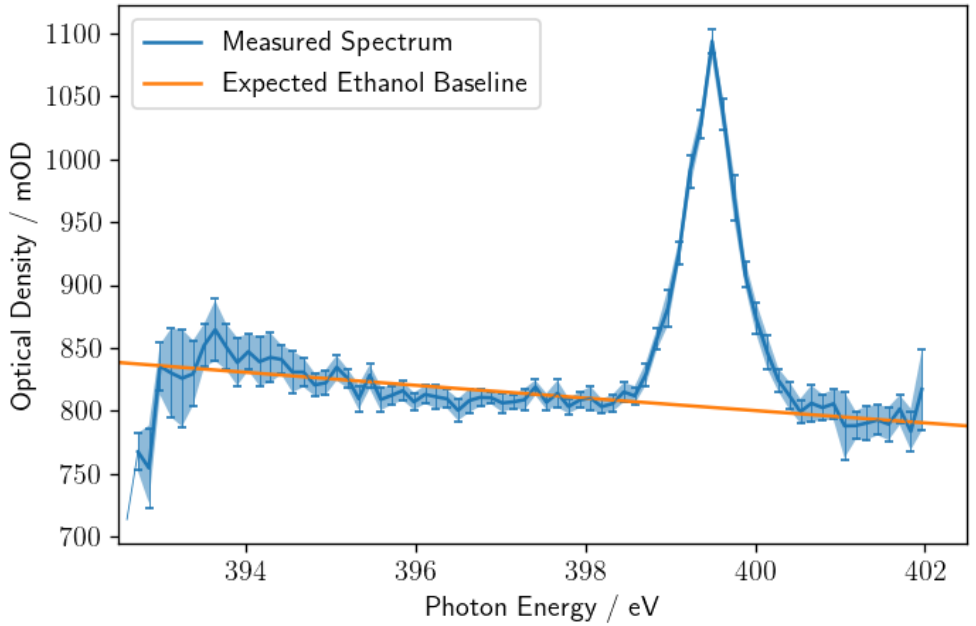


PG2 Beamline

Analysis



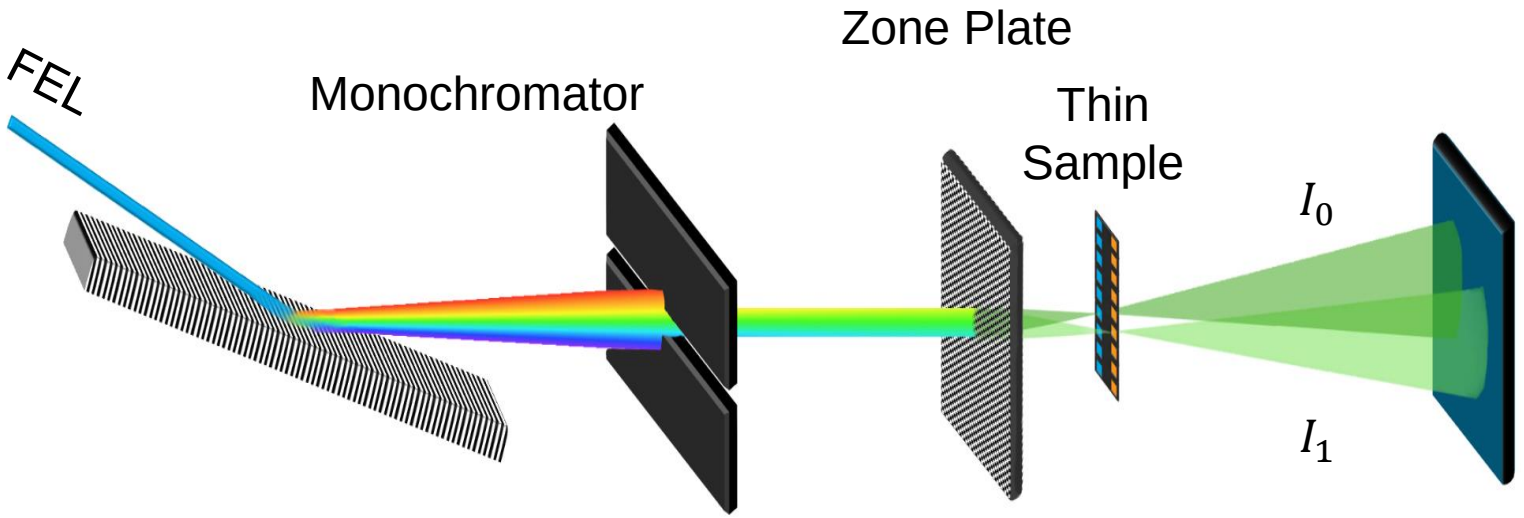


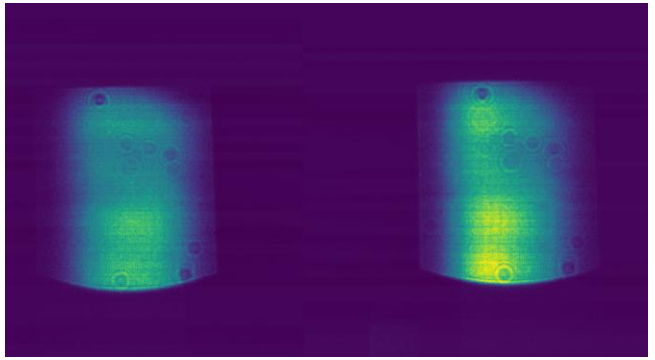


"Shot noise limited soft x-ray absorption spectroscopy in solution at a SASE-FEL using a transmission grating beam splitter"
– *Engel, Robin Y. et al.,* Structural Dynamics **8**, 014303 (2021)

- **Photon shot noise limited**
- Flux **increases** by 10^3 to 10^4 possible
- Next Beamtimes Planned
- **New endstation** in construction: **FLUX**

#2: Zone Plate focused Monochromatic European XFEL – SCS





Science case:
Understand ultrafast **FEL induced distortions** to electronic structure and the impact of **non-linear effects** (multi-photon interactions) on FEL measurements.

Open community proposal at SCS

Proposers

Dr. Martin Beye (DESY Deutsches Elektronen-Synchrotron, Germany), Experiment Role: PI
Prof. Dr. Giacomo Ghiringhelli (Politecnico di Milano, Italy) Experiment role: Co-proposer
Dr. Benedikt Rösner (Paul Scherrer Institut (PSI), Switzerland) Experiment role: Co-proposer
Mr. Adam Kubec (Paul Scherrer Institut (PSI), Switzerland) Experiment role: Co-proposer
Dr. Christian David (Paul Scherrer Institut (PSI), Switzerland) Experiment role: Co-proposer
Prof. Dr. Kai Rossnagel (Christian-Albrechts-Universität zu Kiel (CAU), Germany) Experiment role: Co-proposer
Dr. Ivan Baev (University of Hamburg (UHH), Germany) Experiment role: Co-proposer
Dr. Piter Miedema (European XFEL GmbH, Germany) Experiment role: Co-proposer
PD Dr. Tim Laarmann (DESY Deutsches Elektronen-Synchrotron, Germany) Experiment role: Co-proposer
