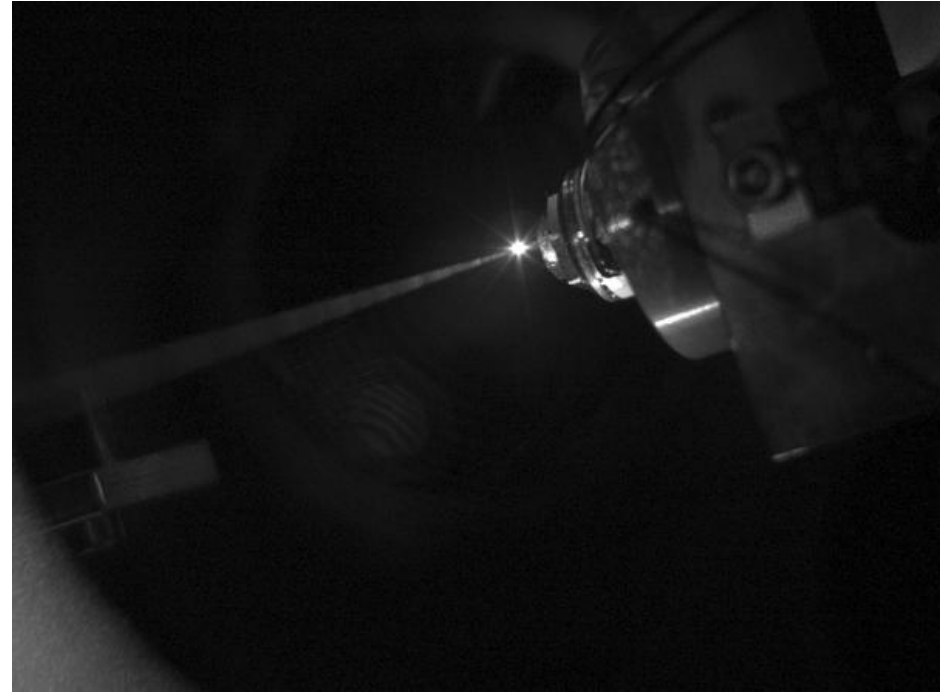
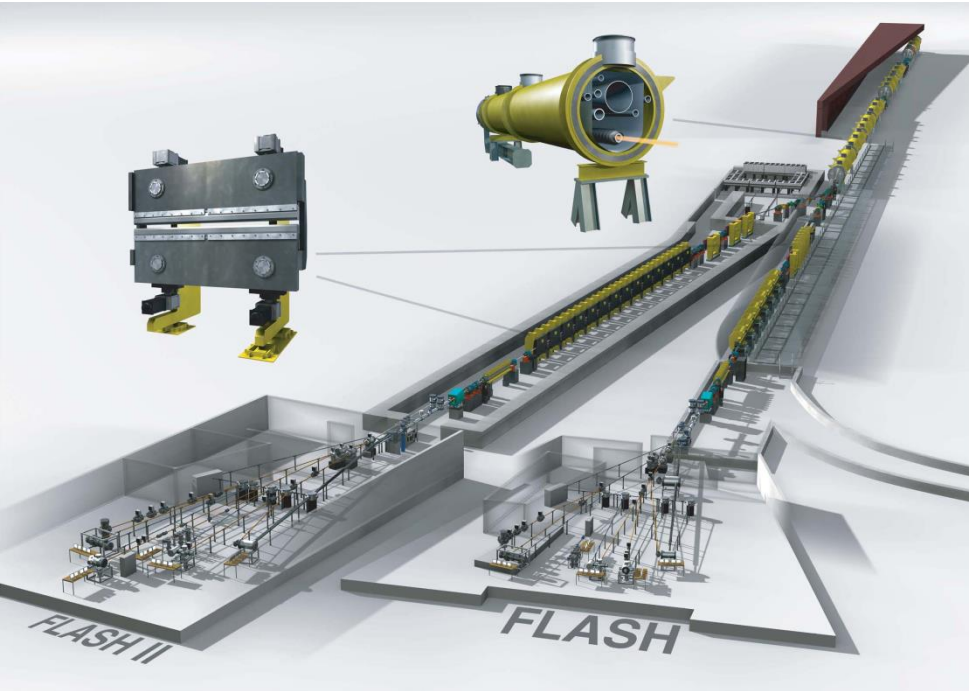


Characterization of Light (Photon Diagnostics @ FLASH).



Sven Toleikis

DESY Ukraine Winter School 2023

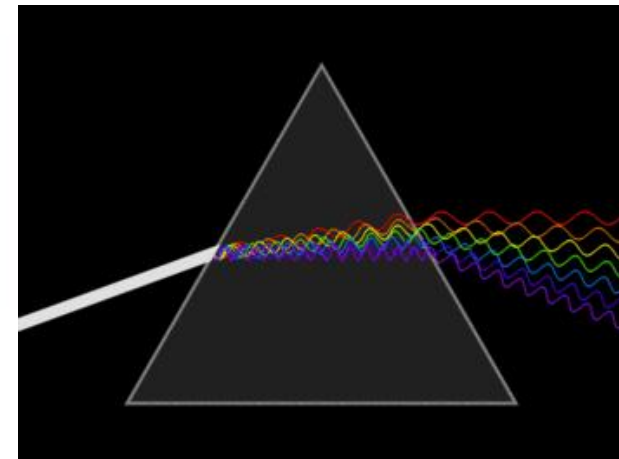
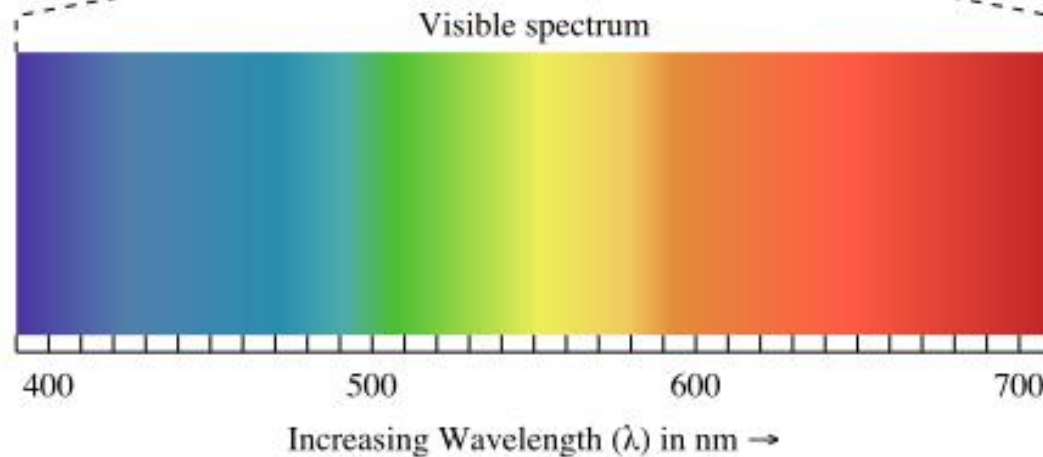
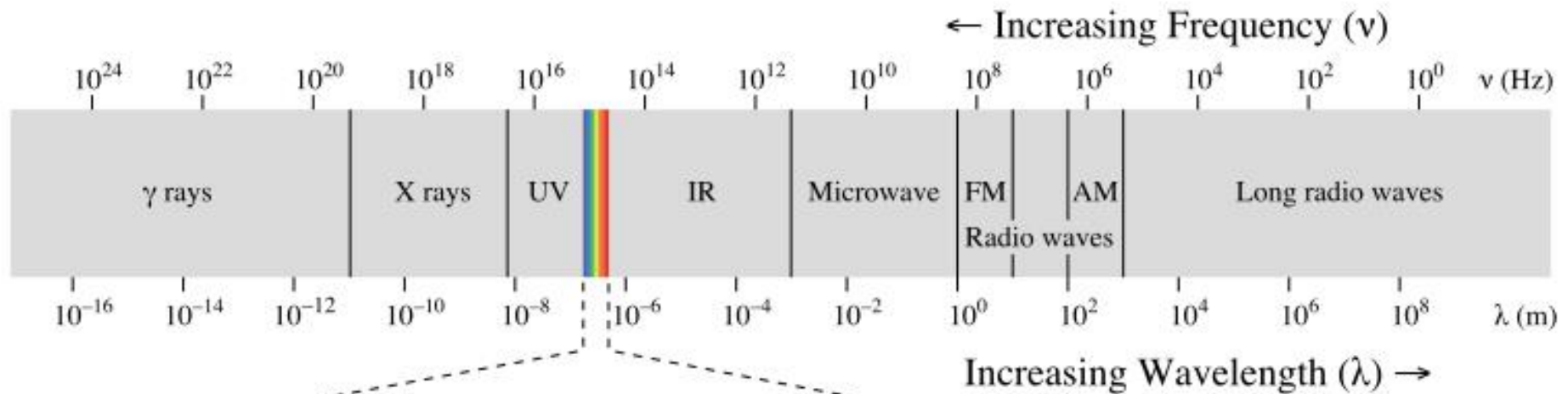
16 Februar 2023

Outline

- Introduction – what is light?
- Light generation
 - XUV FEL light at FLASH
 - THz light at FLASH
- Light transportation
 - Beamlines: beam transport (mirrors), beam manipulation, etc.
- Light characterization
 - Intensity (# of photons) / Beam position
 - Wavelength
 - Pulse duration / Jitter (to another light source)
 - Polarization
 - Coherence / Wavefront
- Light utilization
 - Example experiments

Light – Intro I

- Light -> more general: electro-magnetic radiation



Light – Intro II

- Frequency \leftrightarrow Wavelength \leftrightarrow Energy

CLASS	FREQUENCY	WAVELENGTH	ENERGY
Y	300 EHz	1 pm	1.24 MeV
HX	30 EHz	10 pm	124 keV
SX	3 EHz	100 pm	12.4 keV
SX	300 PHz	1 nm	1.24 keV
EUV	30 PHz	10 nm	124 eV
NUV	3 PHz	100 nm	12.4 eV
NIR	300 THz	1 μ m	1.24 eV
MIR	30 THz	10 μ m	124 meV
FIR	3 THz	100 μ m	12.4 meV
EHF	300 GHz	1 mm	1.24 meV
SHF	30 GHz	1 cm	124 μ eV
UHF	3 GHz	1 dm	12.4 μ eV
VHF	300 MHz	1 m	1.24 μ eV
HF	30 MHz	10 m	124 neV
MF	3 MHz	100 m	12.4 neV
LF	300 kHz	1 km	1.24 neV
VLF	30 kHz	10 km	124 peV
VF/ULF	3 kHz	100 km	12.4 peV
SLF	300 Hz	1 Mm	1.24 peV
ELF	30 Hz	10 Mm	124 feV
ELF	3 Hz	100 Mm	12.4 feV

Sources at DESY:

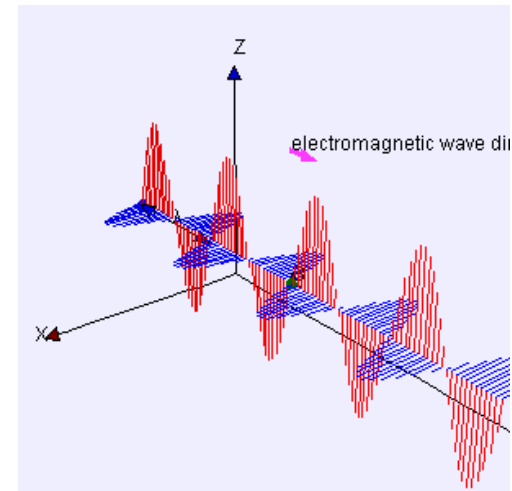
- > PETRAIII:
250 eV (XUV) – 200 keV (hard X-rays)
- > FLASH:
90 nm – 3.5 nm (~14 eV – 350 eV)
- > Optical lasers:
UV to NIR
- > THz@FLASH:
1 μ m – 300 μ m (300 THz – 1 THz)

Light – Intro III

- electro-magnetic radiation – theory
- electric field \mathbf{E} , magnetic field \mathbf{B} , electric potential φ , magnetic potential \mathbf{A}

Maxwell's equations (*vector fields*)

$\nabla \cdot \mathbf{E} = \frac{\rho}{\epsilon_0}$	<u>Gauss' law</u>
$\nabla \cdot \mathbf{B} = 0$	<u>Gauss's law formagnetism</u>
$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}$	<u>Faraday's law</u>
$\nabla \times \mathbf{B} = \mu_0 \mathbf{J} + \mu_0 \epsilon_0 \frac{\partial \mathbf{E}}{\partial t}$	<u>Ampère–Maxwell law</u>



linearly polarized plane wave

Maxwell's equations (*Potential formulation*)

$$\nabla^2 \varphi + \frac{\partial}{\partial t} (\nabla \cdot \mathbf{A}) = -\frac{\rho}{\epsilon_0}$$

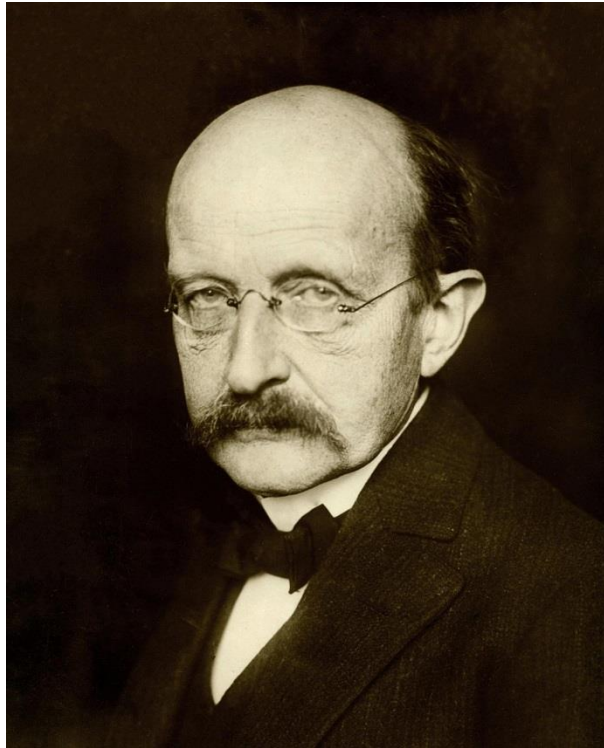
$$\left(\nabla^2 \mathbf{A} - \frac{1}{c^2} \frac{\partial^2 \mathbf{A}}{\partial t^2} \right) - \nabla \left(\nabla \cdot \mathbf{A} + \frac{1}{c^2} \frac{\partial \varphi}{\partial t} \right) = -\mu_0 \mathbf{J}$$

$$\mathbf{E} = -\nabla \varphi - \frac{\partial \mathbf{A}}{\partial t}$$

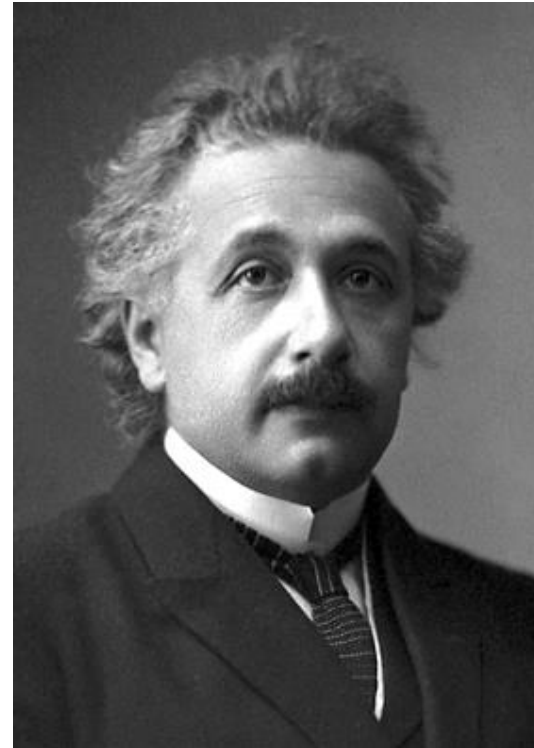
$$\mathbf{B} = \nabla \times \mathbf{A}$$

Light – Intro IV

- Electro-Magnetic radiation is quantized: $E = hf = \frac{hc}{\lambda}$
- Universal particle-wave duality (Louis de Broglie)