Dr. Michael E Stückelberger (DESY Deutsches Elektronen-Synchrotron, Germany) Experiment role: Co-proposer
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Dr. Sam M. Vinko (University of Oxford, United Kingdom) Experiment role: Co-proposer
Dr. Robert E Carley (European XFEL GmbH, Germany) Experiment role: Co-proposer
Dr. Laurent Mercadier (European XFEL GmbH, Germany) Experiment role: Co-proposer
Dr. Justine Schlappa (European XFEL GmbH, Germany) Experiment role: Co-proposer
Dr. Alexander Yaroslavtsev (Uppsala University - UU, Sweden) Experiment role: Co-proposer
Dr. Giuseppe Mercurio (European XFEL GmbH, Germany) Experiment role: Co-proposer
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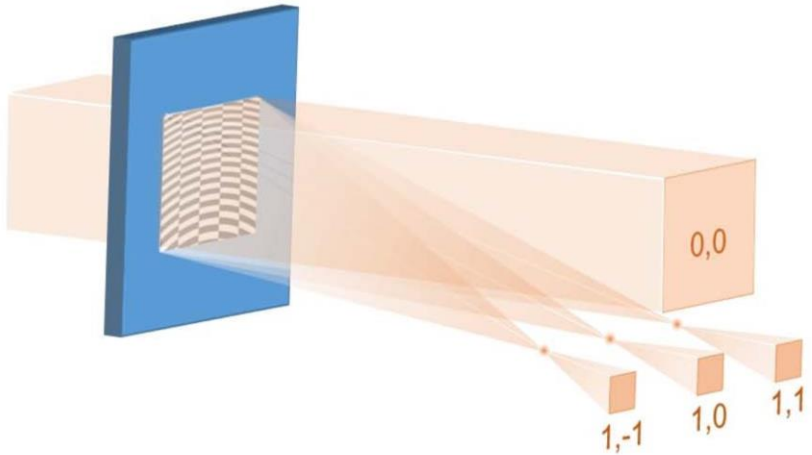
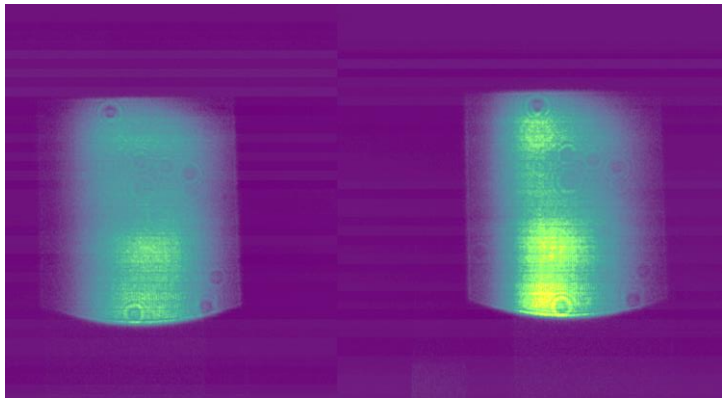
Dr. Thomas R. Preston (European XFEL GmbH, Germany) Experiment role: Co-proposer
Dr. Manuel Izquierdo (European XFEL GmbH, Germany) Experiment role: Co-proposer
Dr. Andreas Scherz (European XFEL GmbH, Germany) Experiment role: Co-proposer
Dr. Loic Le Guyader (European XFEL GmbH, Germany) Experiment role: Co-proposer
Prof. Dr. Artem Rudenko (Kansas State Univ., United States) Experiment role: Co-proposer
Prof. Dr. Jonathan Phillip Marangos (Imperial College London, United Kingdom) Experiment role: Co-proposer
Dr. Zhong Yin (European XFEL GmbH, Germany) Experiment role: Co-proposer
Dr. Marco Moretti (European Synchrotron Radiation Facility - ESRF, France) Experiment role: Co-proposer
Dr. Daniel Higley (SLAC National Accelerator Laboratory, United States) Experiment role: Co-proposer
Dr. William F. Schlotter (SLAC National Accelerator Laboratory, United States) Experiment role: Co-proposer

… and some of their students.

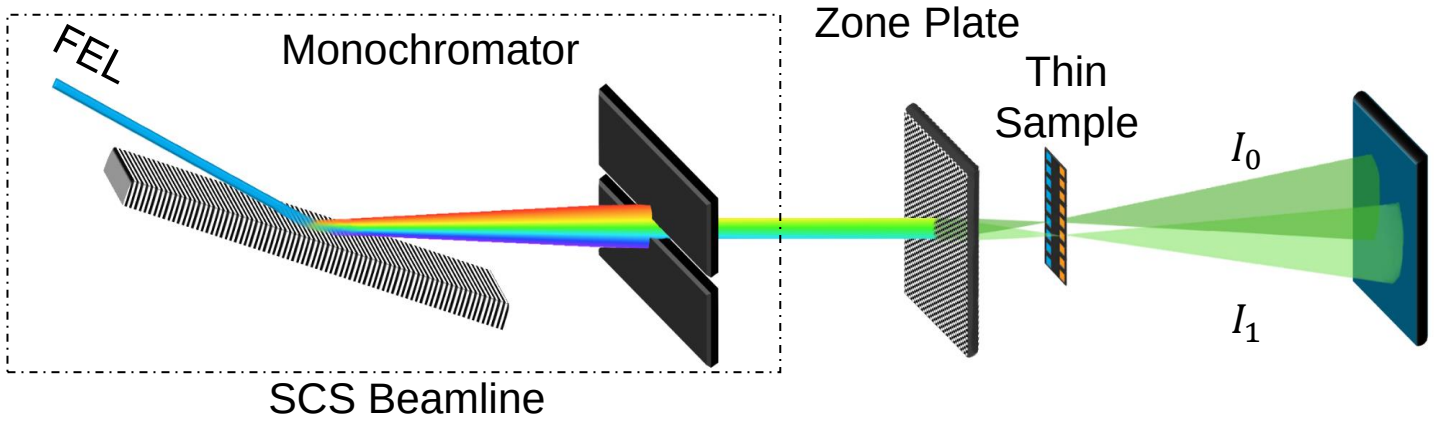
Multifocus Transmission Zone Plate Setup