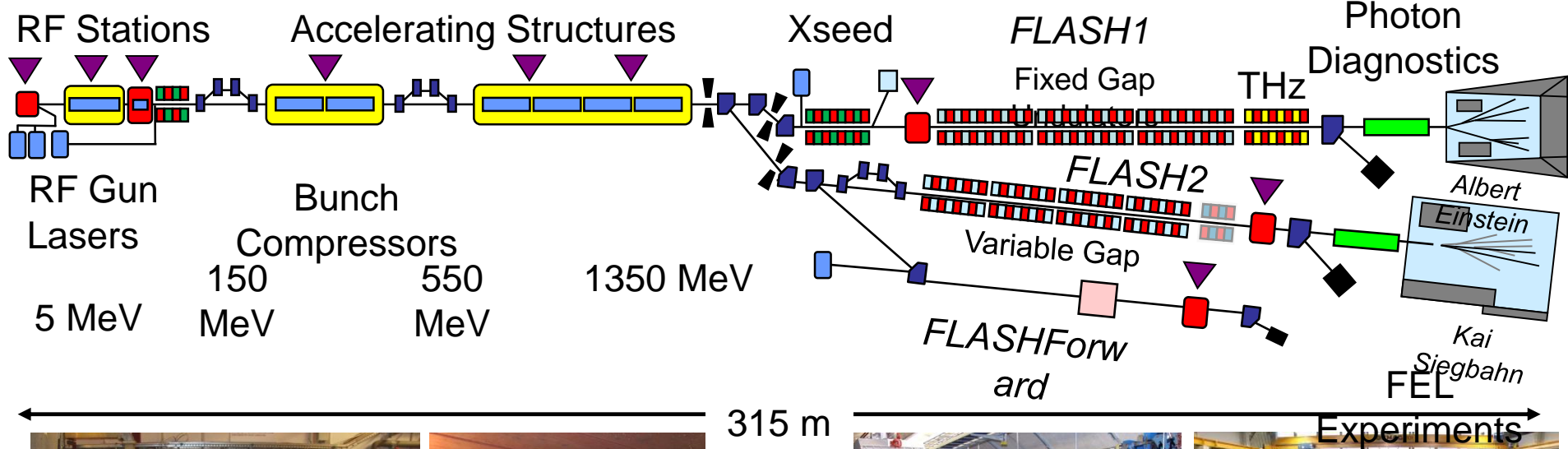


Max Planck (1858-1947)
Nobel Prize in Physics 1918

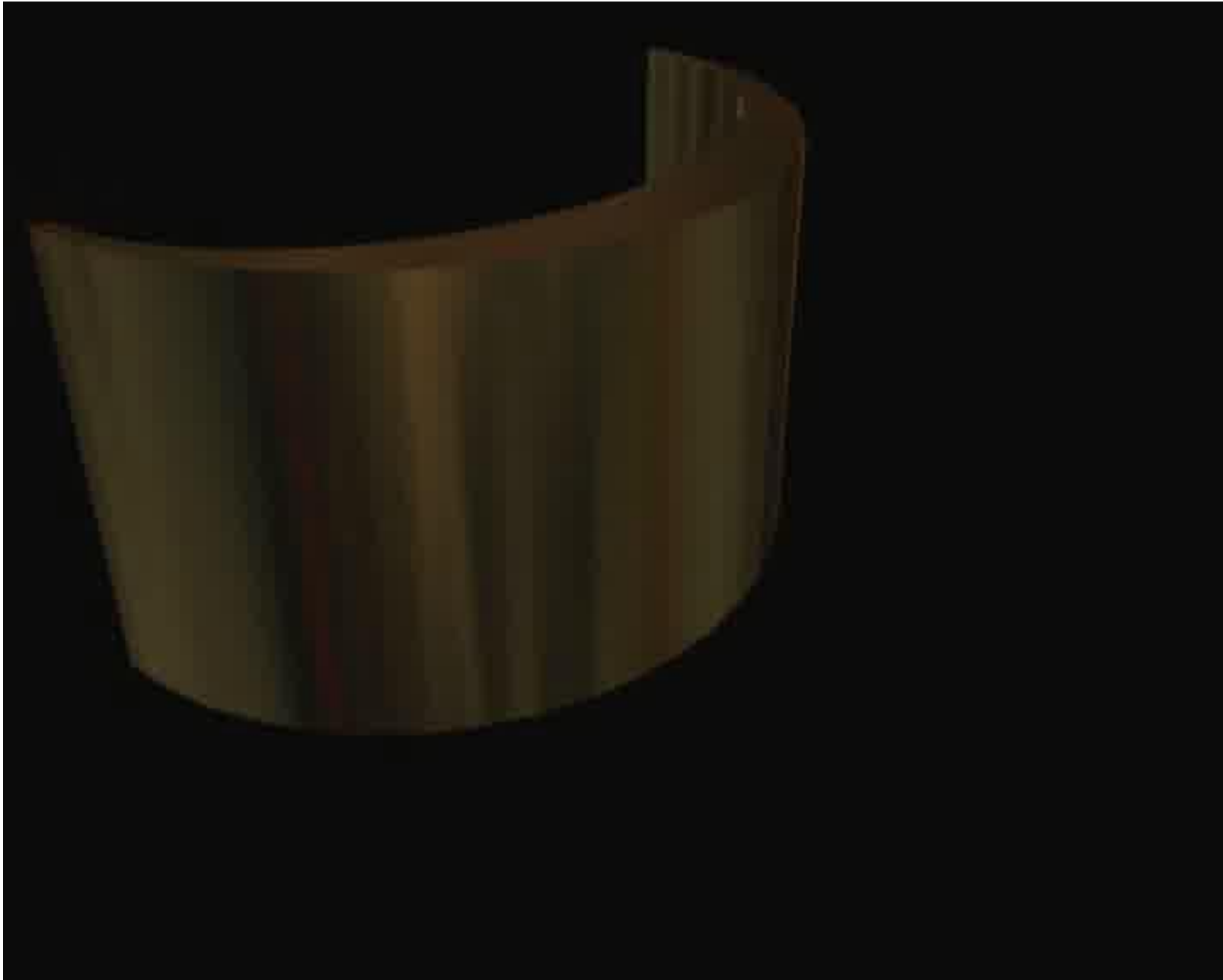


Albert Einstein (1879-1955)
Nobel Prize in Physics 1921

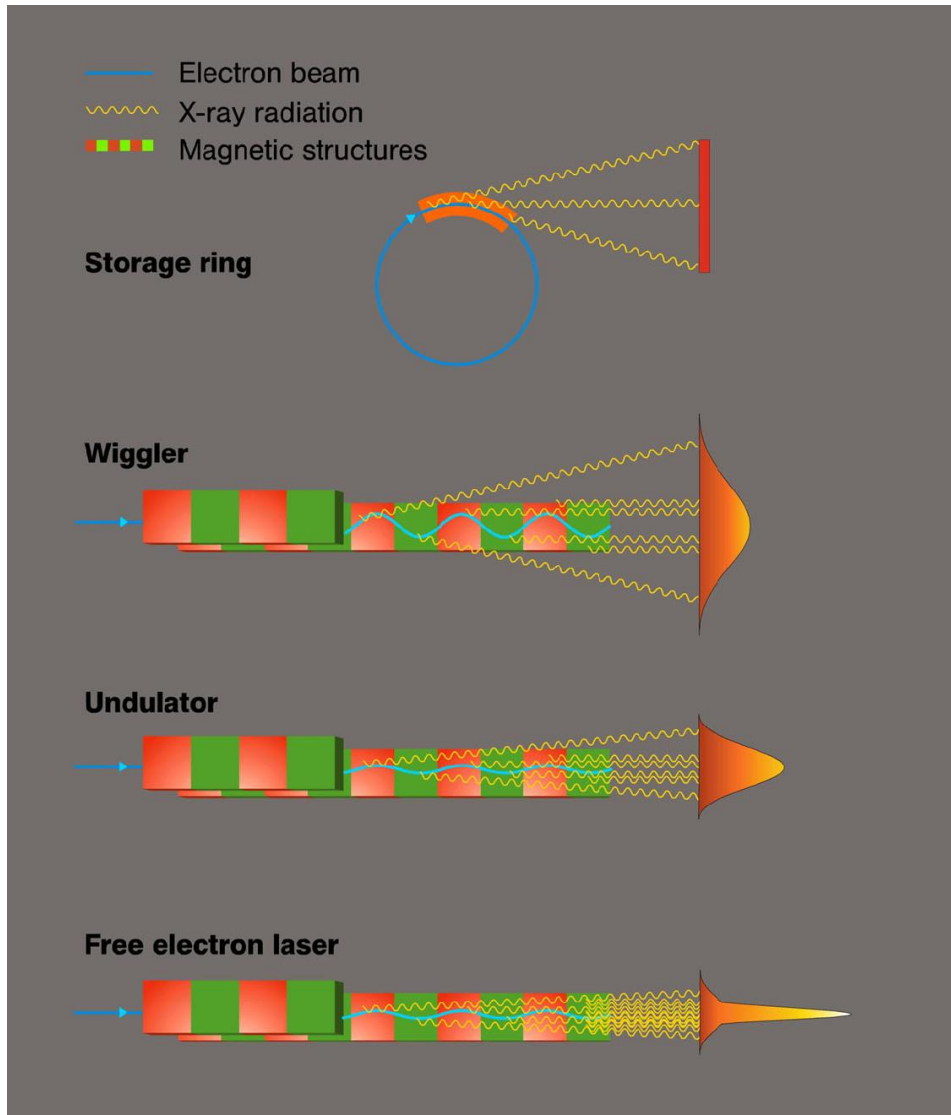
The FLASH facility



SASE movie



Generation of light – accelerator based



N_U or N_W = # of magnetic periods

N_e = # of electrons in a bunch

bending magnet radiation

$\propto N_W \times$ bending magnet

$\propto N_U^2 \times$ bending magnet

$\propto N_U^2 \times N_e \times$ bending magnet

N_e = # of electrons in a bunch; e.g. @ FLASH with typ. 0.5 nC per bunch
-> $N_e = 3.12 \times 10^9$!!!

Current photon parameters of FLASH

Wavelength range (fundamental):

3.5-60 nm (FL1); 3.5-90 nm (FL2)

Spectral width (FWHM):

0.5– 1.5 %

Pulse energy:

**up to 200 μ J (average),
500 μ J (peak), \sim 1 mJ (FL2)**

Pulse duration (FWHM):

\sim 5 fs - 200 fs

Peak power (fundamental):

1-5 GW

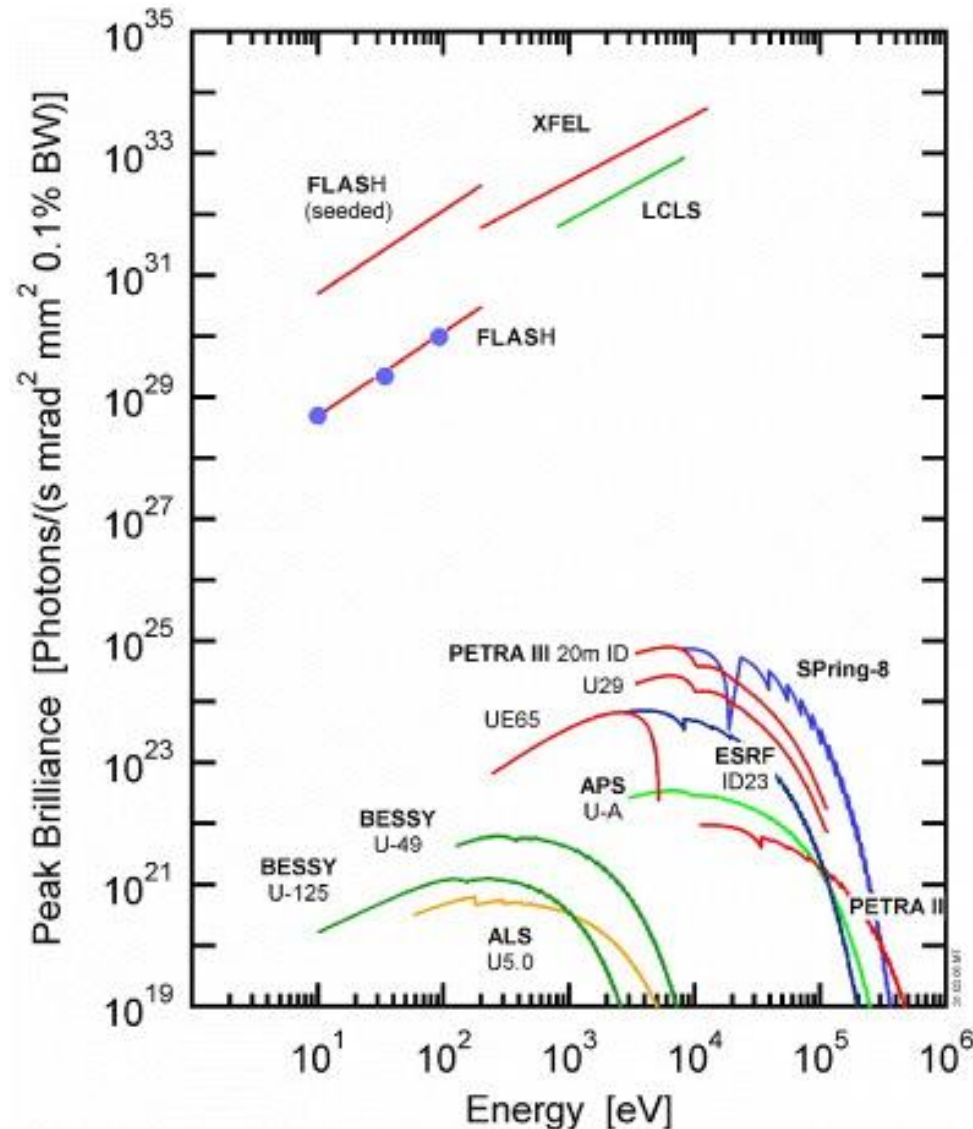
Average power (fundamental):

**up to 5 W
(up to 6000 pulses/s)**

Peak brilliance:

up to $\sim 10^{30}$

Photons/(s mrad² mm² 0.1% BW)



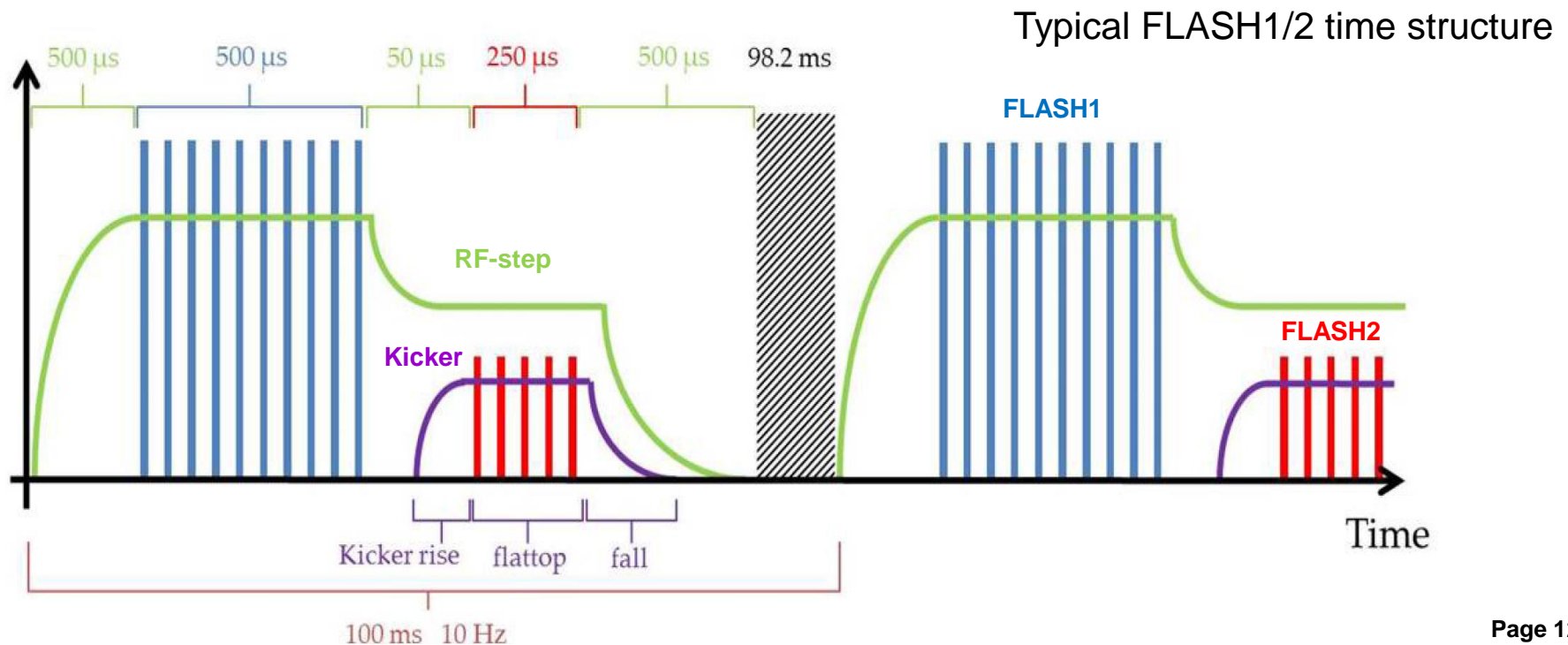
SASE FEL Properties

- > high intensity (GW peak power)
- > coherence (laser-like radiation)
- > femtosecond pulses!
- > narrow bandwidth!
- > full wavelength tunability!
- > down to X-rays!

- > But: shot-to-shot fluctuations (w/o seeding)
 - > very good photon diagnostics are mandatory!

Simultaneous FLASH1 and FLASH2 operation

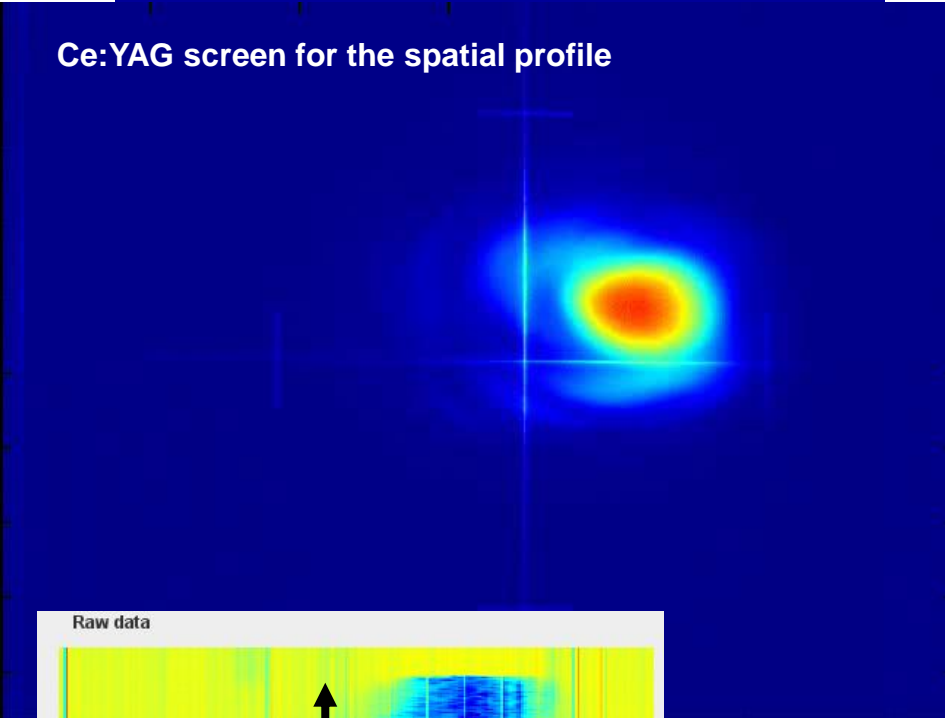
- Superconducting accelerator technology allows long RF-pulses (10 Hz, up to 800 μs)
→ long electron bunch train, which can be shared between FLASH1 and FLASH2 with 10 Hz
- Fast kicker and Lambertson septum used to extract a part of the bunch train to FLASH2
- Two injector lasers: FLASH1 and FLASH2 bunch pattern and charge selected independently
- Flexible RF-system: amplitude and phase is adjusted independently for FLASH1 and FLASH2



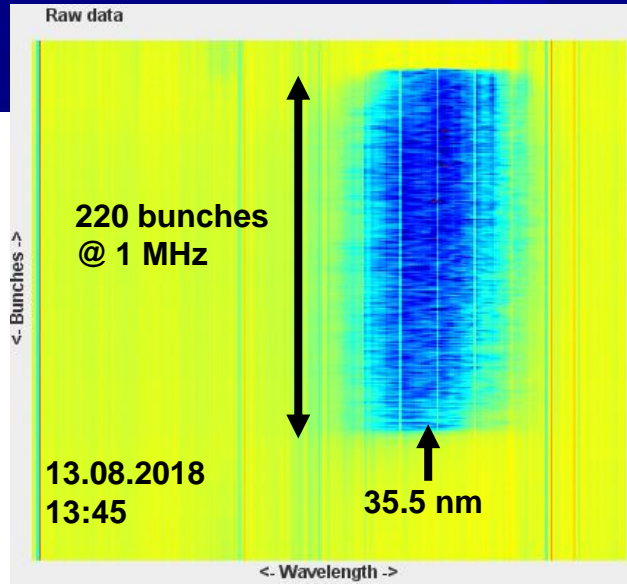
FLASH performance

Spatial Profile

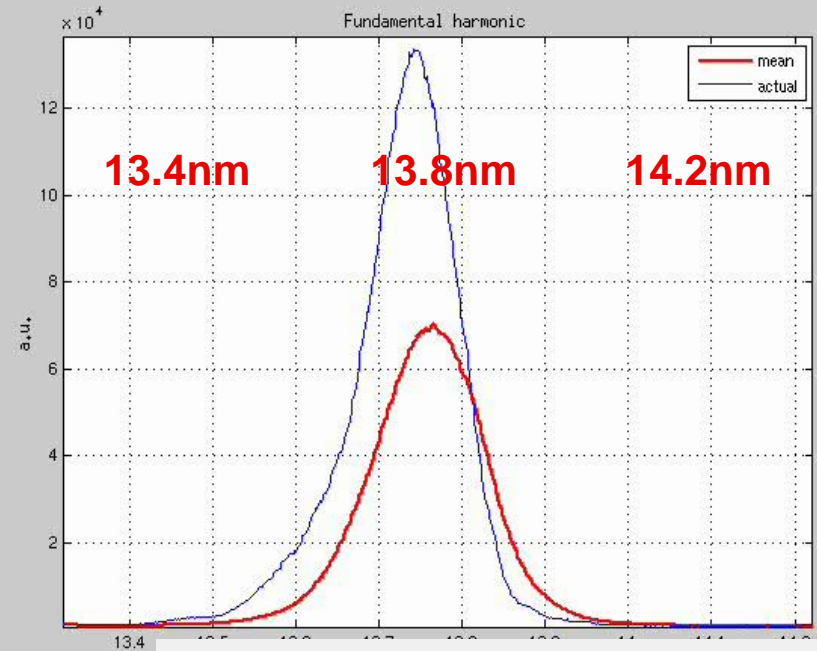
Ce:YAG screen for the spatial profile



Raw data

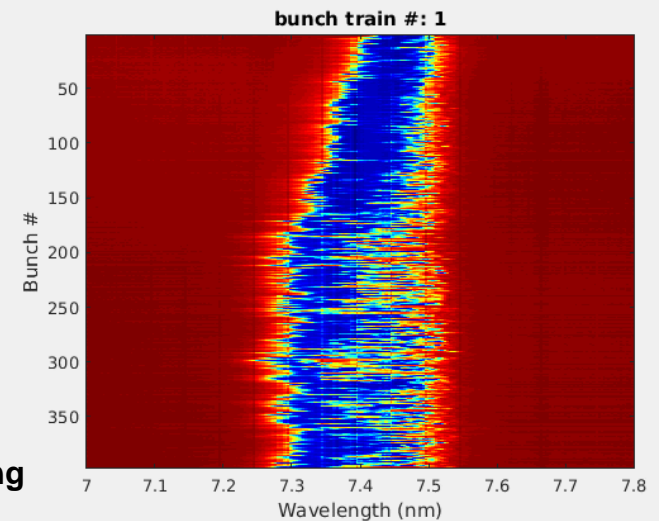


Spectral distribution



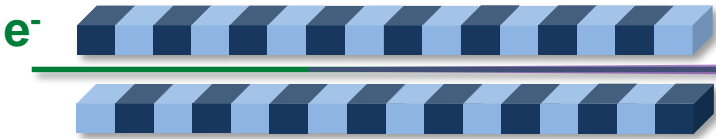
VLS spectrometer
with Kalypso line detector
@ 1 MHz

28.05.2018
During tuning



THz @ FLASH1

XUV undulators



Undulator:

- Intensity $\sim N_e^2$ (up to 100 μJ)
- Synchronized to XUV pulse (<5 fs jitter)
- narrowband ($\sim 10\%$)
+ tunable (0.6 – 300 μm)
- polarization: linear

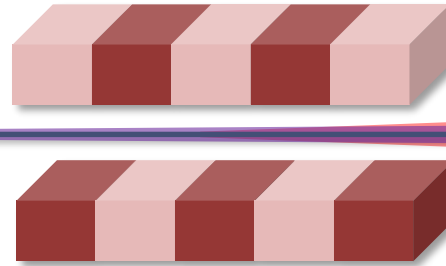
Dump magnet:

- Intensity up to 10 μJ
- broadband (single cycle)
 $\sim 100\%$ @ 1.5 THz
- polarization: Radial

Peak fields:

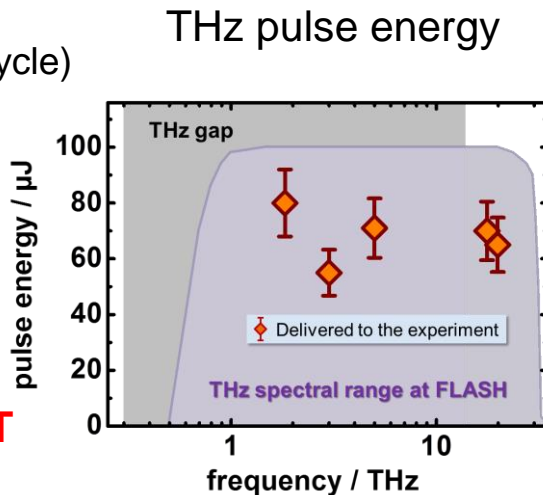
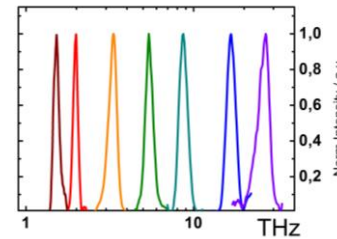
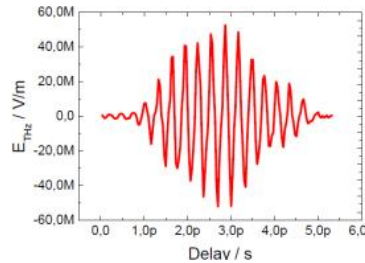
$E \sim \text{GV/m}$ & $B \sim 1 \text{ T}$
DESY.