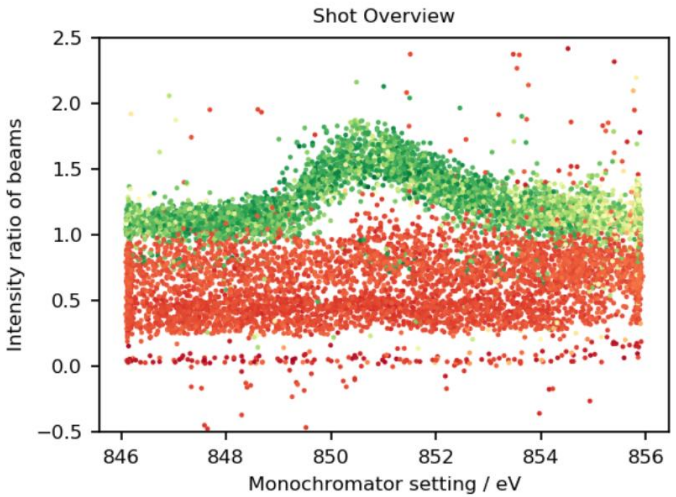
Combining a grating and an off-axis zone plate in one element



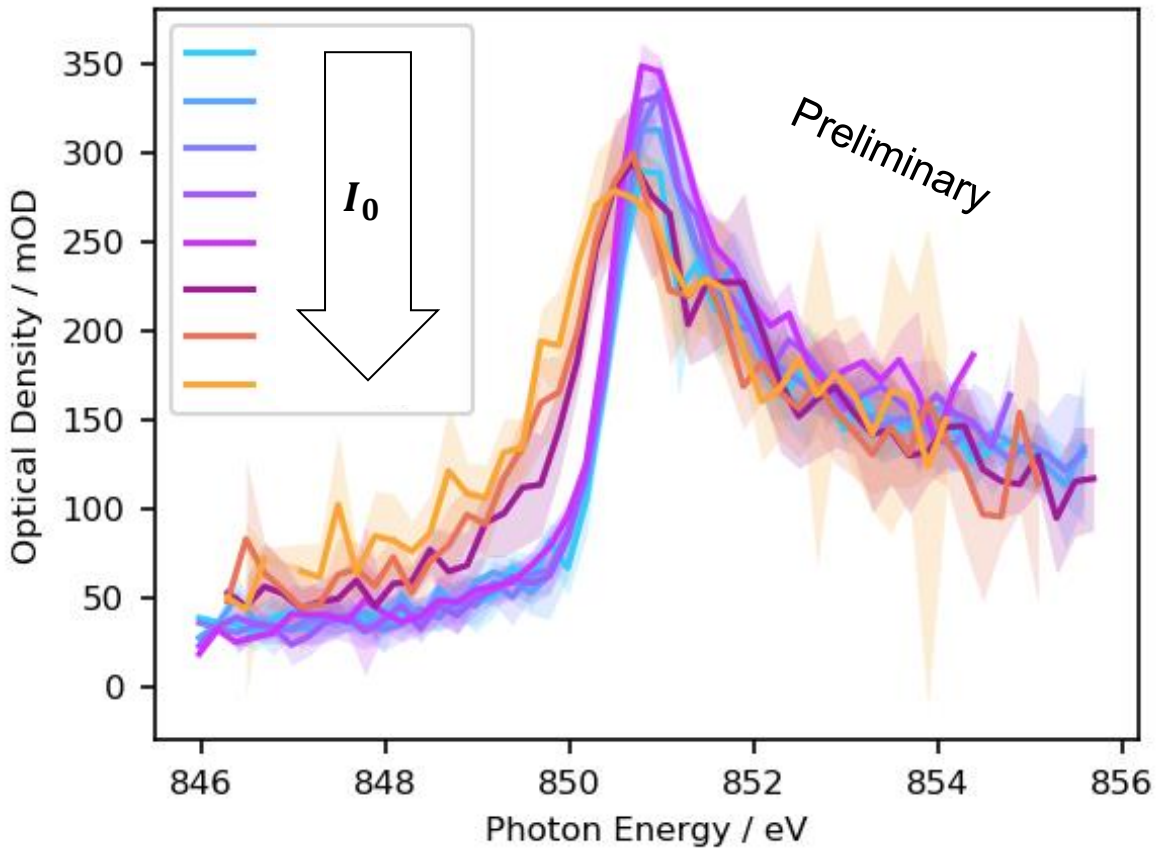


Multifocus off-axis zone plates for x-ray
free-electron laser experiments - F. Döring, B. Rösner et al.