THz undulator

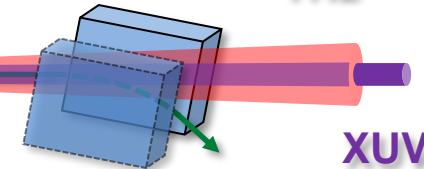


Tunable + narrow band:

1 – 300 μm (4 meV – 1 eV)

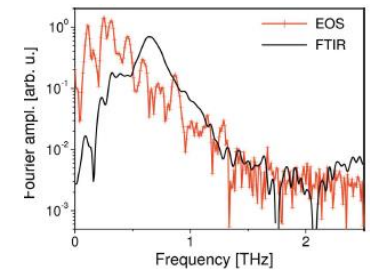
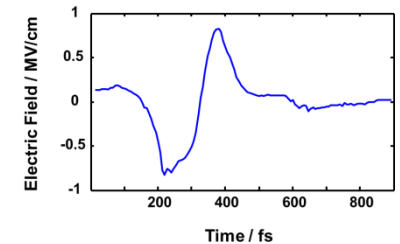


Dump magnet



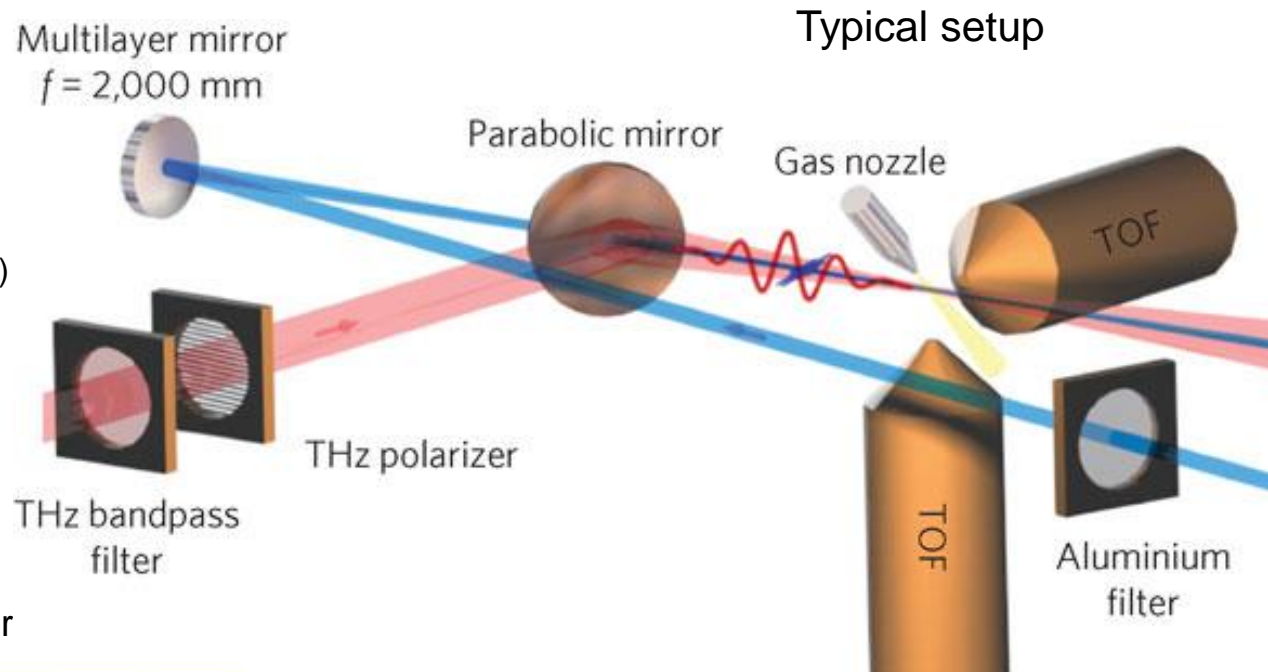
+ Single cycle

(50 – 300 mm) (4 – 24 meV)

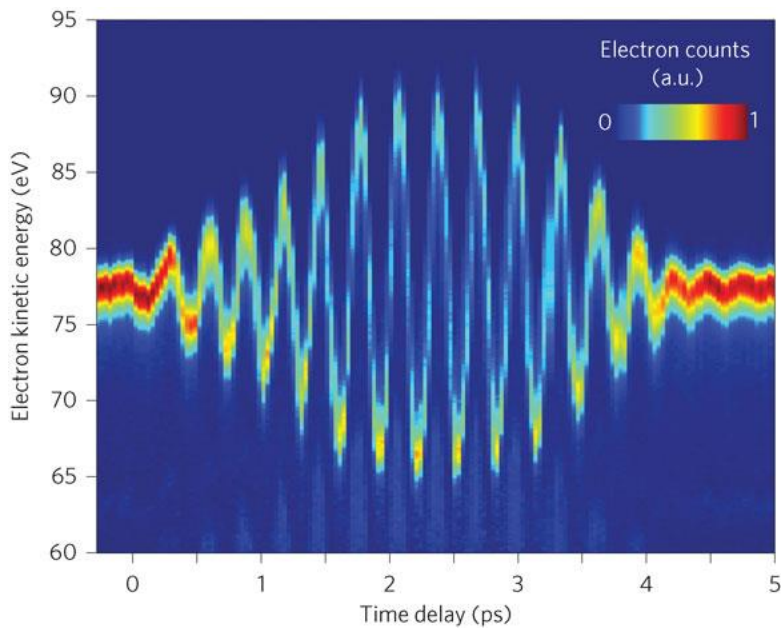


THz streaking experiment

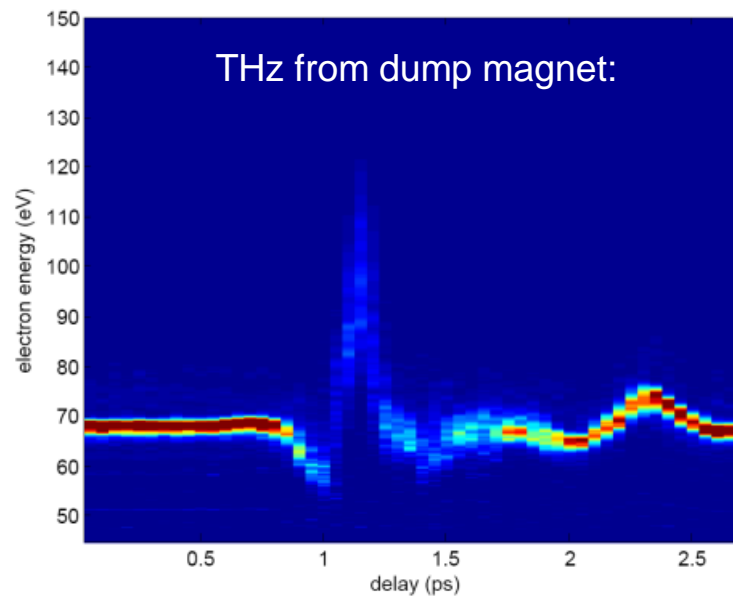
Nature Photonics **3**, 523 (2009)
Phys. Rev. Lett. **108**, 253003 (2012)



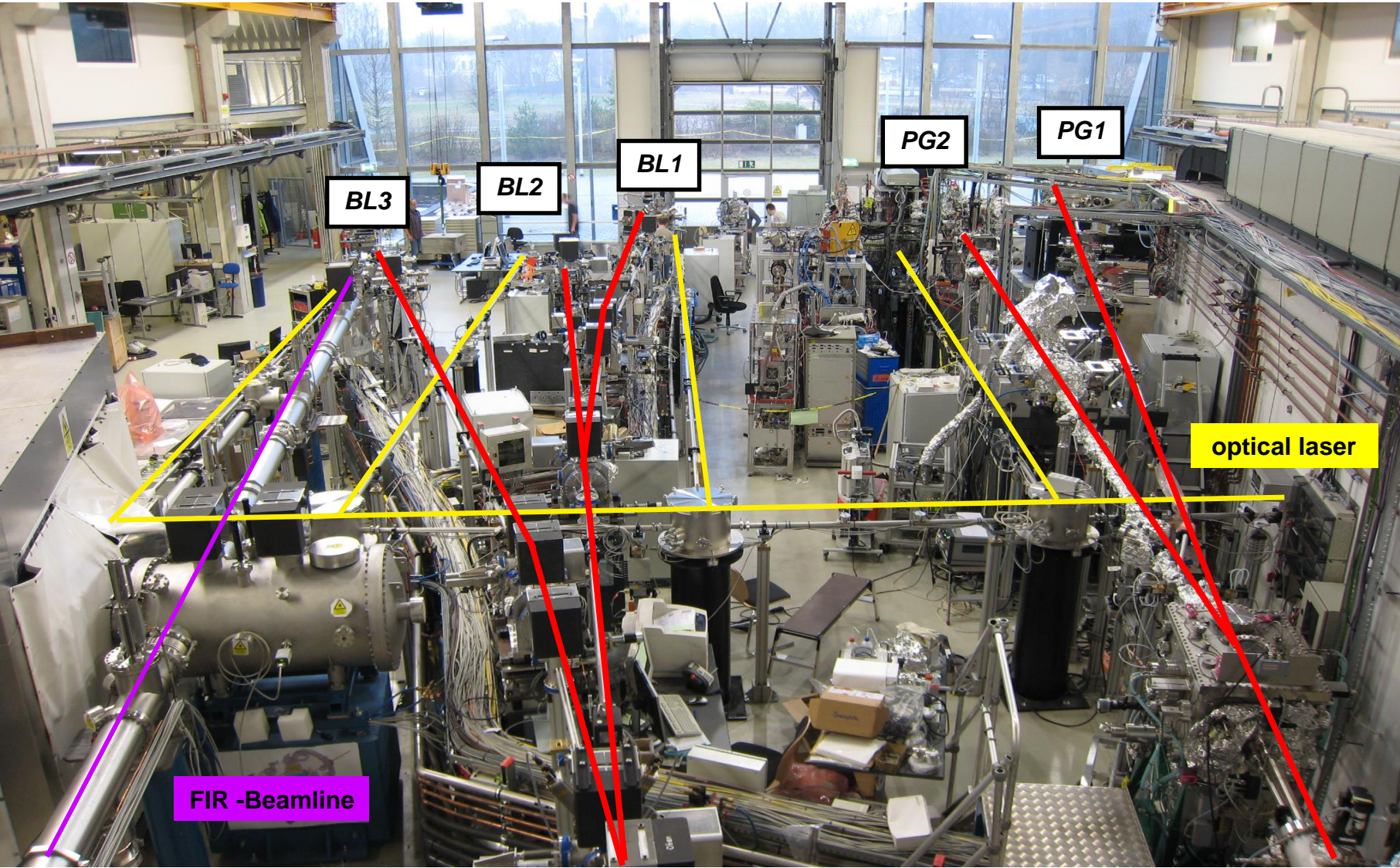
THz from undulator



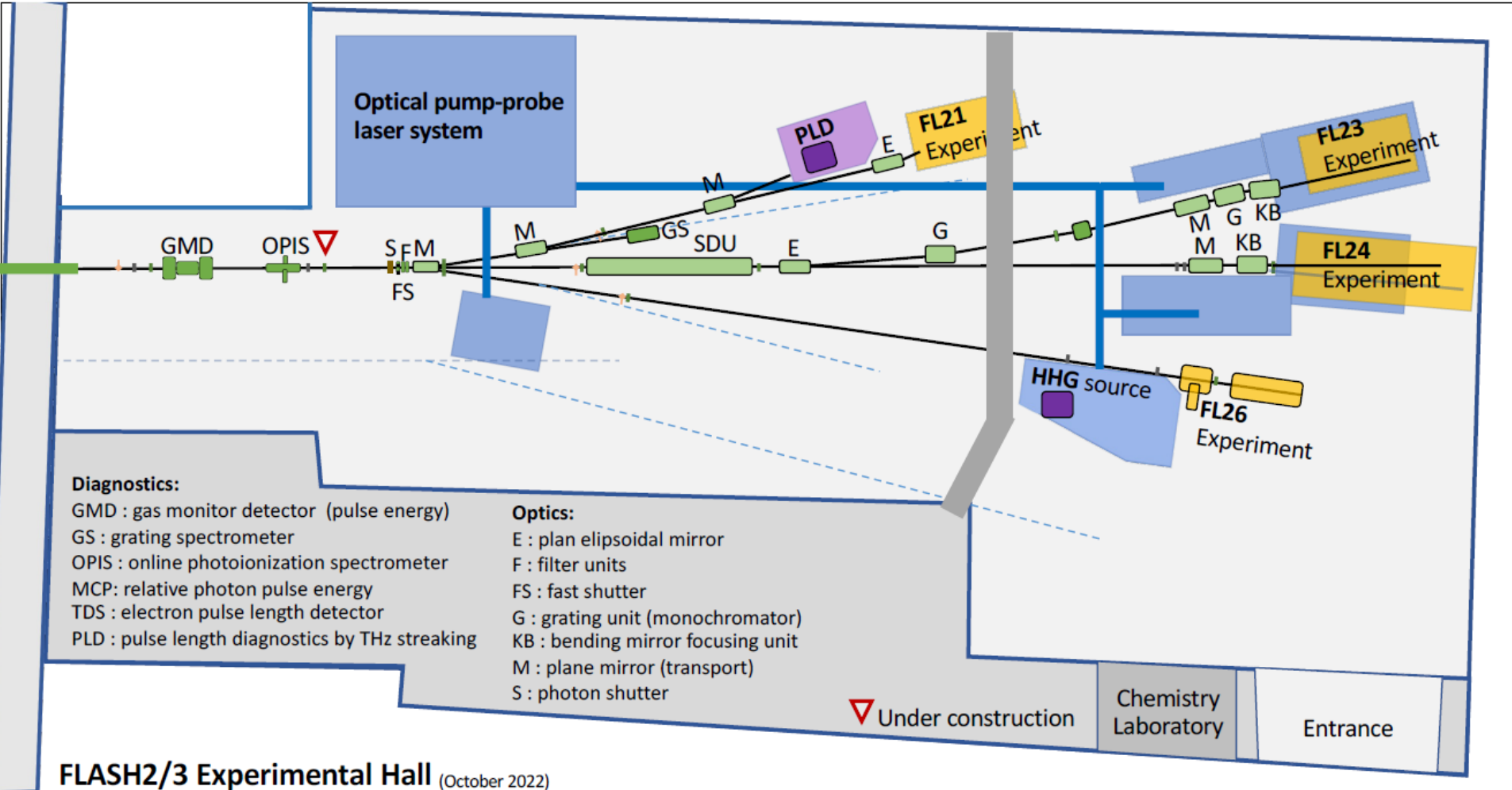
THz from dump magnet:



FLASH1 experimental hall – Albert-Einstein hall



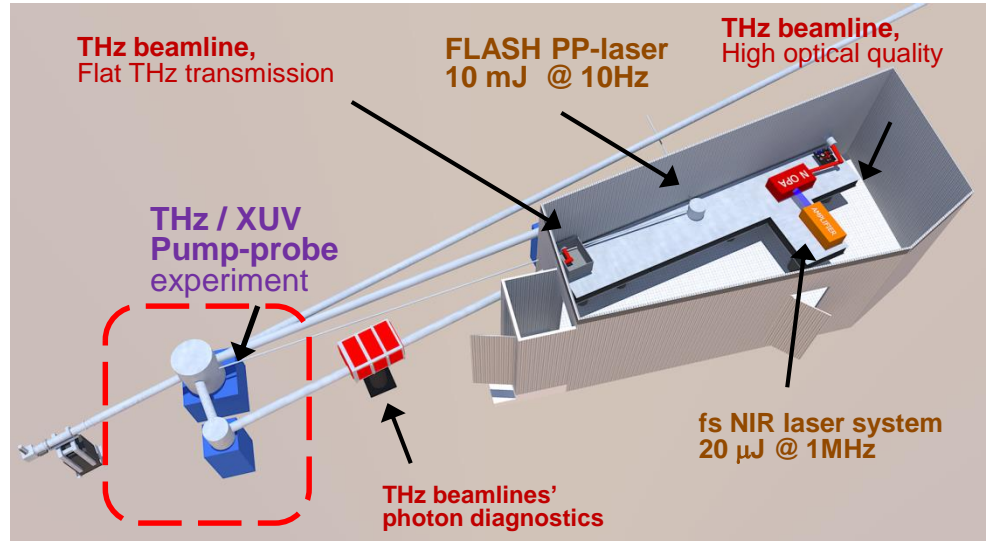
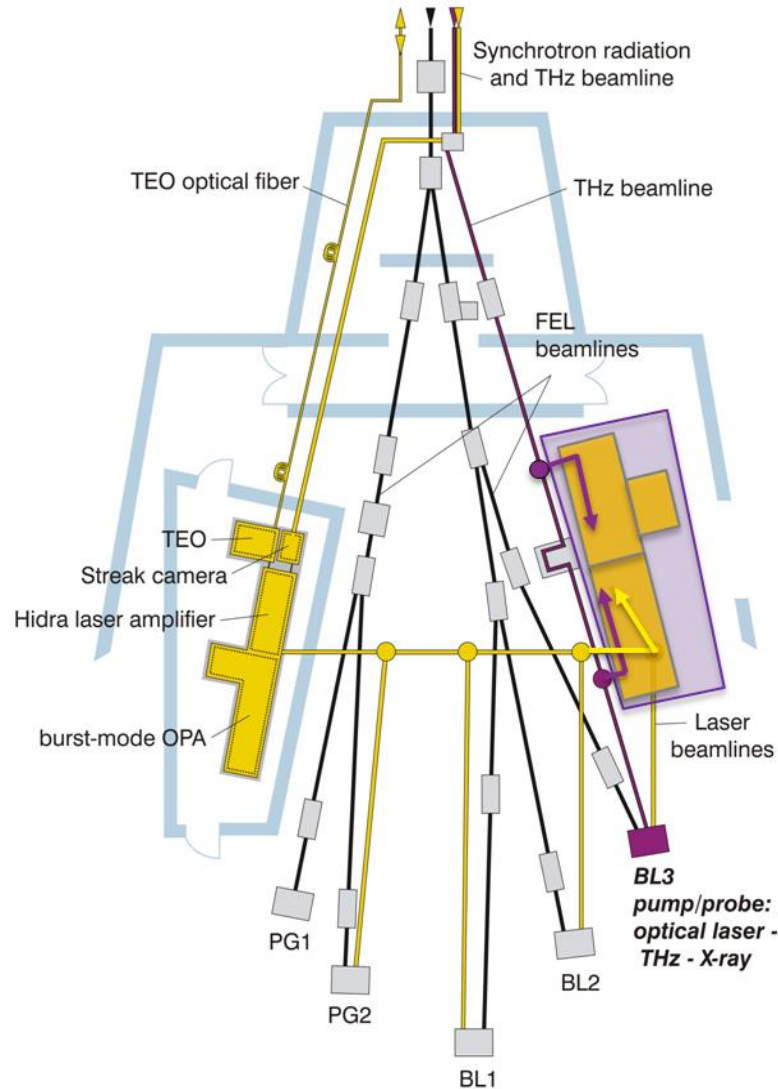
FLASH2 experimental hall – Kai Siegbahn hall



FLASH2/3 Experimental Hall (October 2022)

THz beamline at FLASH

THz laboratory and THz/XUV beamline



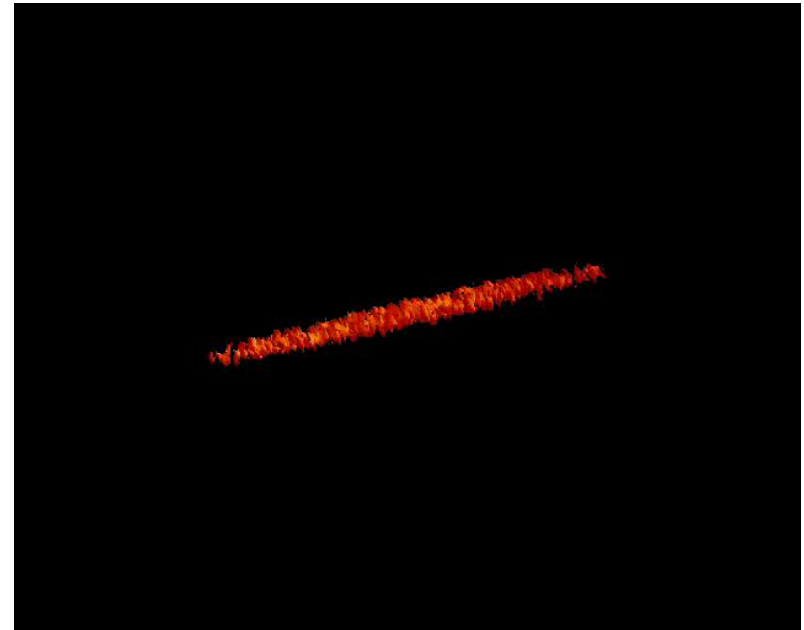
THz hutch laser (synchronized to FLASH):

- Wavelength: 1030 nm, 515 nm
- Rep. rate: 2 MHz
- Pulse energy: 100 μJ
- Avg. power: 20 W

Diagnostic tools for FELs.

What kind of diagnostic tools do user need to make efficient use of FELs?

- intensity
- beam position
- focus size
- spectral distribution
- temporal radiation pulse profile
- coherence
- polarization



Courtesy S. Reiche

**Due to the SASE specific shot-to-shot fluctuation the users need most of this information for every single pulse
=> online, non-destructive**

Light characterisation at FLASH - Introduction.

The *Atomic Photoionization Process* is a perfect candidate for non-destructive, pulse-resolved photon metrology tools.

- intensity Gas-Monitor Detectors (GMD)
- beam position GMD Split Electrodes
- focus size Wigner-Distribution Measurement
- spectral distribution Photoionization spectra
- temporal radiation pulse profile Non-linear autocorrelation or (THz or angular) streaking
- coherence Wigner-Distribution Measurement
- polarisation Angular photoemission distribution

The effort for developing such detector systems is extremely high, in particular due to the tight requirements on robustness and reliability.