Optica, Vol. 7, Issue 8, pp. 1007-1014 (2020)

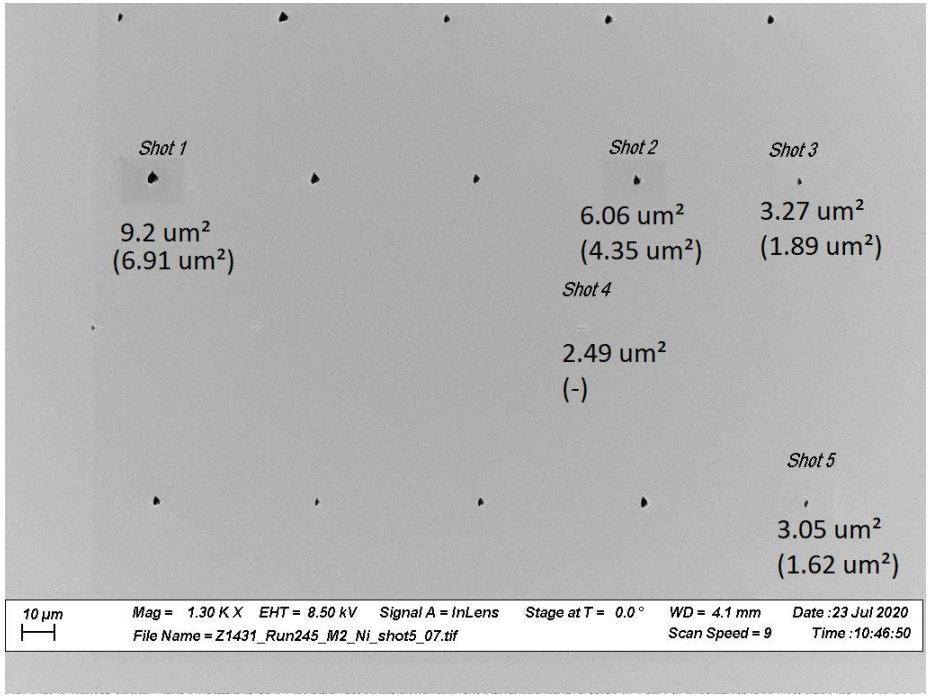




Determining the Fluence Dependency

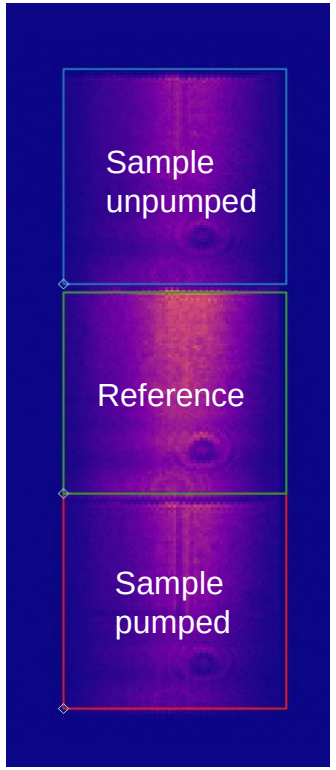


Imprints of the beam on the sample



Analysis ongoing.