Requirements for Intensity and Beam Position Detectors

- cover full dynamic range: ~ 6 - 7 orders of magnitude from spontaneous emission to SASE in saturation
- on-line pulse resolved detectors (non-destructive with respect to the beam)
- low degradation under radiant exposure by FEL beam with a peak power of few GW; high linearity
- ultra-high vacuum compatibility

No commercial detectors available!

Gas-monitor detectors for online intensity and beam position monitoring

Based on atomic photoionization =>
no degradation, indestructible

Low particle density =>
transparent

Calibrated in the PTB laboratory
Uncertainty for the pulse
energy: less than 10%

Photon beam

$10^{-6} - 10^{-4}$ mbar

electron signal
(position)

ion signal
(position)

ion signal
(intensity)

electron signal
(intensity)

PTB

Loeffe
Physico-
Technical
Institute

Reference number at the German Patent Office: 102 44 303

Equation behind the Gas-Monitor Detector

Number of particles detected (electrons or ions). Average photoionization charge needed to evaluate.

Quantum Efficiency

$$N_{\text{particle}} = N_{\text{photon}} \cdot \sigma(\hbar\omega) \cdot z \cdot \eta \cdot n = N_{\text{photon}} \cdot Q.E.(\hbar\omega)$$

Cross Section

Detection Efficiency

Atomic Gas Density (requires temperature and pressure info)

Detector Acceptance Length

Charge accumulated by the detector

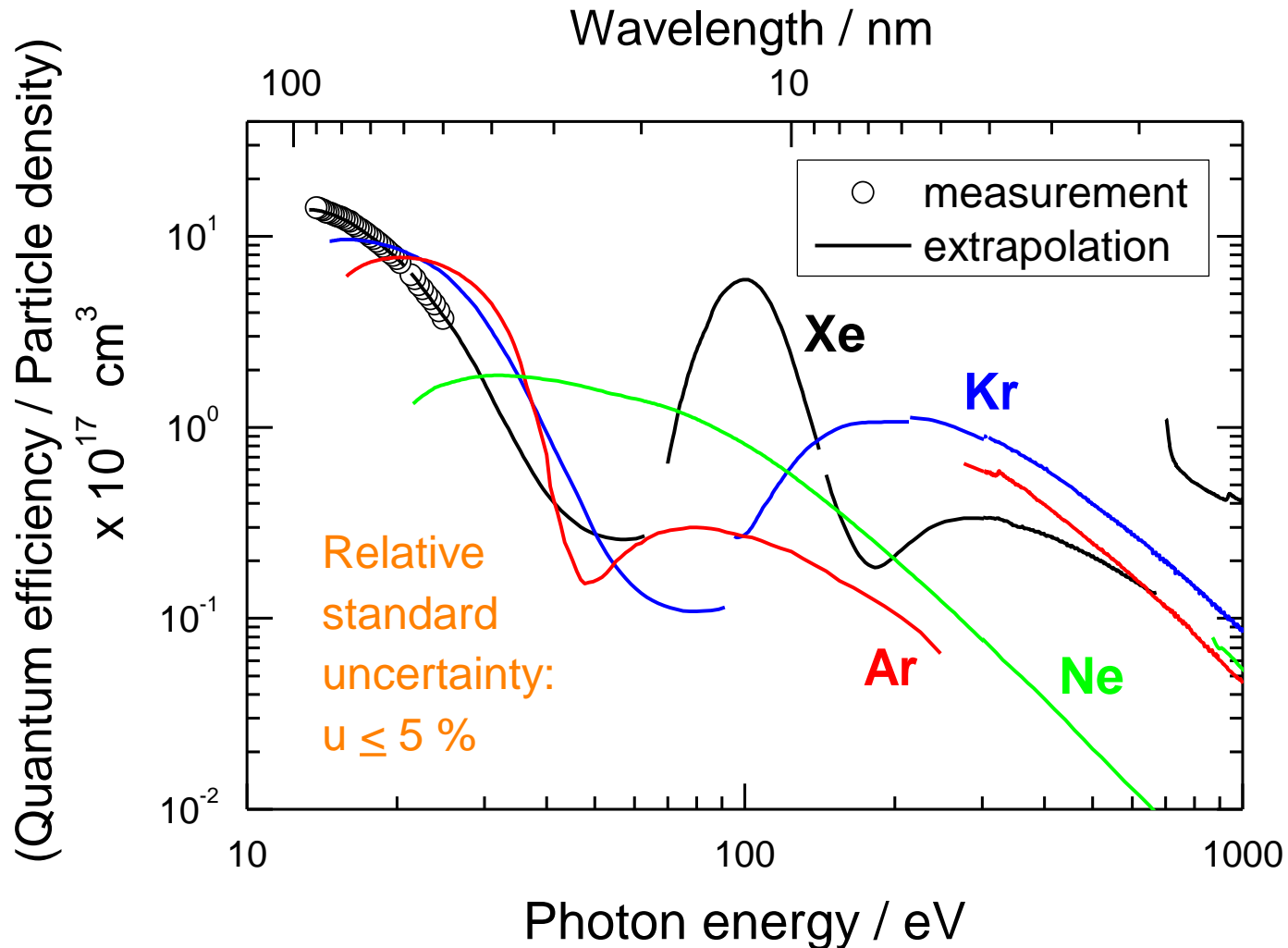
$$N_{\text{particle}} = \frac{Q}{e \cdot \gamma}$$

Elementary charge

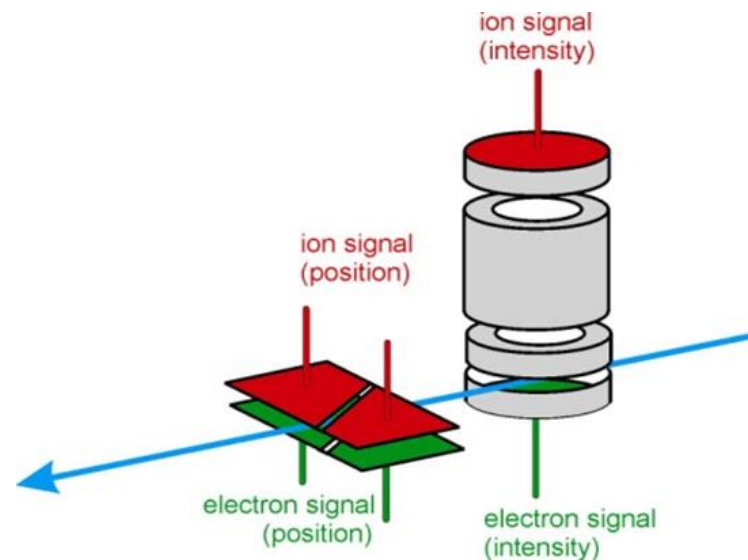
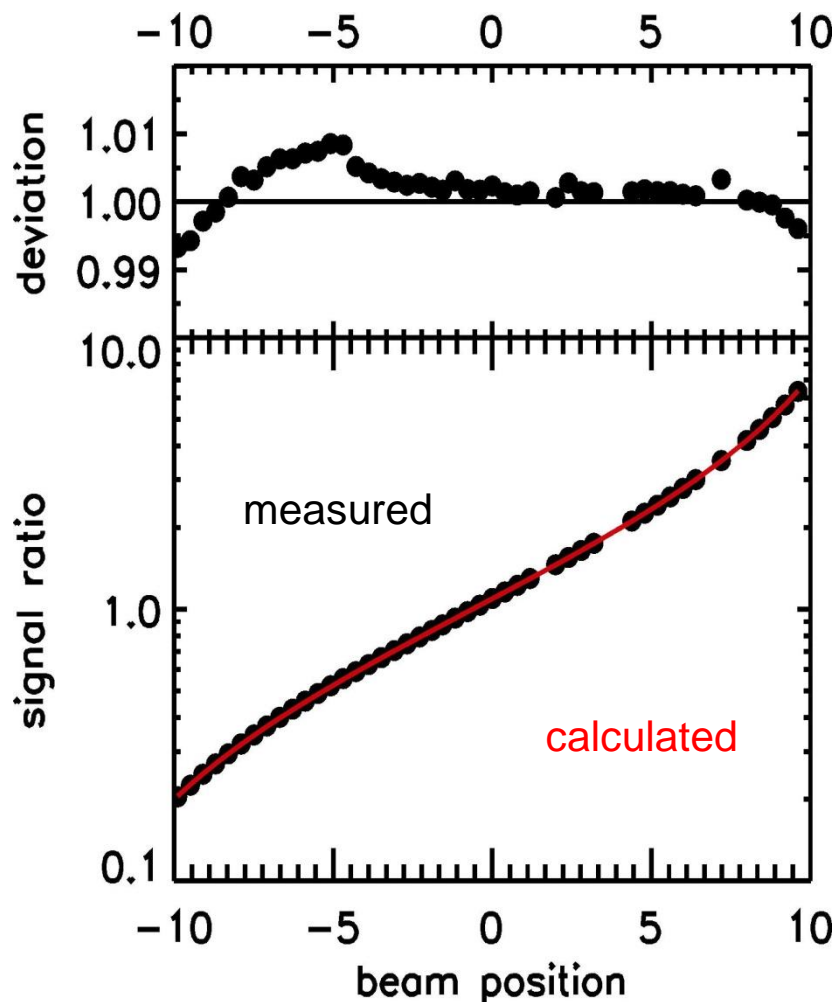
Mean ion charge

Quantum Efficiency of the FLASH GMD

calibrated in the PTB laboratory at BESSY II.



Beam position monitor



Accuracy for on-line measurements of relative beam positions: $\sim 20 \mu\text{m}$

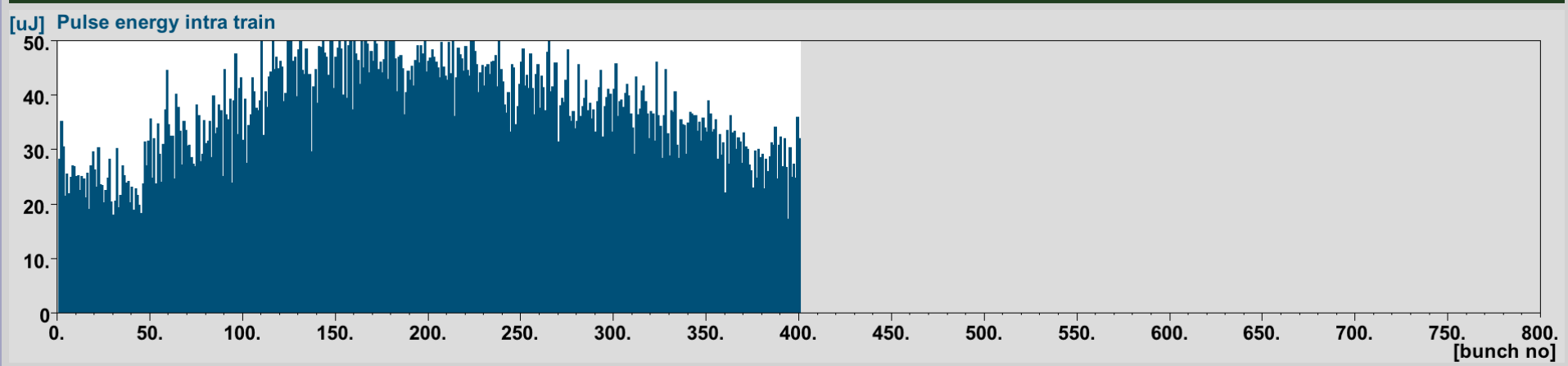
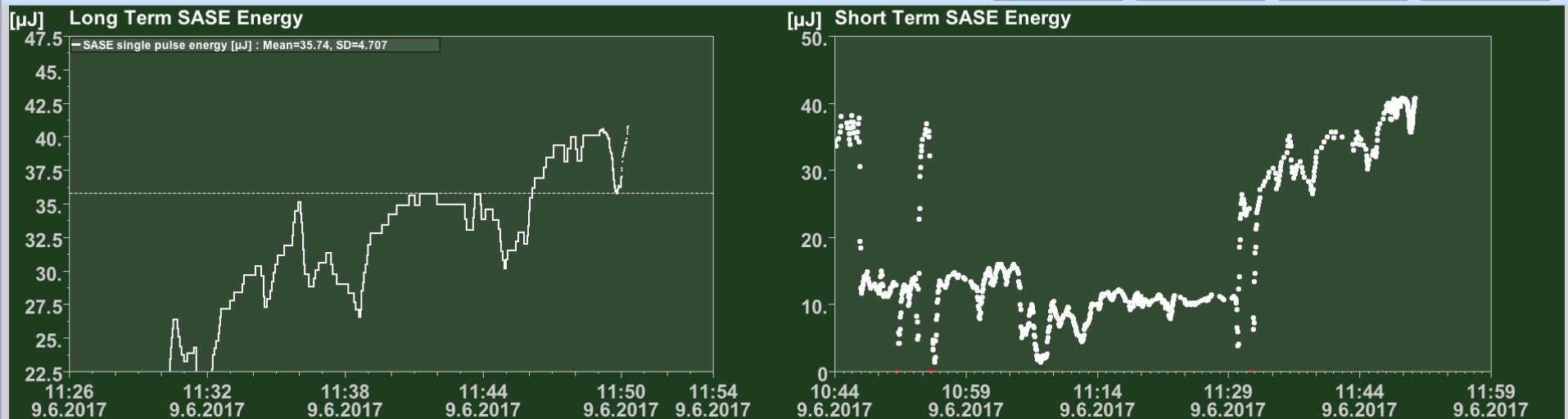
Two gas-monitor detector sets, which are 20 m apart, allow on-line monitoring of the angle: $\sim 1 \mu\text{rad}$

In collaboration with PTB in Berlin and IOFFE institute St. Petersburg

Example performance – FLASH 1

/svn/FLASH/diag/SASE/FLASH_DOCS_SASE_Viewer.xml TTF2.FEL//

FLASH. FLASH 1 - SASE viewer 1020.4 MeV GMD-Tunnel 3.00 mm / 3.00 mm 40.72 μJ +/- 9% 6.10 nm
163.23 mW



Example performance – FLASH 2

/svn/FLASH/diag/SASE/FLASH2_DOOCS_SASE_Viewer.xml FLASH.FEL/XGM.PHOTONFLUX/FL2_TUNNEL/*



FLASH 2 - SASE viewer

FL2_TUNNEL

680.67 MeV

GMD ok? ●

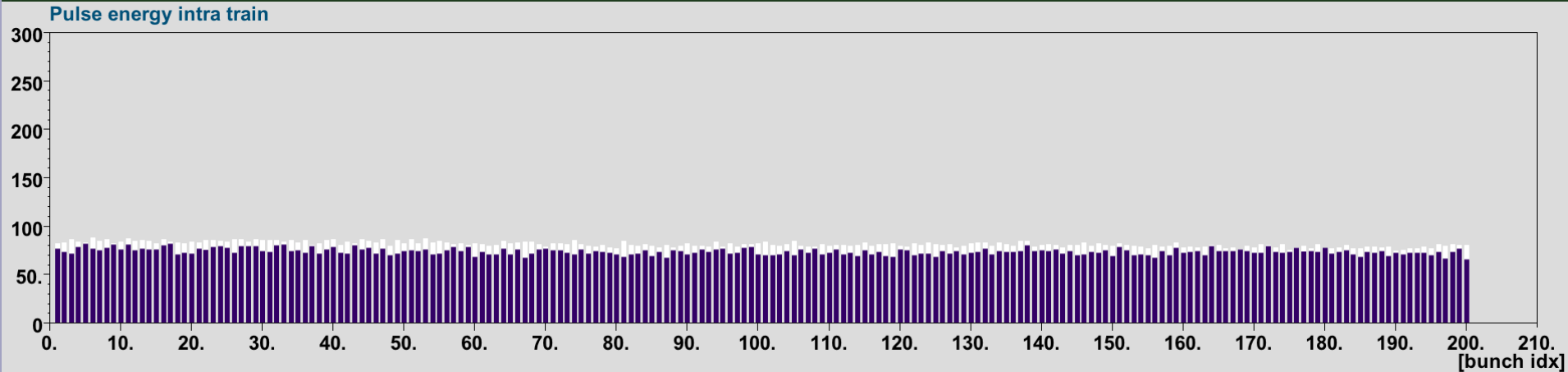
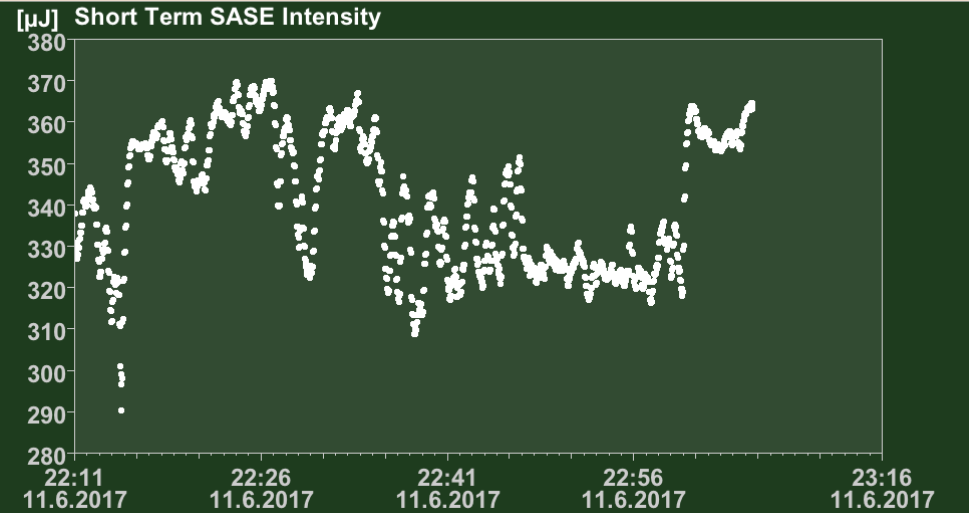
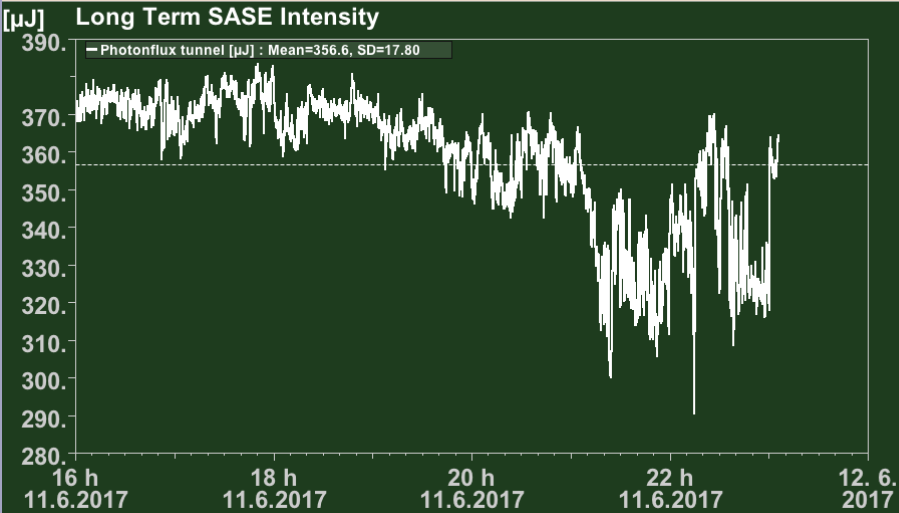
Apertures
10 mm / 10 mm

363.25 μ J @ 25.90 nm

726.49 mW

Print

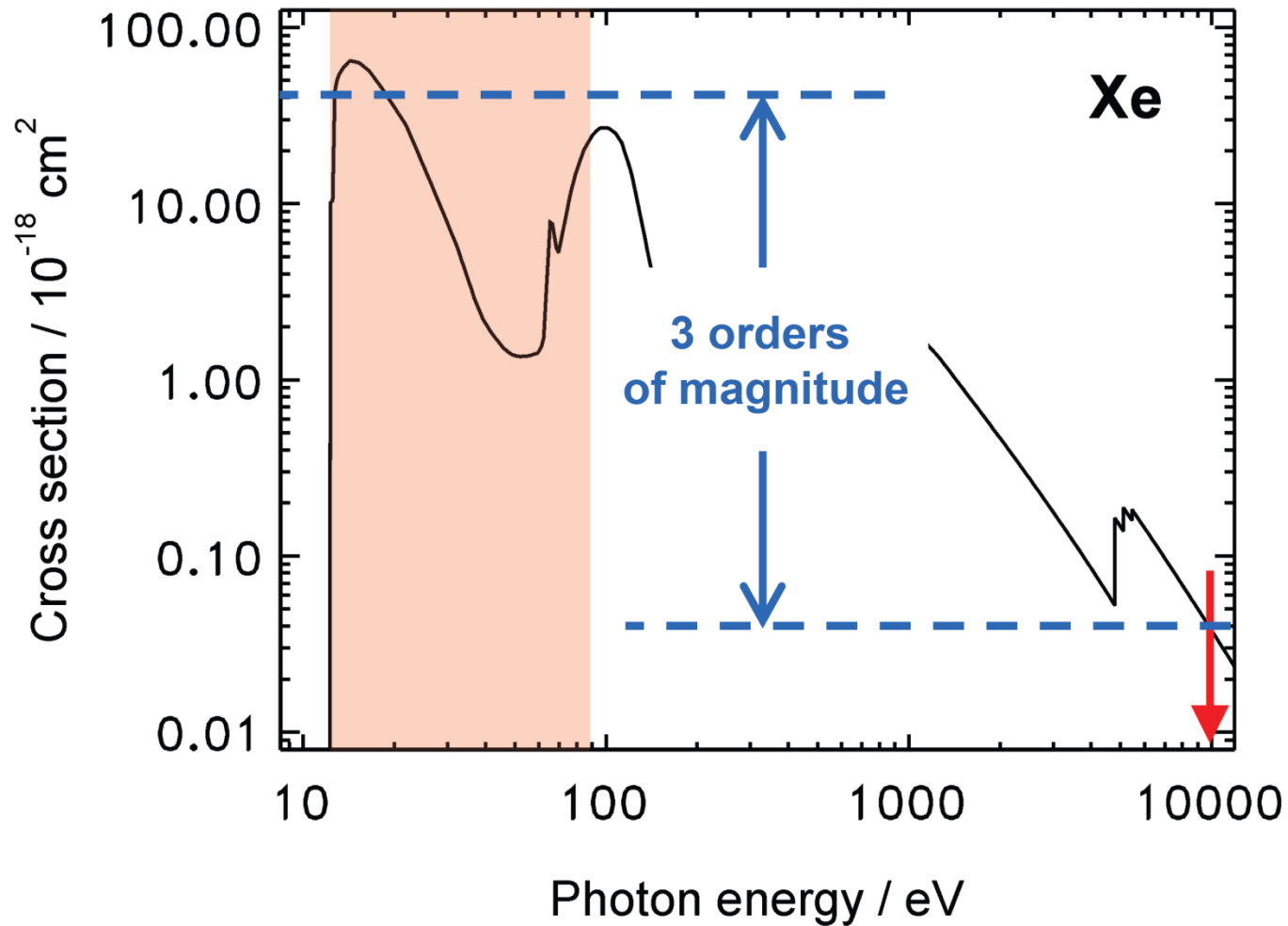
photon-diag



Specification of the intensity measurements for European XFEL

- Photon energy range: up to 12.4 keV (TDR) **(presently >24 keV)**
- Number of pulses per second: 30000
- Time resolution: < 200 ns
- Uncertainty for the pulse energy: <10 %
- Relative uncertainty (pulse to pulse): < 1 % (for more than 10^{10} photon per pulse)
- Operating pressure: 10^{-6} mbar – 10^{-4} mbar

Photoabsorption cross sections of Xenon



B.L. Henke et al., Atomic Data and Nuclear Data Tables **54**, 181-342 (1993).

3rd generation GMDs for European XFEL

- Measured uncertainty due to statistical nature of photoionization:

$$\delta = \frac{\sqrt{N_{\text{ion}}}}{N_{\text{ion}}} = \frac{1}{\sqrt{N_{\text{ion}}}} \quad \Rightarrow \quad \delta = 1 \% \quad \text{if } N_{\text{ion}} = 10^4 \text{ ions generated per pulse}$$

- What the detector size should be?

Photon energy: 12.4 keV

Target gas Xe: $\sigma = 0.021 \text{ Mb}$ ($q \approx 8$)

Pressure: $p = 10^{-4} \text{ mbar}$ ($n_{\text{atom}} = 2.4 \times 10^{12} \text{ cm}^{-3}$)

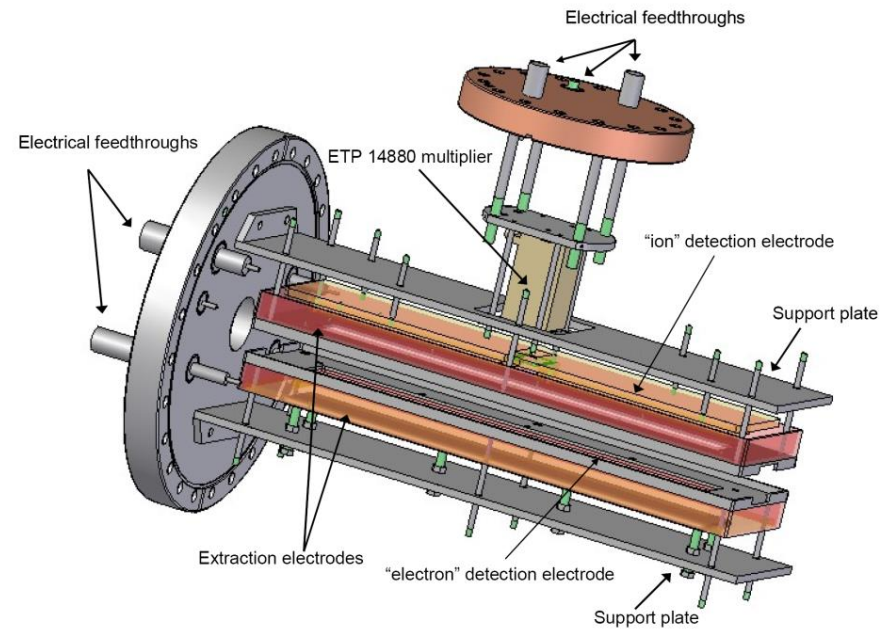
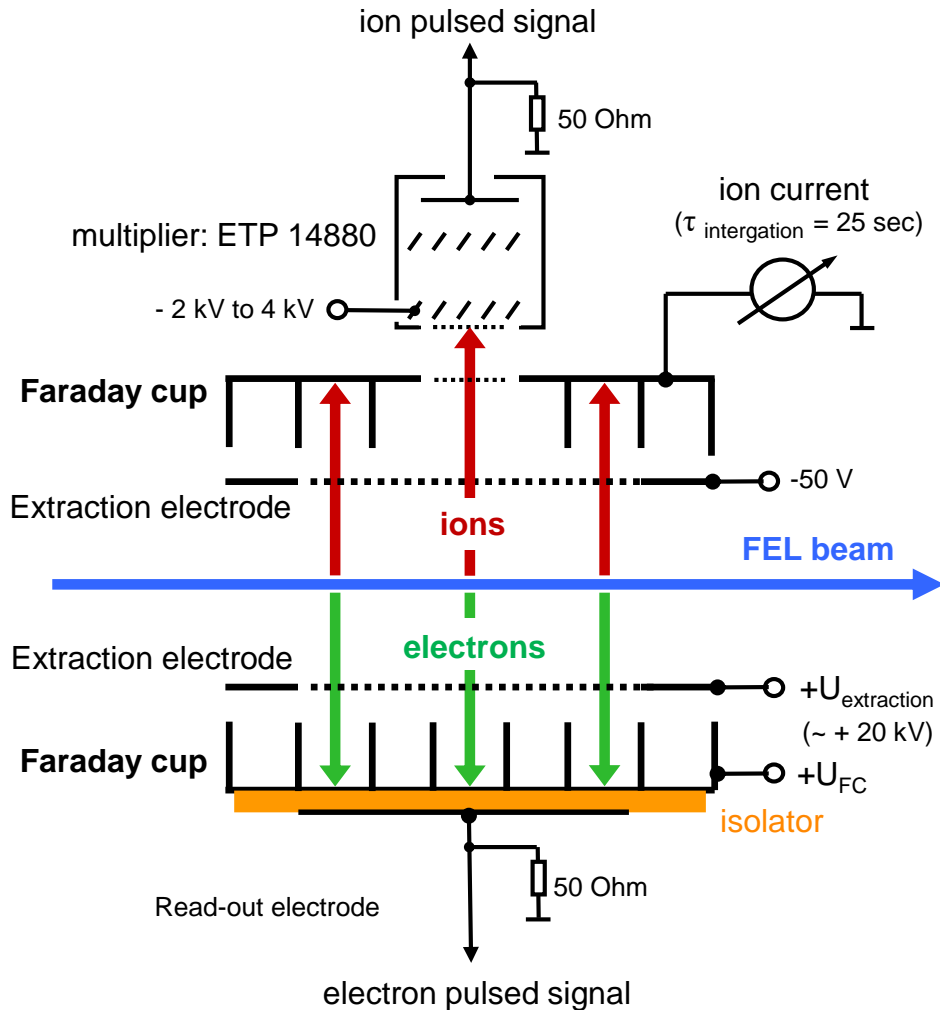
Number of photons per pulse: $N_{\text{photon}} = 10^{10}$

Effective length z :

$$z = \frac{N_{\text{ion}}}{N_{\text{photon}} \cdot \sigma_{\text{ph}}(\hbar\omega) \cdot n_{\text{atom}}}$$

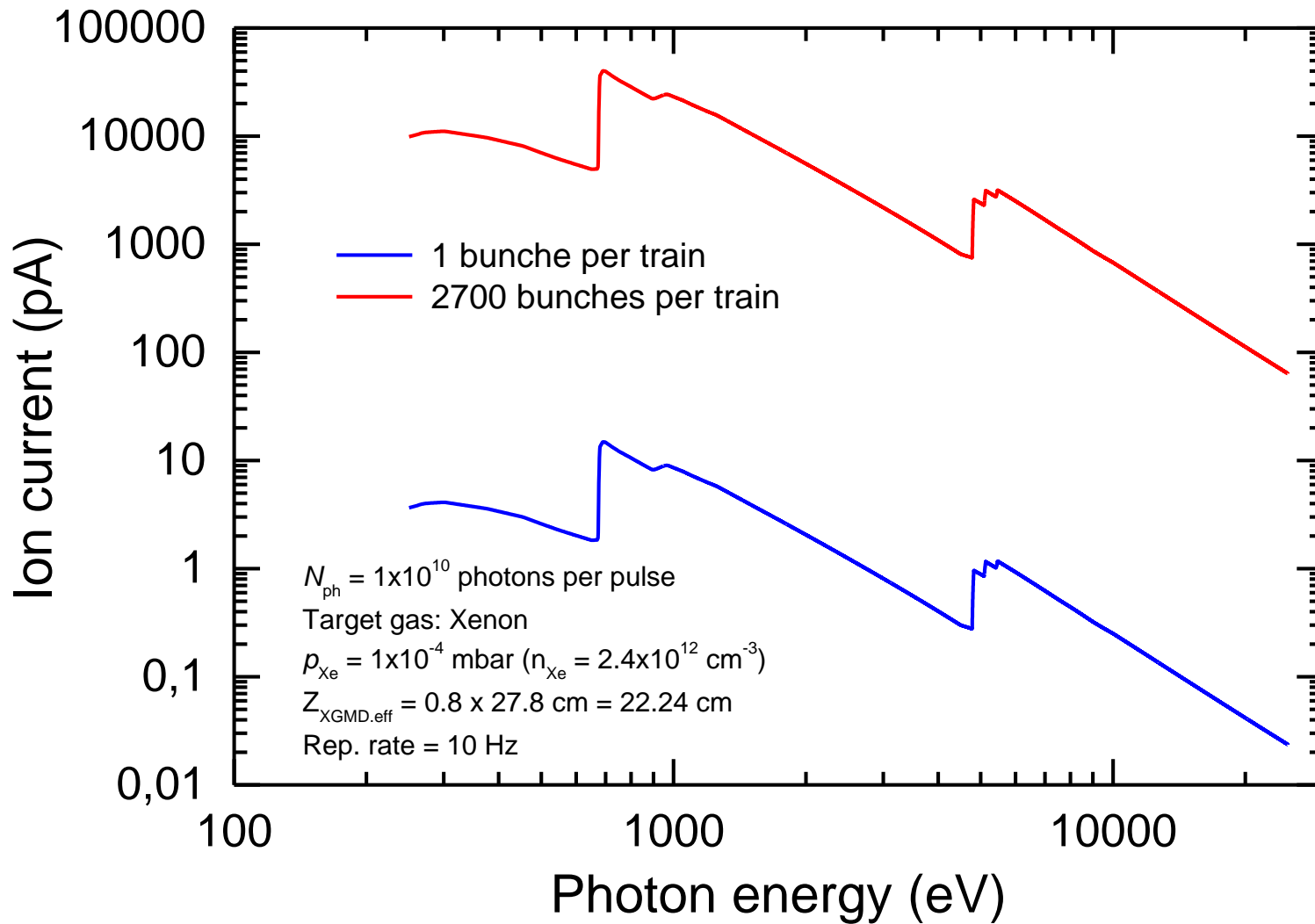
Minimum length : $Z = 20 \text{ cm} !!!$

3rd generation GMD for European XFEL

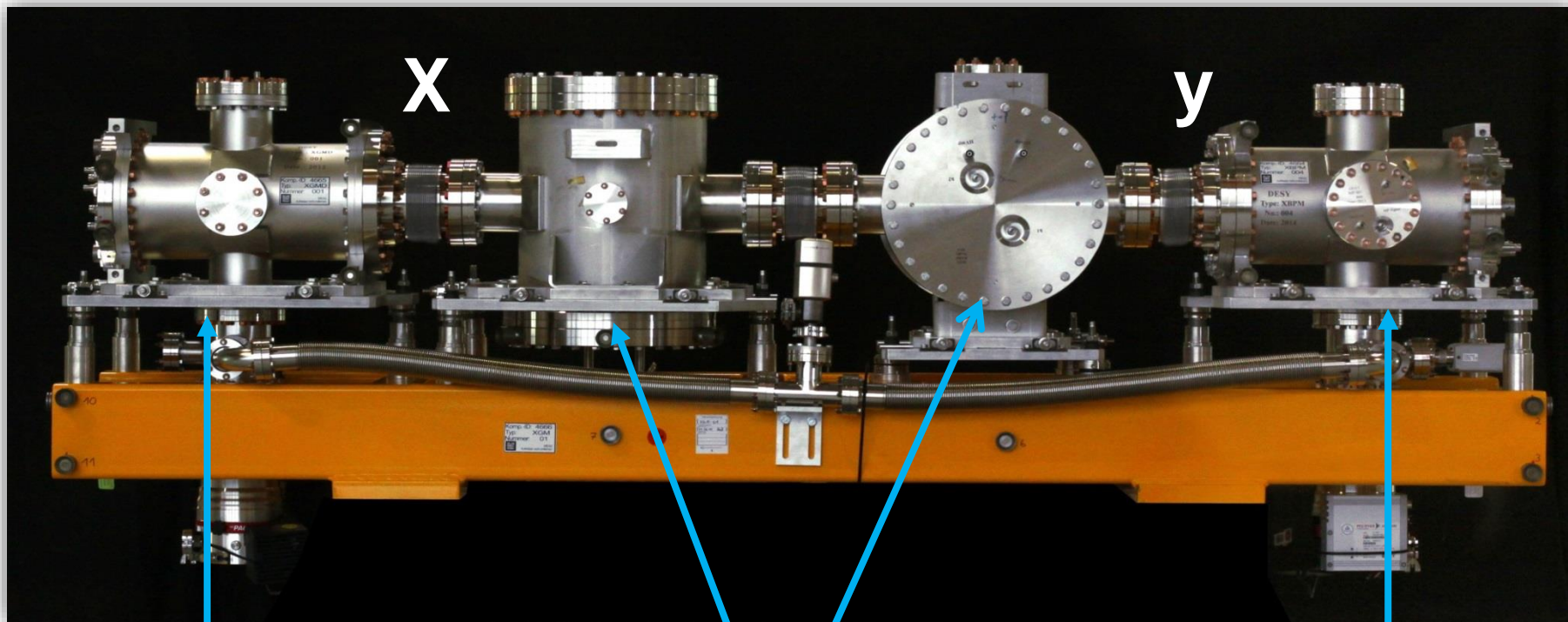


High extraction voltage of up to 20 kV – 30 kV has to be applied to prevent detection of highly energetic photoelectrons by the ion detector.

XGMD signal: average ion current from Faraday



XGM for European XFEL and SwissFEL: Intensity and beam position with an extended dynamic range.



XGMD with split Faraday cup detection electrodes (horizontal beam position)

Two HAMPs with huge area open electron multipliers each equipped with a pair of split anodes (horizontal X and vertical Y beam position)

XGMD with split Faraday cup detection electrodes (vertical beam position)

Example – European XFEL – SASE 1,2,3



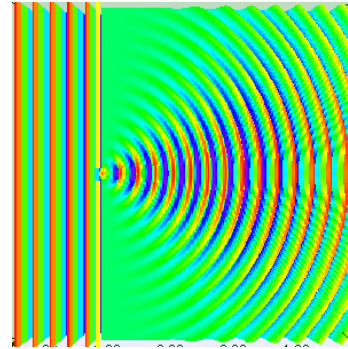
Spectral distribution

Principle of a diffraction grating

- > Light is a wave -> diffraction occurs
- > Huygens–Fresnel principle
-> principle of superposition of waves
- > Grating equation

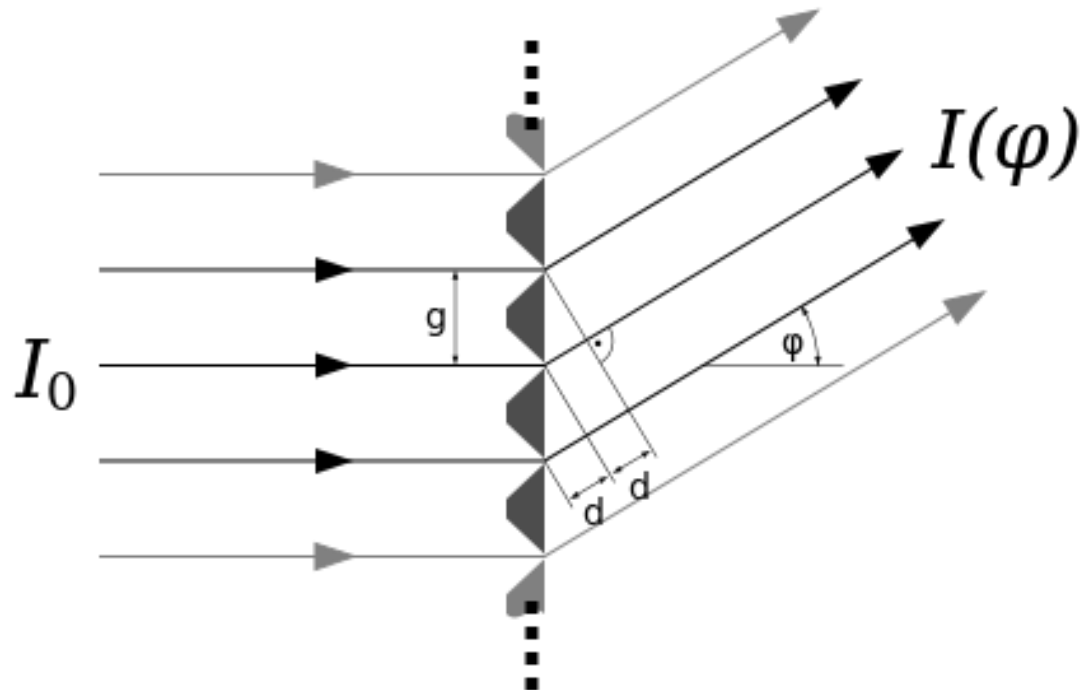
$$d_m = g \cdot \sin \varphi_m = m \cdot \lambda, \quad m \in \mathbb{N}$$

- > Angle of diffracted light is wavelength dependent!

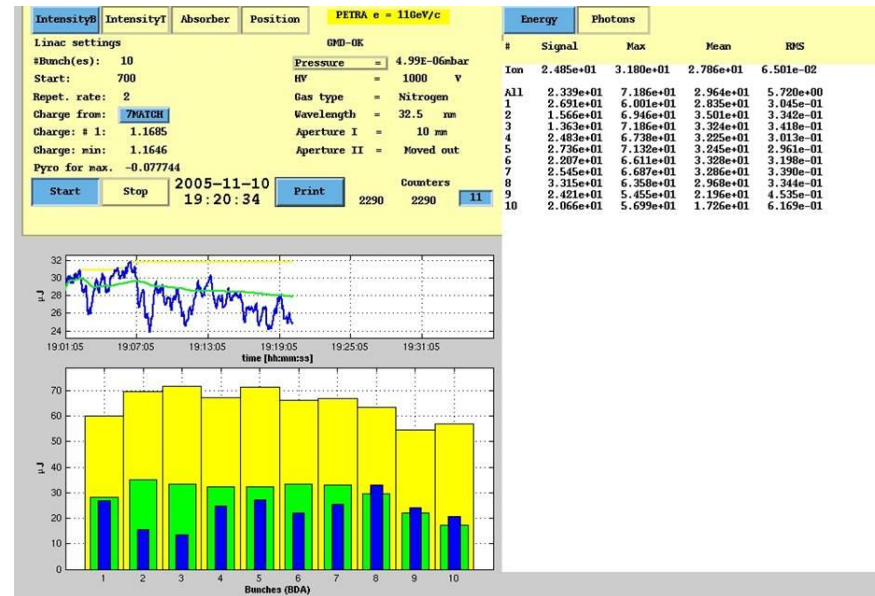
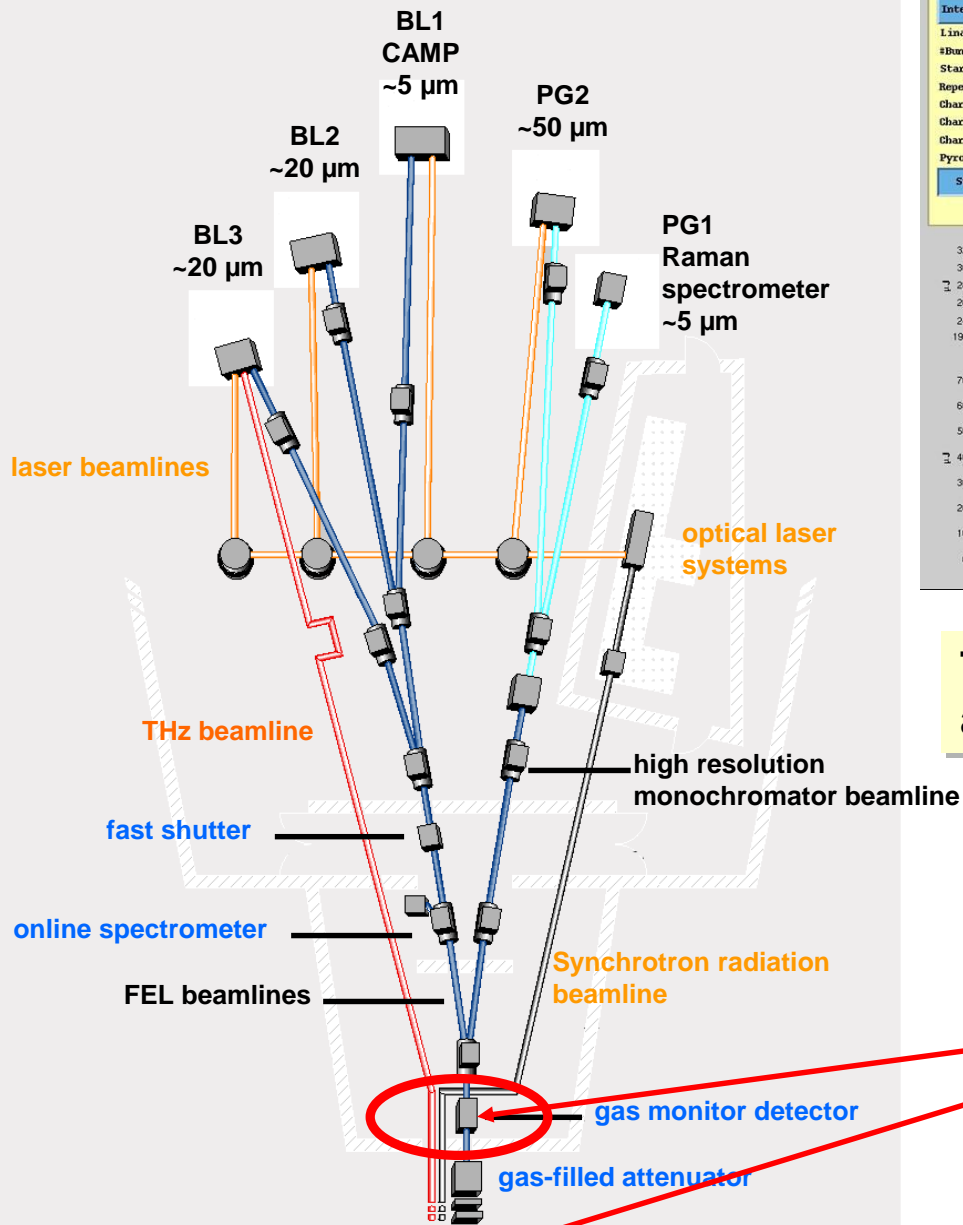


Example: num.
approx.