Pump-Probe Studies

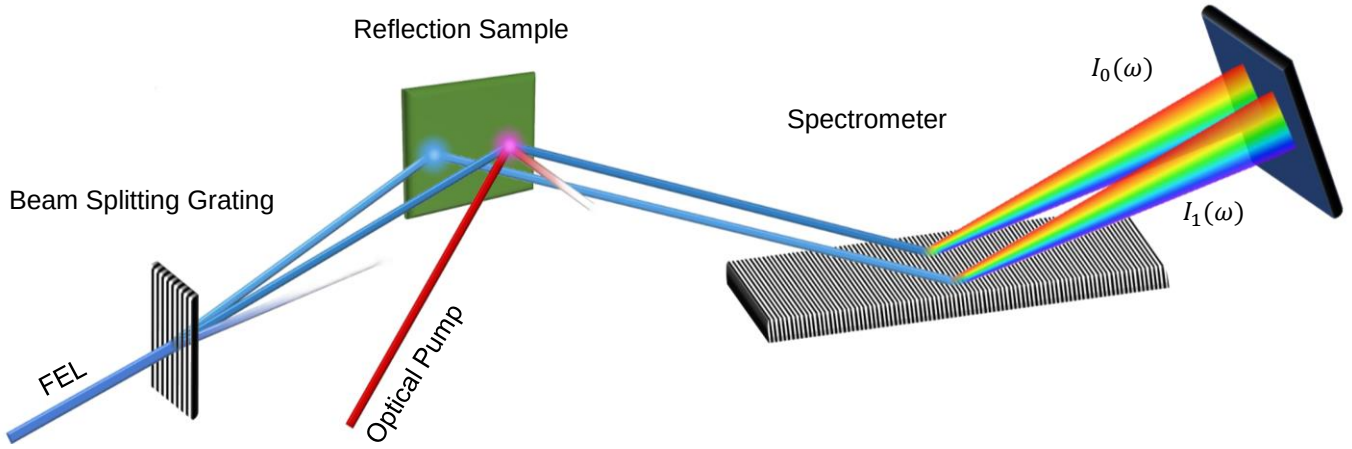


Non-destructive IR-pump FEL-probe study on
NiO – European XFEL, SCS

See upcoming publication
T. Lojewski, et al. publication in progress
Group of A. Eschenlohr University of Duisburg-Essen

Plots are a courtesy of A. Eschenlohr

#3: Parallel Broadband reflection FLASH II – FL24



Science case:
Explore the response of **a correlated insulator** (NiO) to **sub-bandgap excitations**.

People Involved

Many thanks to:

Martin Beye
Jan O. Schunck
Piter Miedema
Siarhei Dziarzhytski
Günter Brenner
Marion Kuhlmann