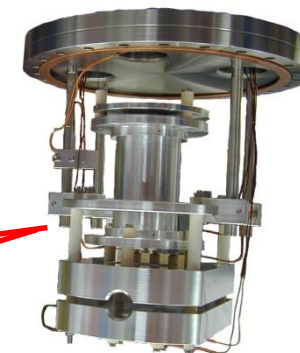
slit width =
wavelength



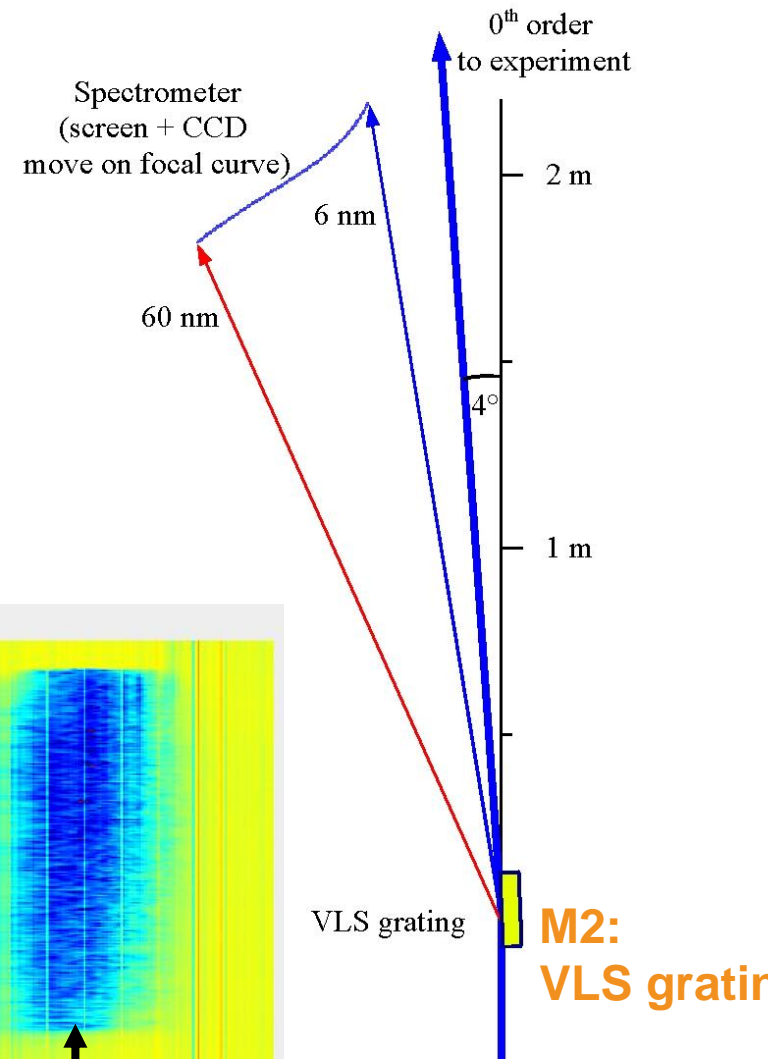
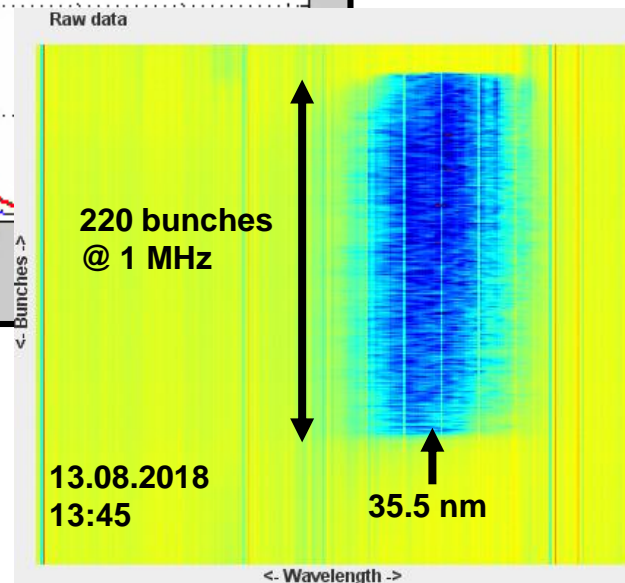
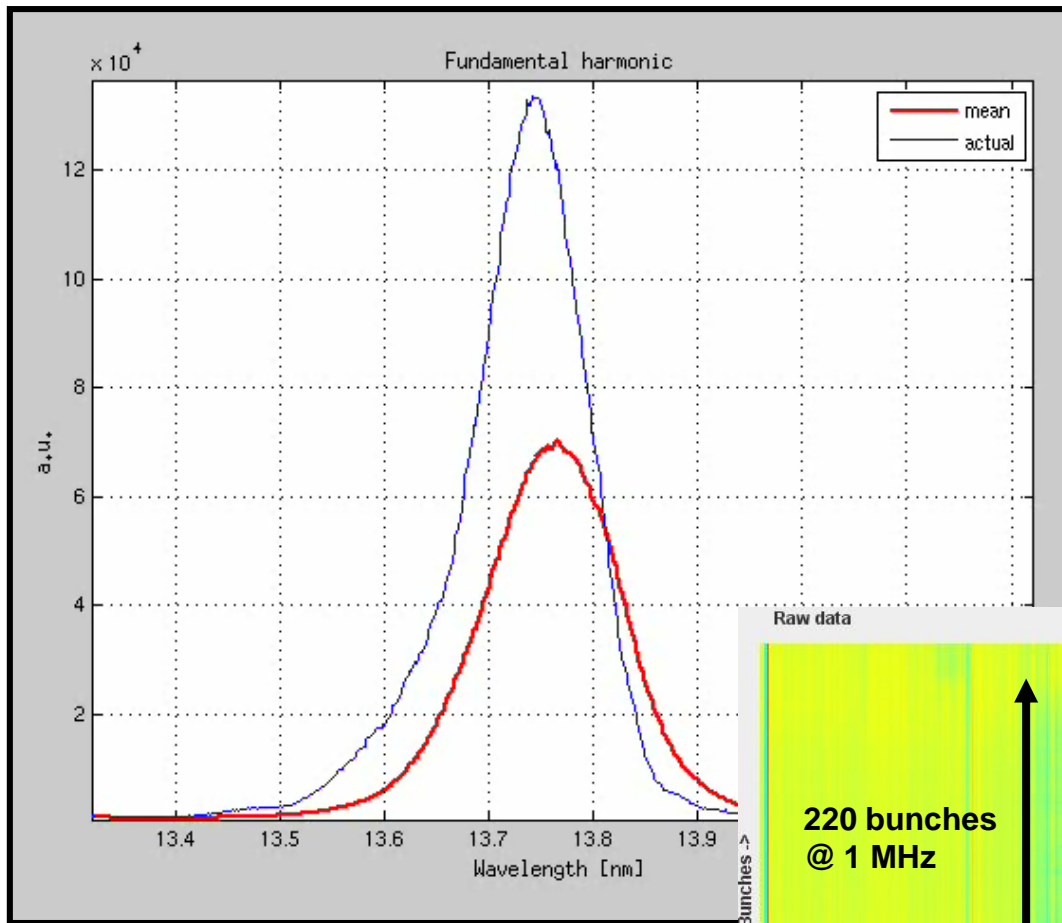
Light transportation – FLASH1 beamlines



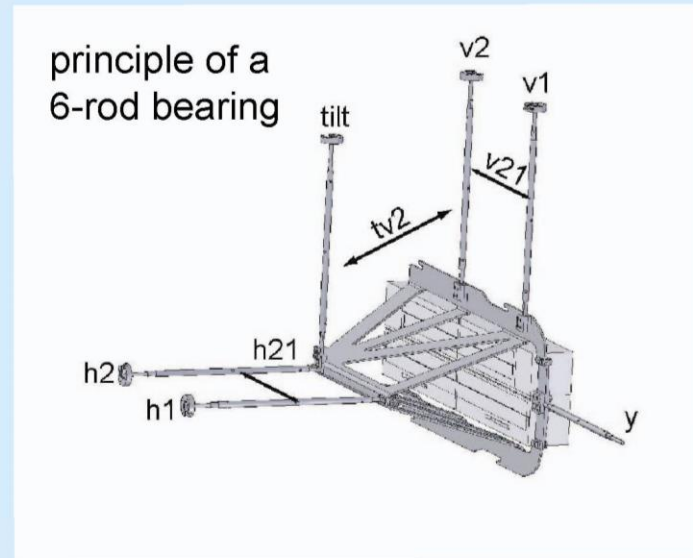
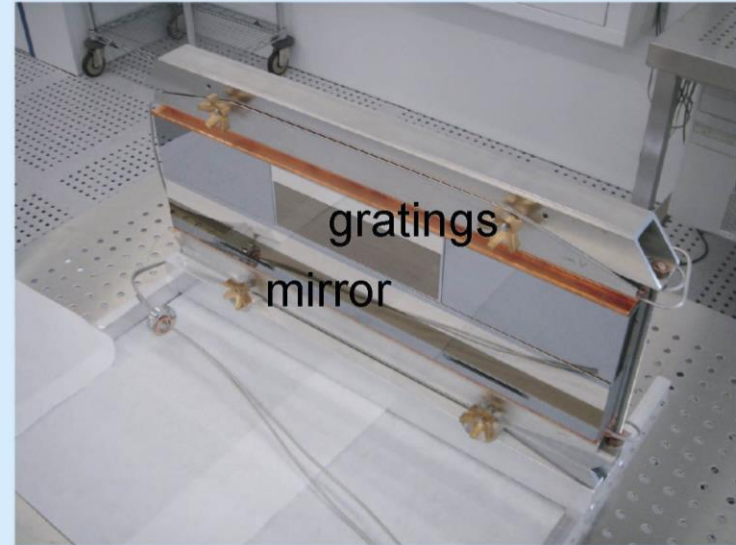
Two gas monitor detector sets: before and behind the gas attenuator



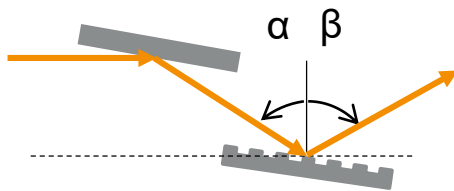
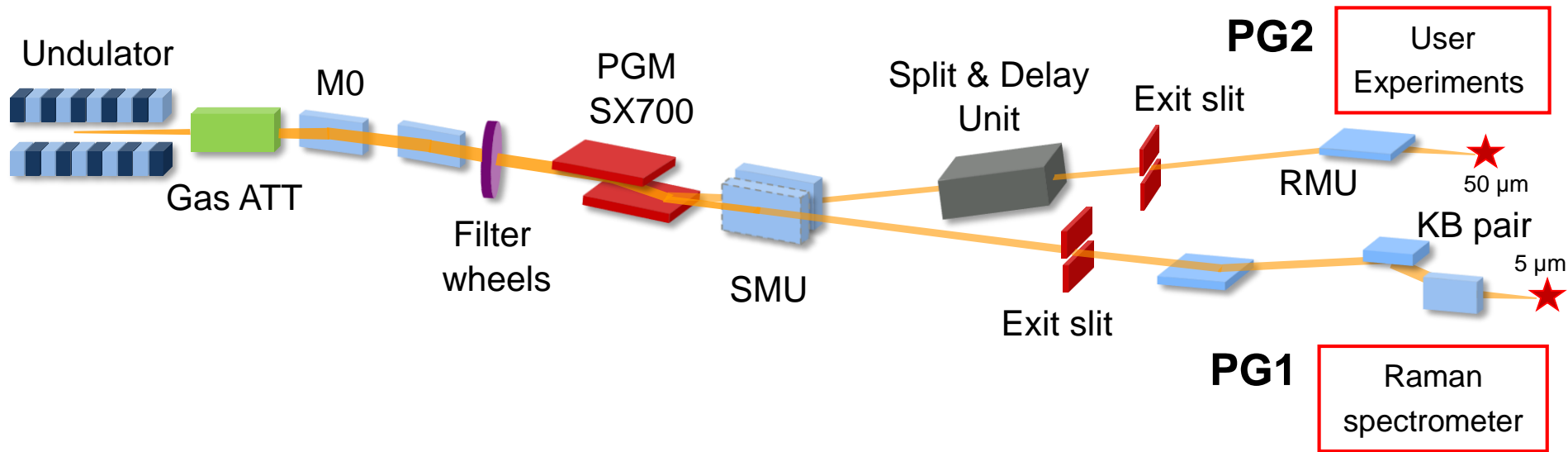
VLS online Spectrometer



VLS online spectrometer for single pulses



Plane Grating (PG) Monochromator Beamline



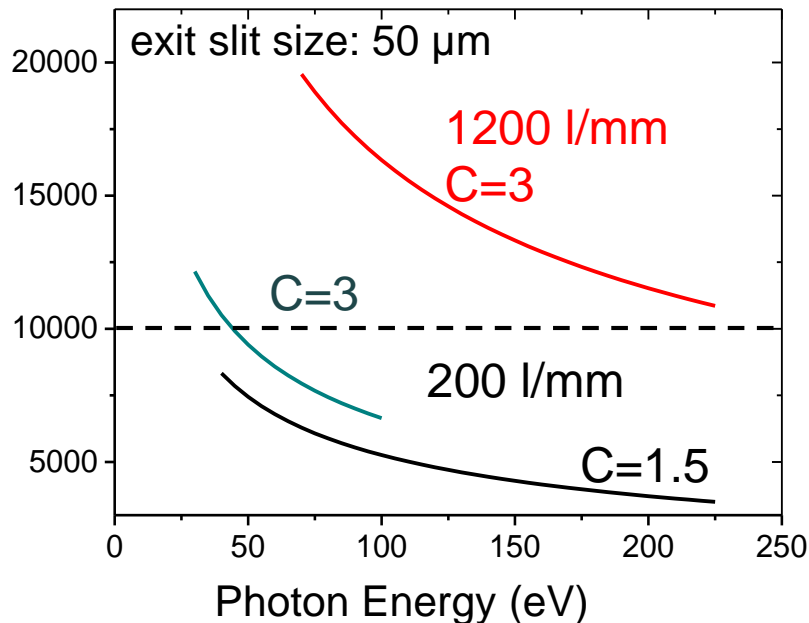
$$c_{ff} = \frac{\cos \beta}{\cos \alpha}$$

- 2 beamline branches PG1 & PG2
- Slitless operation
- High flux mode/high resolution mode
- 0th order operation
- PG2 spectrometer mode available

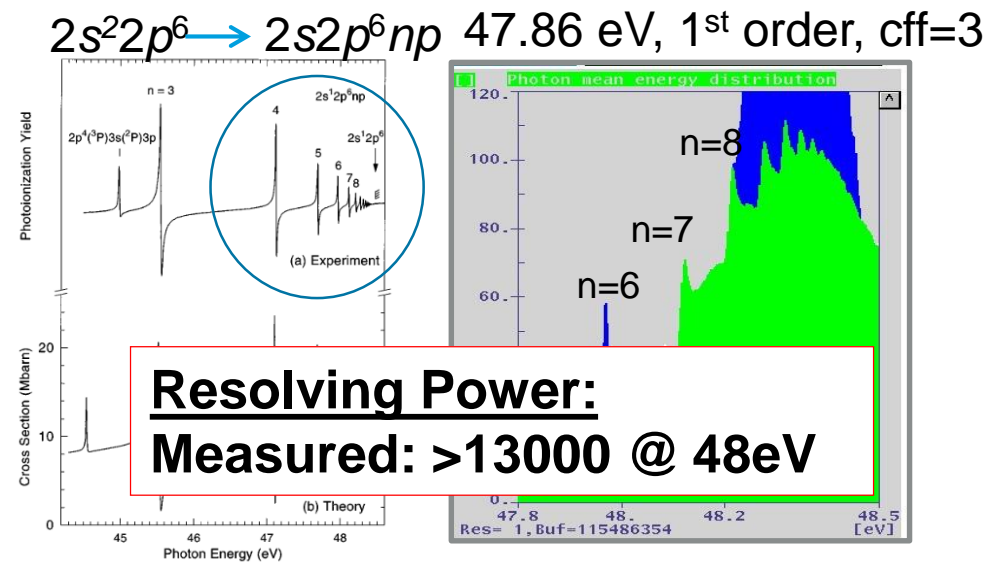
PG Monochromator Beamline - Performance

- Photon energy range: 20...250 eV (fund.) (...600eV 3rd harm.)
- Resolving power : $>10^4$
- Photon Flux: $10^9...10^{12}$ photons/pulse
- Spot size at sample: 40 x 100 μm (h x v)

Resolving Power



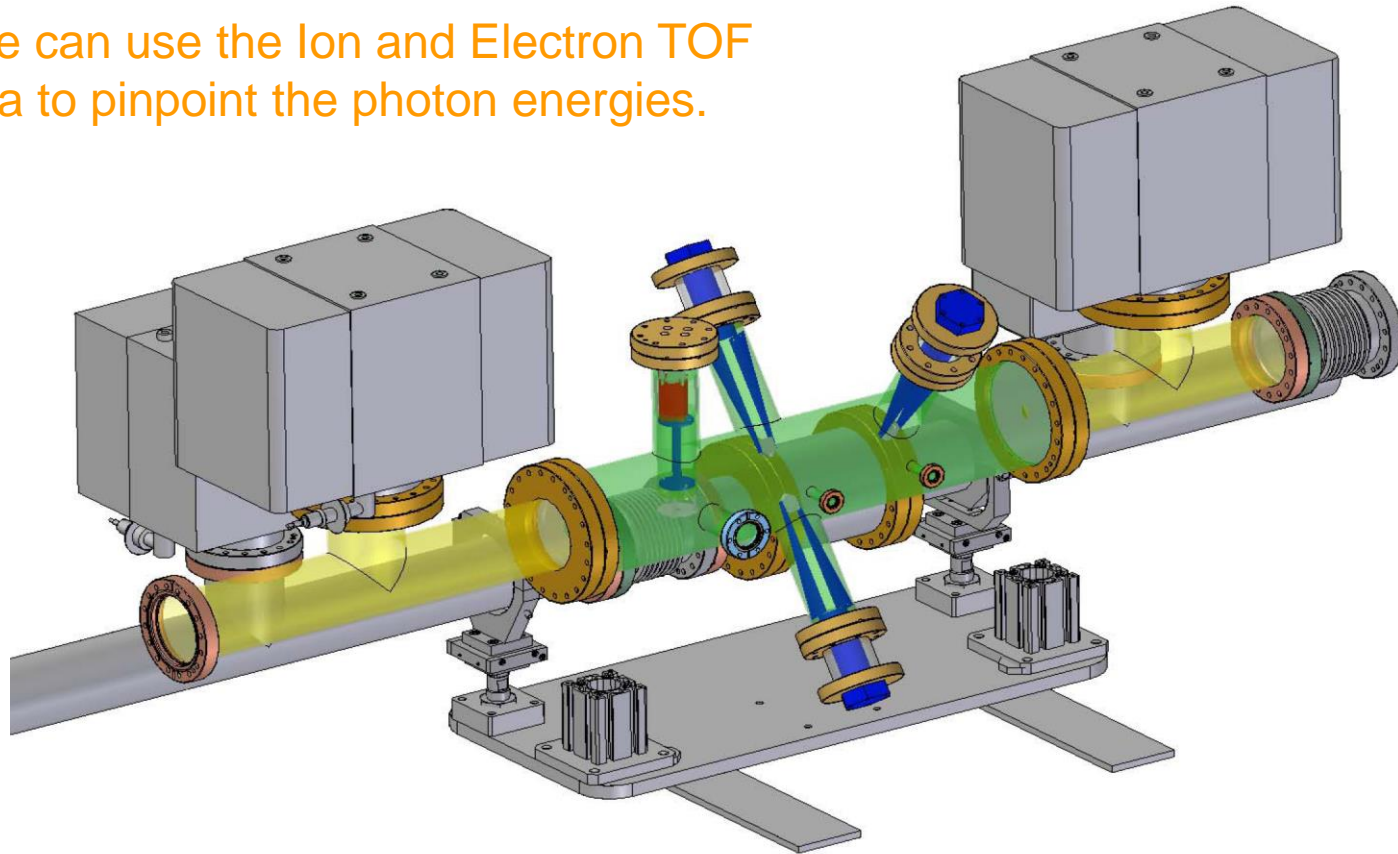
Calibration: using Ne gas absorption lines



PG in spectrometer mode

Online determination of the spectral distribution using ion and electron TOF spectrometer

One can use the Ion and Electron TOF data to pinpoint the photon energies.