Xiaocui Wang
Igor Vaskivskyi
Diego Turenne
Hermann A. Dürr

Christian Schüßler-Langenheine

Max Born Institute for Nonlinear Optics and Short Pulse Spectroscopy

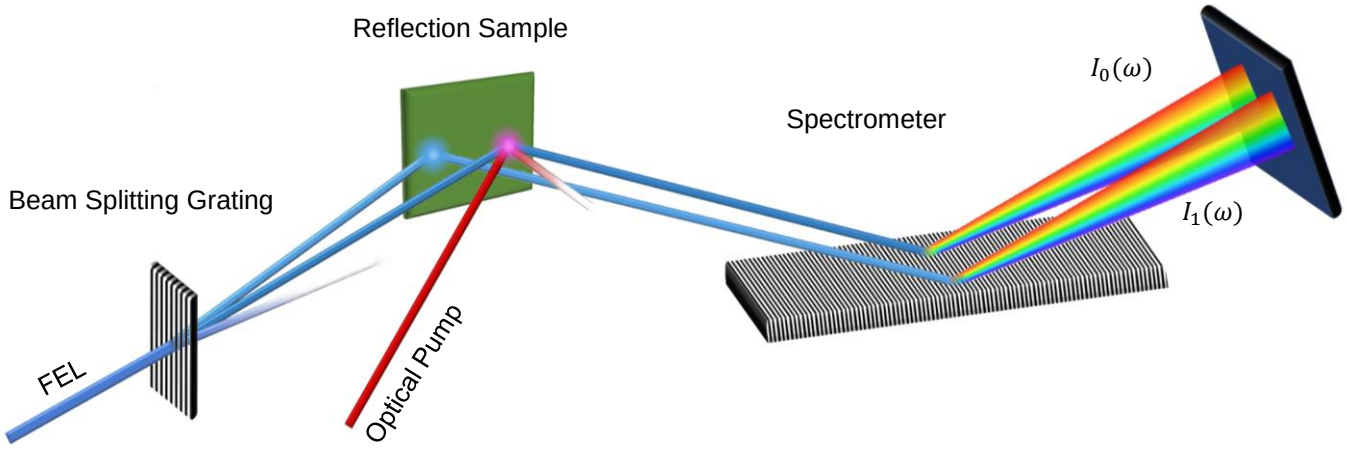
UPPSALA UNIVERSITET

Andriy Styervoyedov
Stuart S. P. Parkin

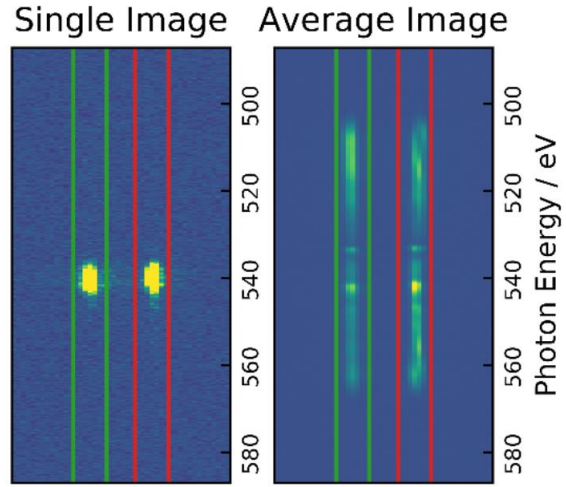
Florian Döring
Benedikt Rösner
Christian David

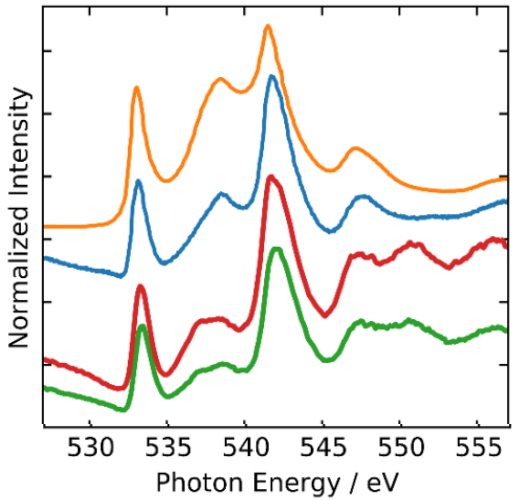
Parallel Broadband Reflection Spectroscopy

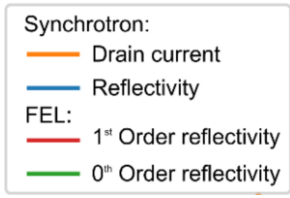
- Reflectivity
  - Possible on bulk samples (no thin films needed)
  - Equivalent information to absorption
- Spectrometer behind sample
- 3rd harmonic of FEL



*"Parallel Broadband Femtosecond Reflection Spectroscopy at a Soft X-Ray Free-Electron Laser.", Engel, Robin Y., et al. – Applied Sciences **2020** 10(19), 6947.*







Normalization of pump probe differences over two spots

- Naive pump-probe normalization:

$$\Delta_{XAS} = I^{Pumped}_{Spot\ sig} - I^{Unpumped}_{Spot\ ref}$$

  - *This neglects sample inhomogeneities!*

- Alternative: Switching opt. laser on and off

$$\Delta_{XAS} = I^{Pumped}_{Spot\ sig} - I^{Unpumped}_{Spot\ sig}$$

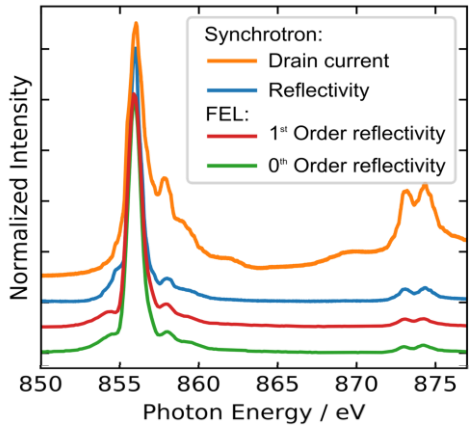
  - *This neglects FEL fluctuations!*

- Combined approach:

$$\Delta_{XAS} = I^{Pump\ on}_{Spot\ sig} - I^{(Pump\ on)}_{Spot\ ref} \cdot \frac{I^{Pump\ off}_{Spot\ sig}}{I^{Pump\ off}_{Spot\ ref}}$$

FEL fluctuations

Spot difference

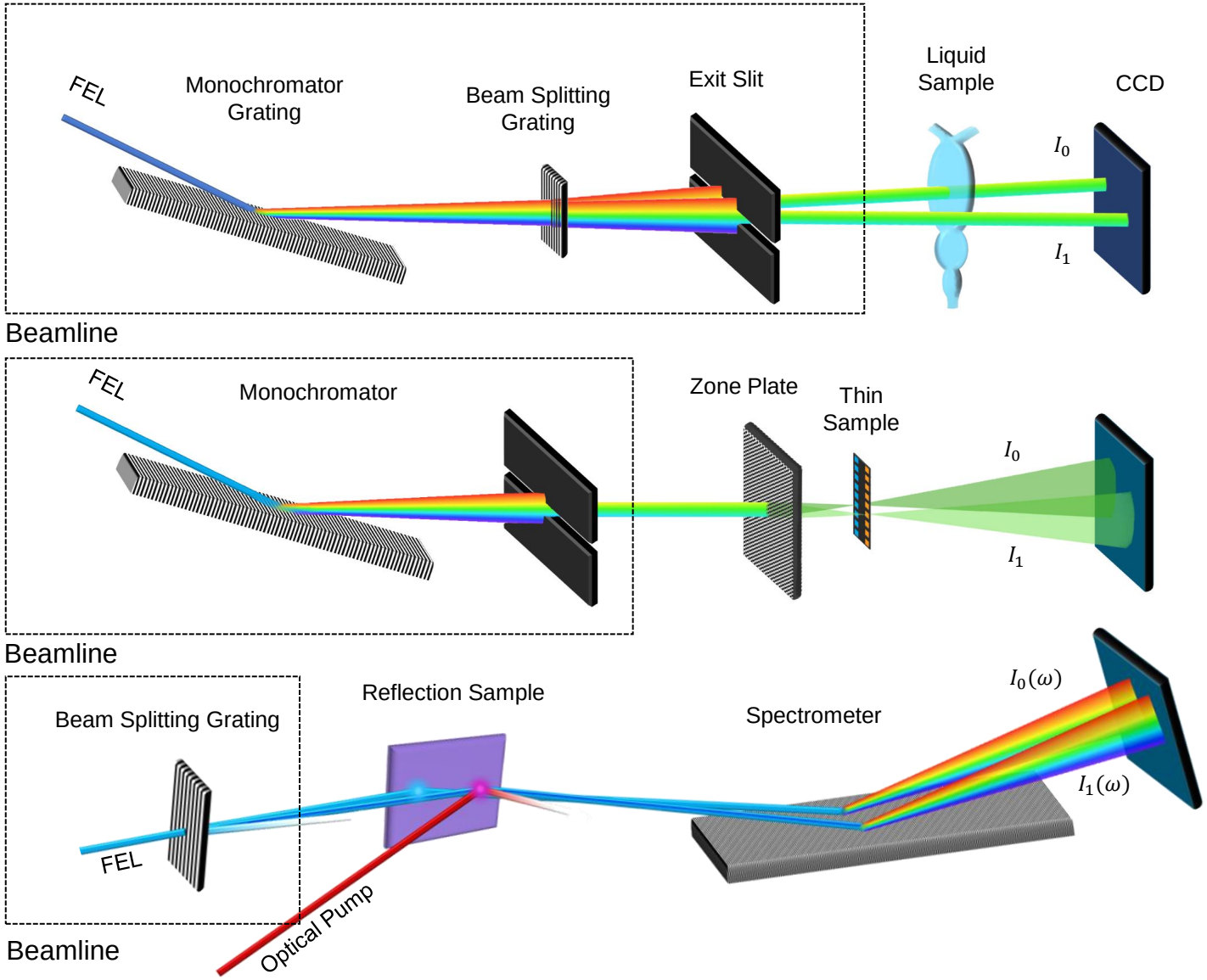


See upcoming publication

See upcoming publication

Pump-probe plots by C. Wang,
Publication in progress
Group H. Dürr, Upsala University

Summary



Liquid Jet – PG 2, FLASH I

+ Integrated into beamline
+ Shot noise limited measurement
- Beam transport of 2 beams

Zone Plate – SCS, European XFEL

+ Clean, efficient setup
+ Large sensitivity and dynamic range
+ Excellent focus
- Dedicated setup

Broadband – FL24, FLASH II

+ Utilizes full FEL spectrum
+ Bulk samples possible
- Sample inhomogeneity

Thank You!