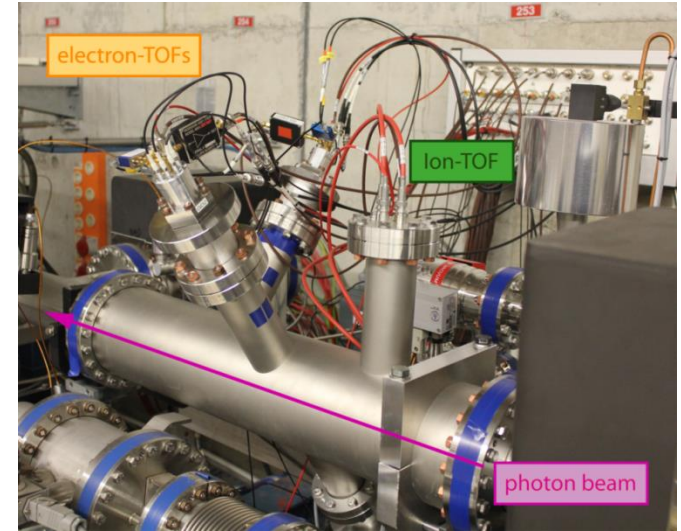
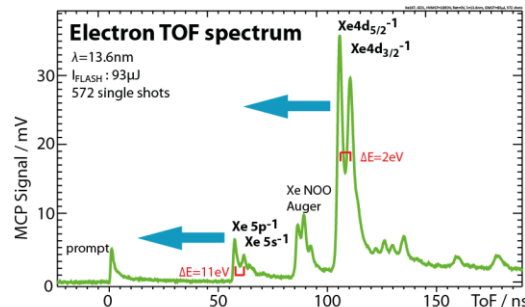
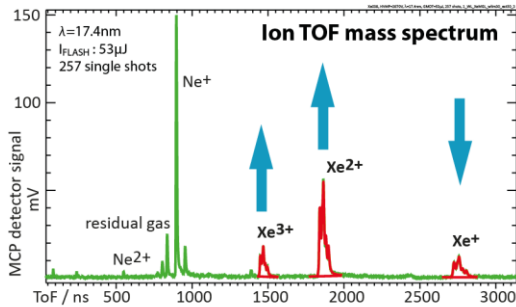
M. Wellhöfer, J. T. Hoeft, M. Martins, W. Wurth, M. Braune, J. Viefhaus, K. Tiedtke, M. Richter,
Photoelectron spectroscopy as a non-invasive method to monitor SASE-FEL spectra. JINST 3, P02003 (2008)

Cross-calibration campaign of OPIS using PG2

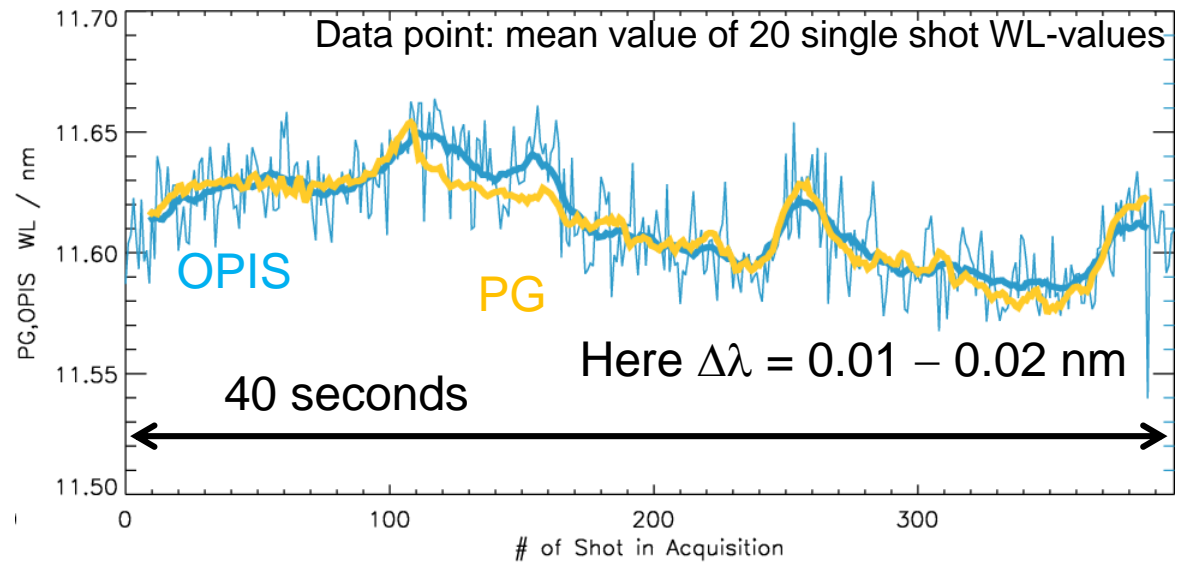
OPIS - Online Photoionization Spectrometer:

- installed in FLASH1 tunnel
- Online wavelength monitoring

Using Ion and Electron TOF to determine the FEL wavelength



FEL wavelength (moving average)

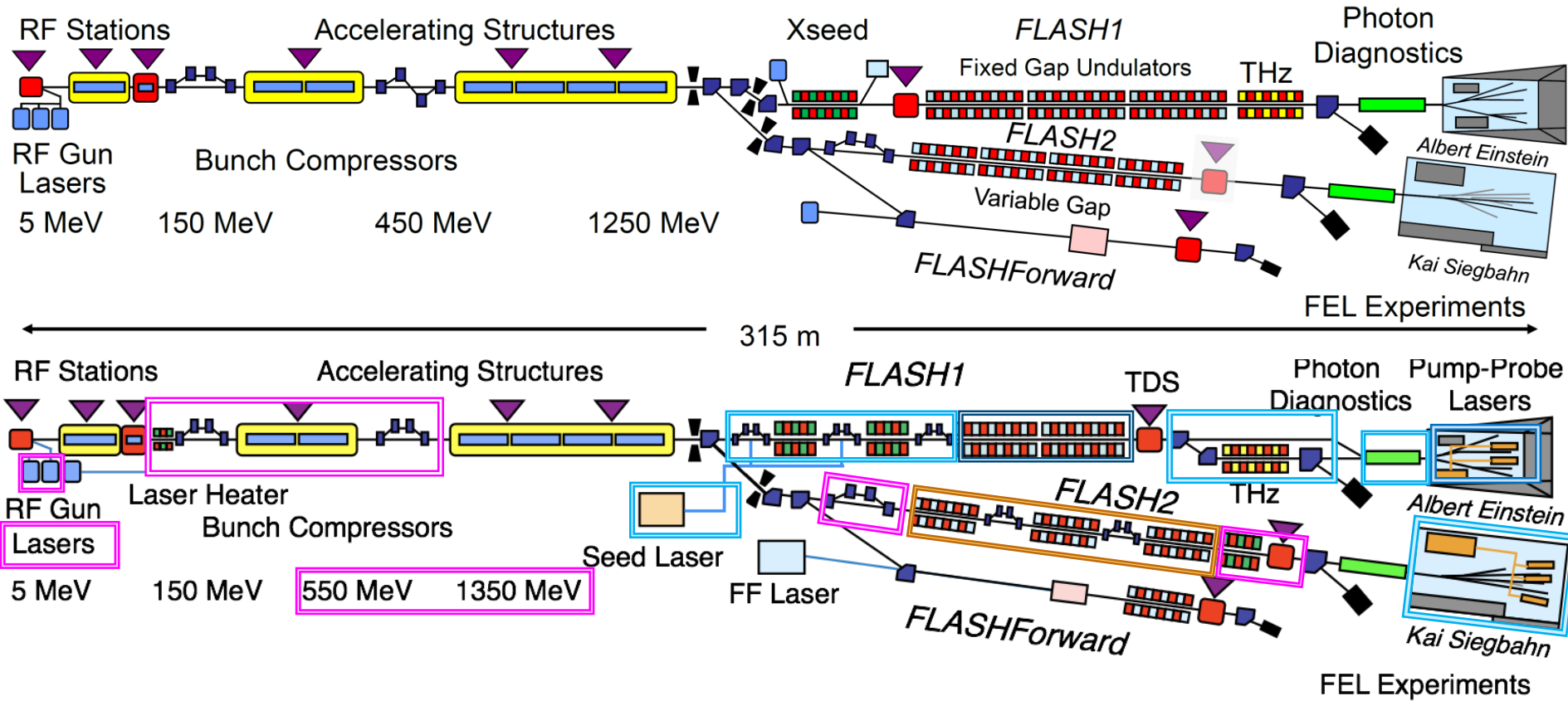


- Cross calibration campaign
- OPIS, PG spectrometer, CS spectrometer & VLS spectrometer
- Range 5-31 nm

FLASH2020+

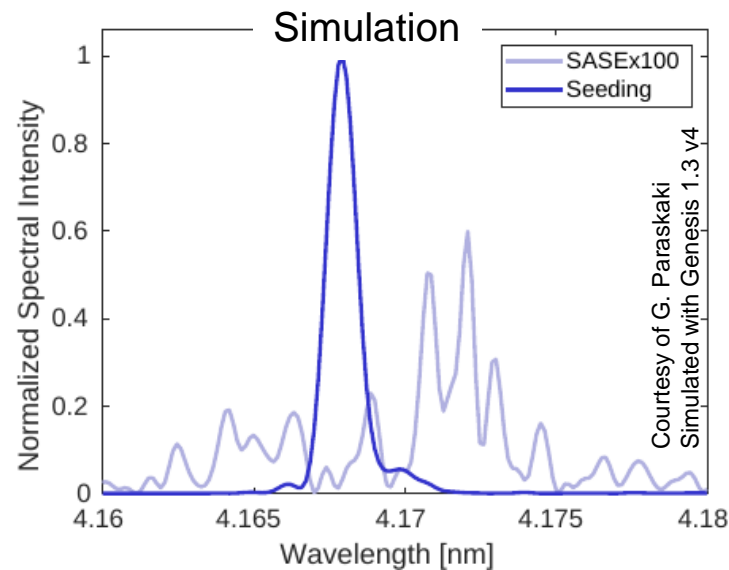
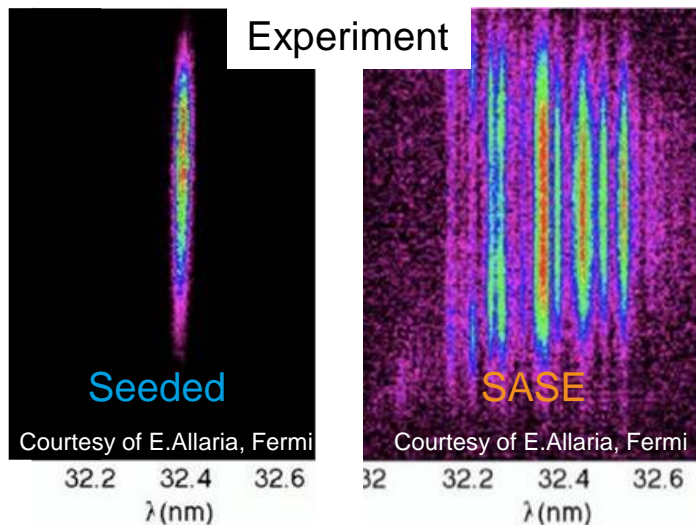
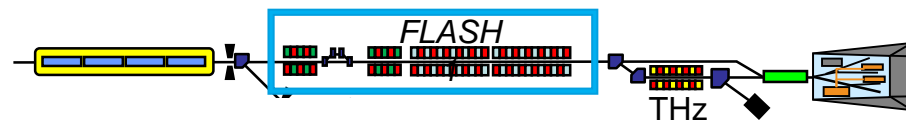
FLASH 'now' vs 2025

FLASH2020+ Upgrades of the facility



Difference between seeded and SASE pulses

Advantages of seeded FELs



Seeded

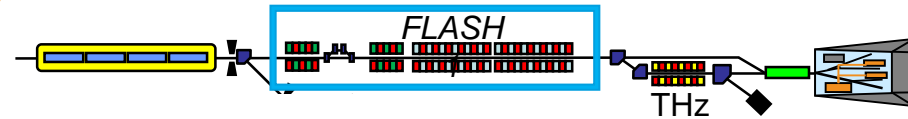
- Narrow bandwidth
- Stability
- Longitudinal coherence
- Brilliance
- Laser controlled pulse properties
- Synchronisation to seed laser

SASE

- Pulse energy
- Repetition rate

FLASH2020+ seeding in FLASH1

Starting from 2025 1 MHz coherent pulses in soft-X-ray

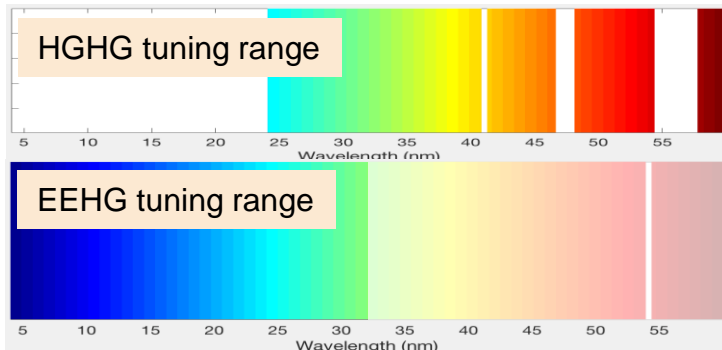
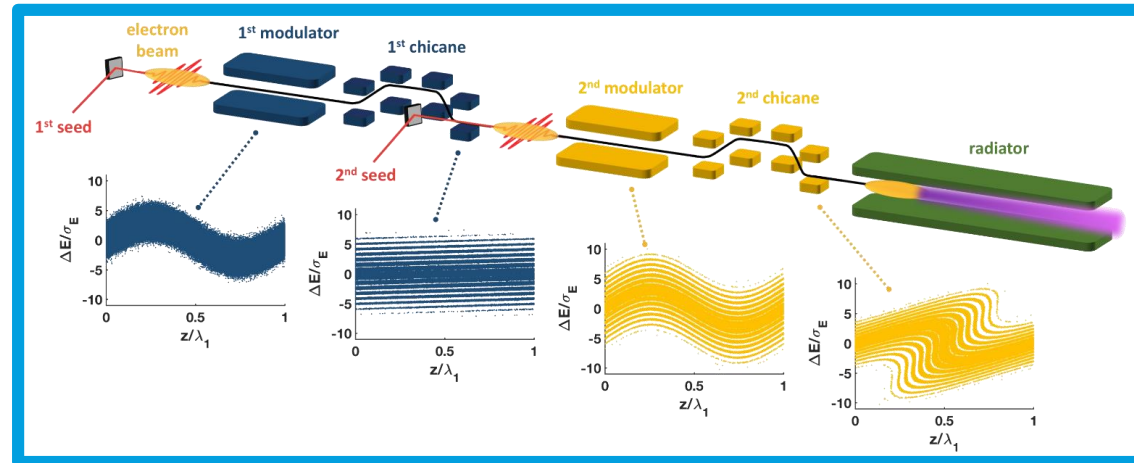


Combination of HGHG and EEHG:

Fully coherent pulses with
variable wavelength (60 – 4 nm)
tens of fs duration and
1 MHz repetition rate.

Apple III undulators:

Variable polarization



Successful seeding relies on **high quality** e-beam and seed lasers:

- Linac upgrade
- R&D for optimal lasers
- Seeding development

Seed 1: ~343 nm, 100 MW, 500 fs

Seed 2: 297-317 nm, 300 MW, 50 fs

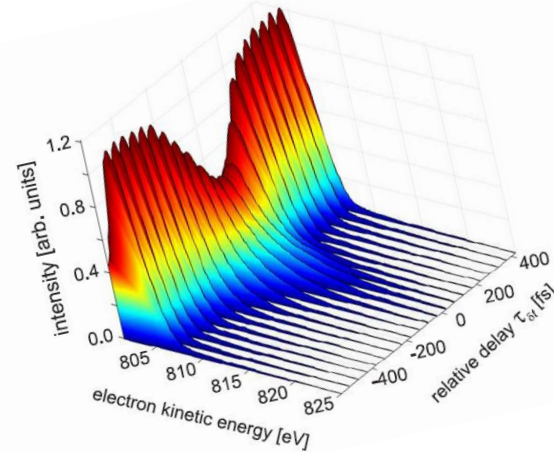
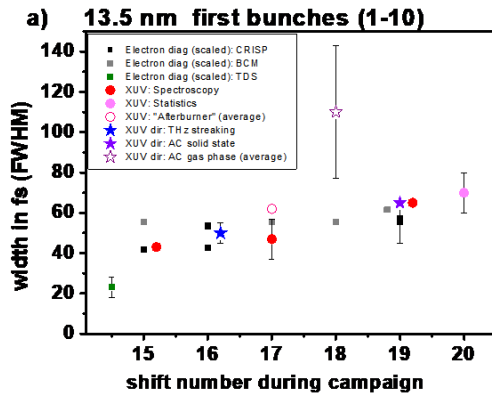
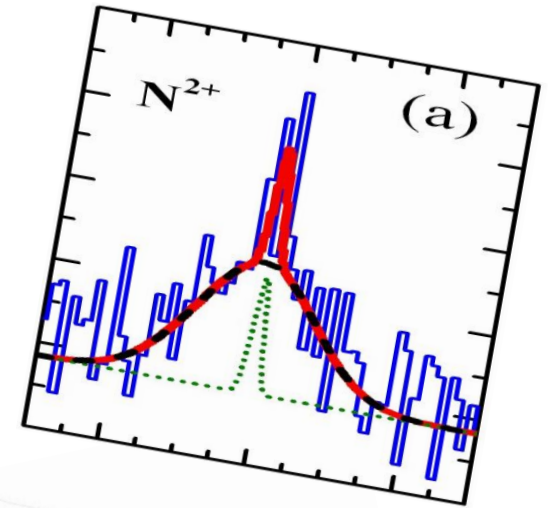
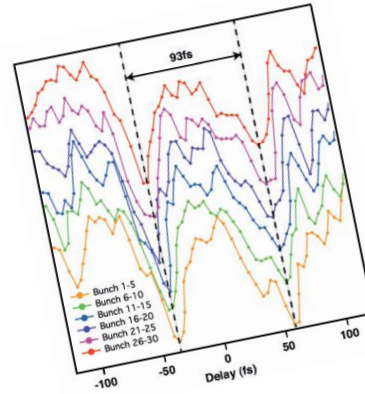
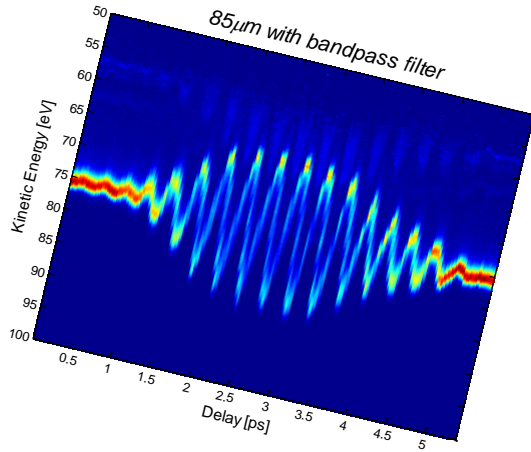
Thanks to S. Ackermann, T. Lang, M. Kazemi, M. Tischer

Thank you for your attention



Temporal distribution

Simultaneous Measurement of Electron and Photon Pulse Duration at FLASH



Goals for the short pulse studies

1. Can we setup the FEL to a **defined** pulse duration
2. Calibrate “**indirect**” methods against “**direct**” ones
3. Measure the scaling factor between **photon** pulse length and **electron** bunch length
4. Find out **advantages / disadvantages** of different methods

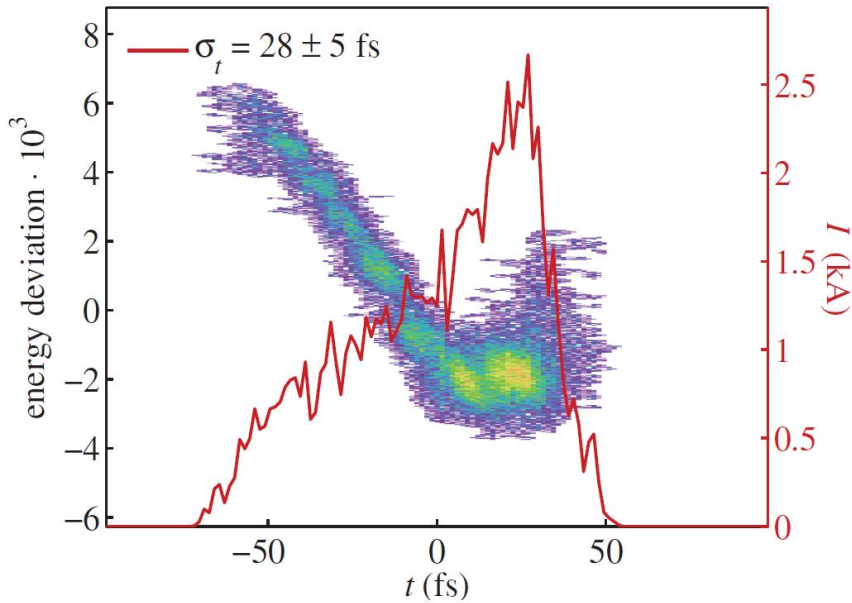
Outline – temporal distribution

- > Electron beam diagnostics
 - Transverse Deflecting Structure (TDS)
 - (THz spectroscopy (CRISP))
 - (Bunch Compression Monitor (BCM))

- Indirect photon based methods
 - Spectral characteristics
 - (Pulse energy fluctuations – statistics)
 - (Mapping SASE to visible light: “afterburner”)

- Direct photon based methods
 - Autocorrelation
 - THz streaking
 - Optical-XUV cross-correlation

Electron Diagnostics: transverse deflecting cavity

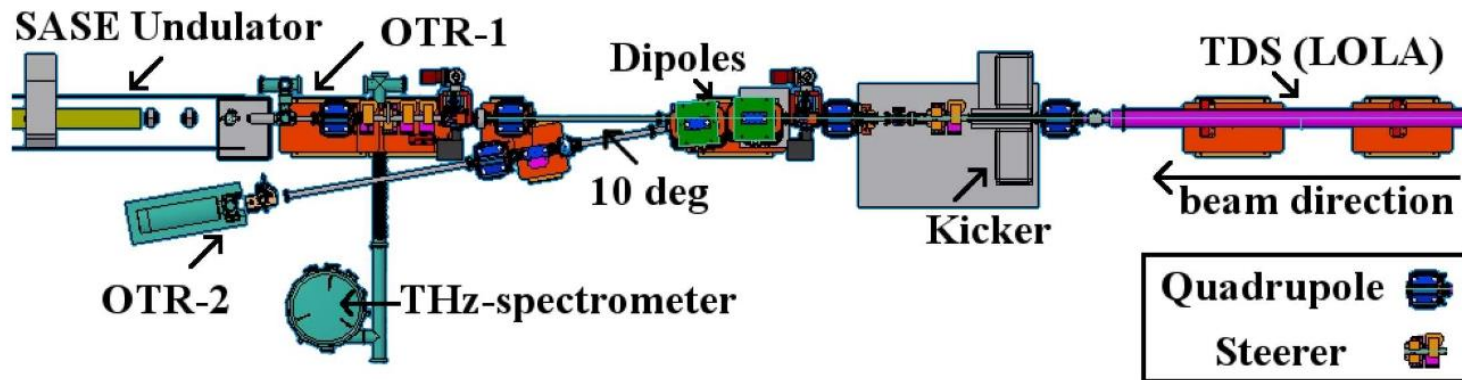


PRO:

- very good resolution (few fs)
- (meanwhile) online diagnostic
- Arbitrary pulse in bunch train can be measured

CON:

- only 1 bunch out of bunch train
-> destructive
- dispersive measurements (chirp)
-> not online



Courtesy: M. Yan, Ch. Gerth

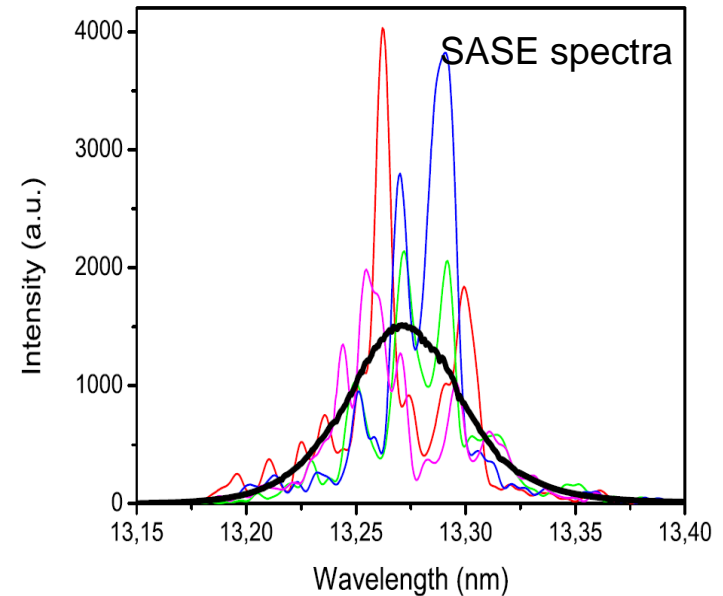
Photon pulse length diagnostics at PG2

Different methods/tools under development to measure photon pulse length & temporal distribution

- CRISP
- LOLA
- THz streaking
- Reflectivity method
- optical afterburner
-
-

Spectral analysis

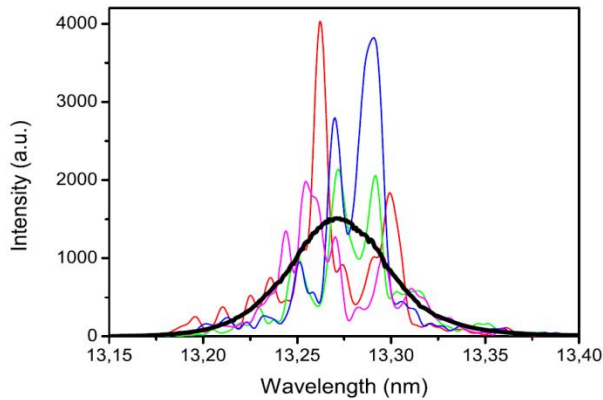
Employ Fourier relation between spectral distribution and temporal properties



- requires high resolution PG2 spectrometer
- Spectral correlation yields pulse duration

From spectra to photon pulse duration

Set of FEL spectra
measured with PG2
beamline

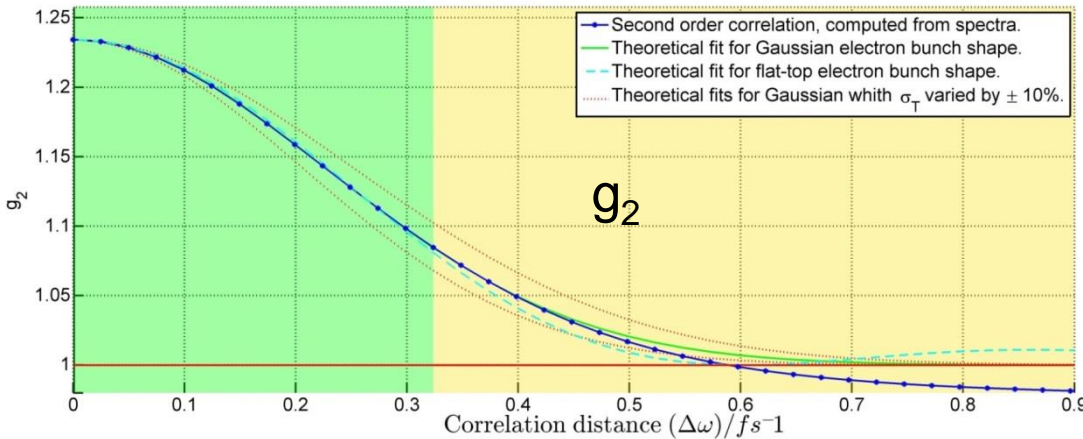


2nd order correlation function
(from spectra):

$$g_2(\Delta\omega) \propto \frac{\langle S(\omega_0 - \frac{\Delta\omega}{2}) S(\omega_0 + \frac{\Delta\omega}{2}) \rangle}{\langle S(\omega_0 - \frac{\Delta\omega}{2}) \rangle \langle S(\omega_0 + \frac{\Delta\omega}{2}) \rangle}$$

Expected 2nd order correlation function for given σ_T
and electron bunch shape:

$$g_2(\Delta\omega) = 1 + |\bar{F}(\Delta\omega, \sigma_T)|^2 \quad \text{Fit parameter}$$



Electron bunch shape

$$\bar{F}^g = e^{-\frac{\Delta\omega^2 \sigma_T^2}{2}} \quad \text{and} \quad \bar{F}^{ft} = \frac{\sin(\Delta\omega \frac{\sigma_T}{2})}{\Delta\omega \frac{\sigma_T}{2}}$$

A least squares fit optimizes the rms
pulse duration σ_T for maximum
agreement within a set correlation
window.

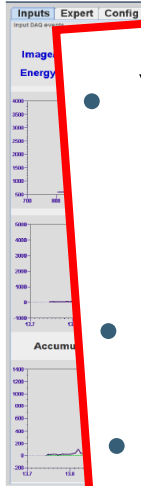
R. Engel (Bachelor Thesis)

S. Serkez (XFEL)

Lutman et al, *PRST* 15, 030705 (2012)

Real time analysis of FLASH spectra

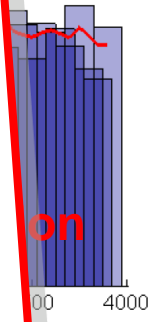
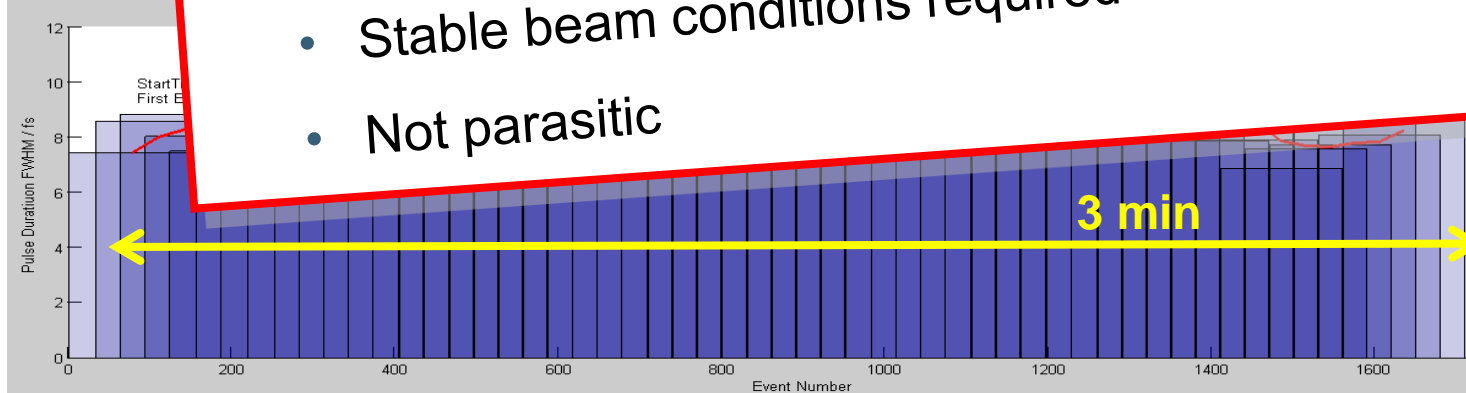
Photon pulse length (PPL) server Multiple run



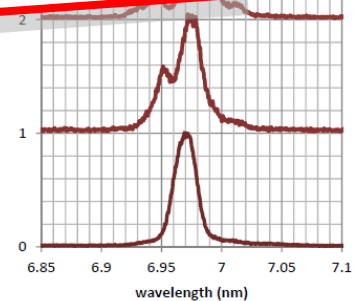
- Very useful tool for detailed photon pulse diagnostics
- Pulse length estimations during run time possible
- No additional setup required
- Limitations:

- Sensitive to electron bunch shape & energy chirp
- Stable beam conditions required
- Not parasitic

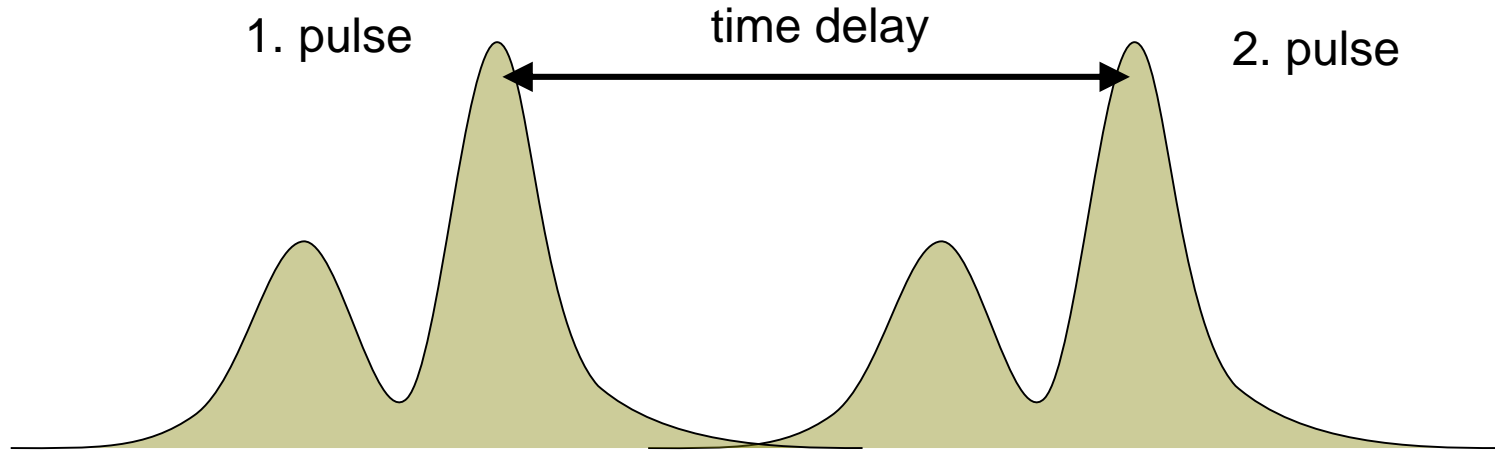
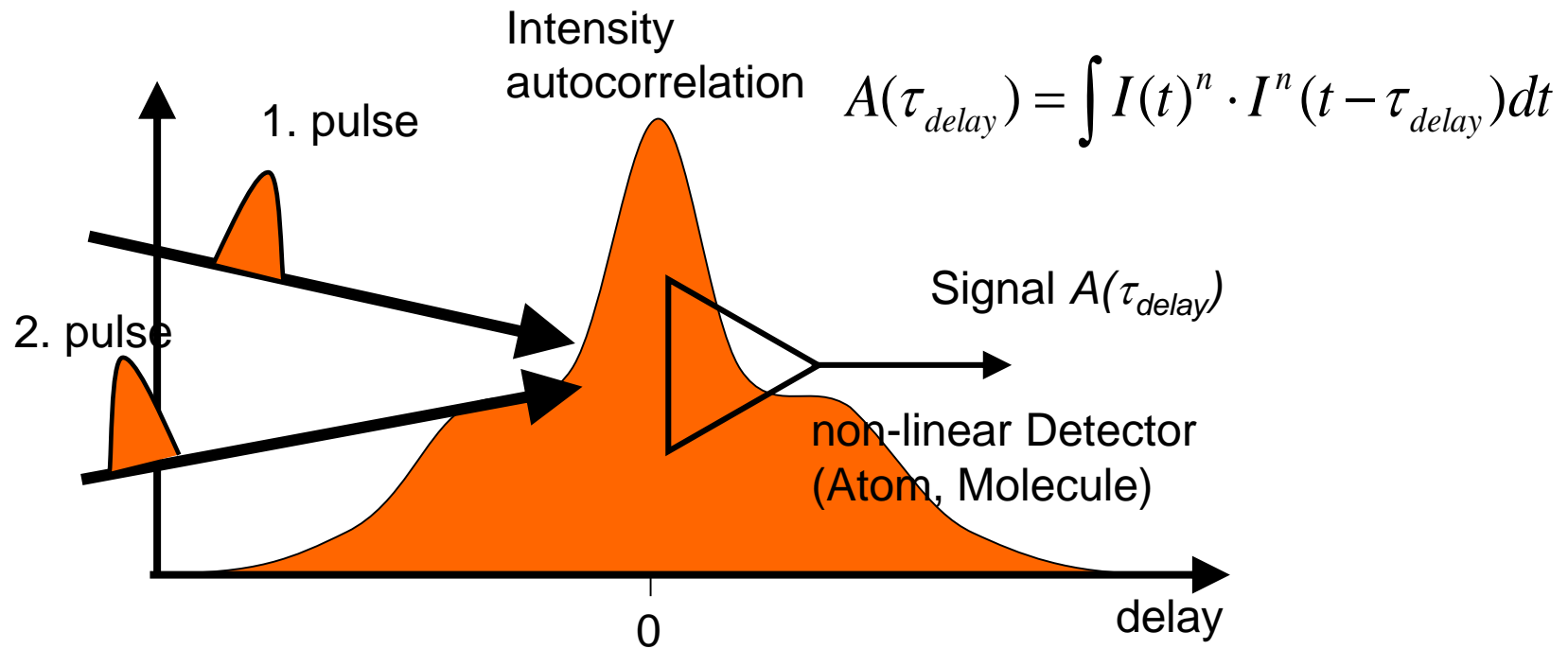
FLASH
(J. Roer)



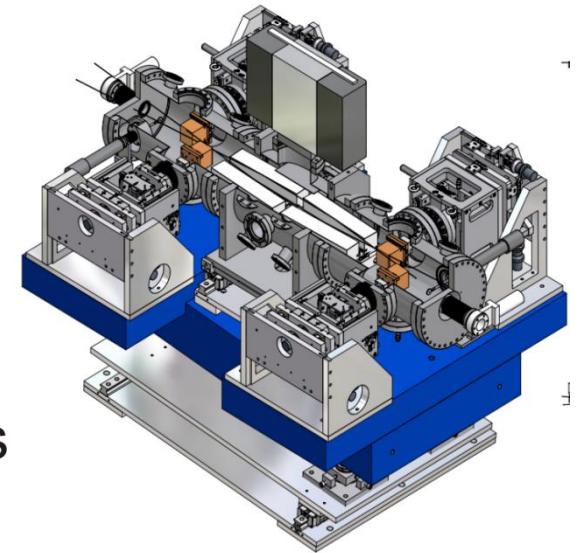
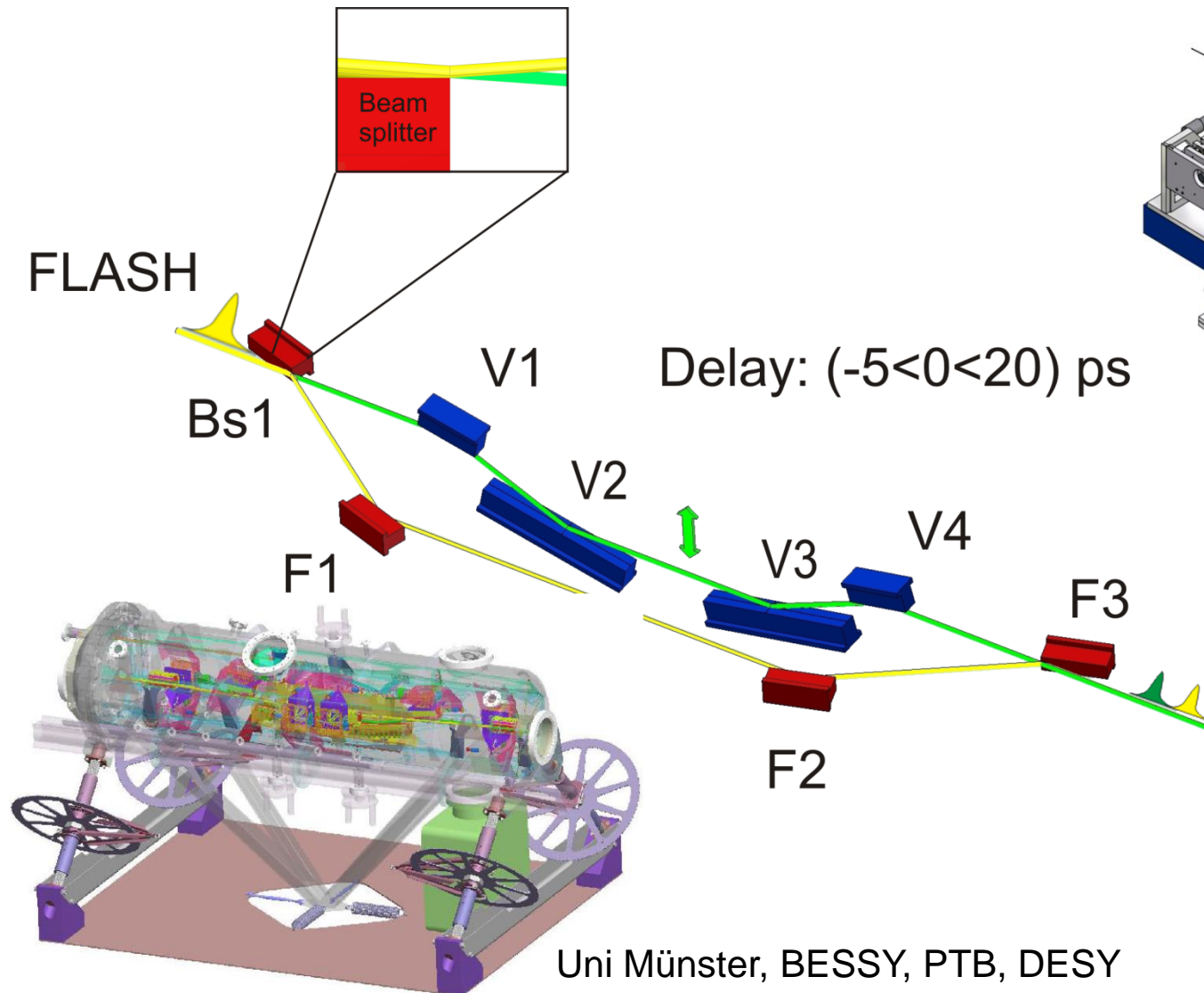
element
spectra)
single
spectra



Direct PHOTON methods: auto correlation

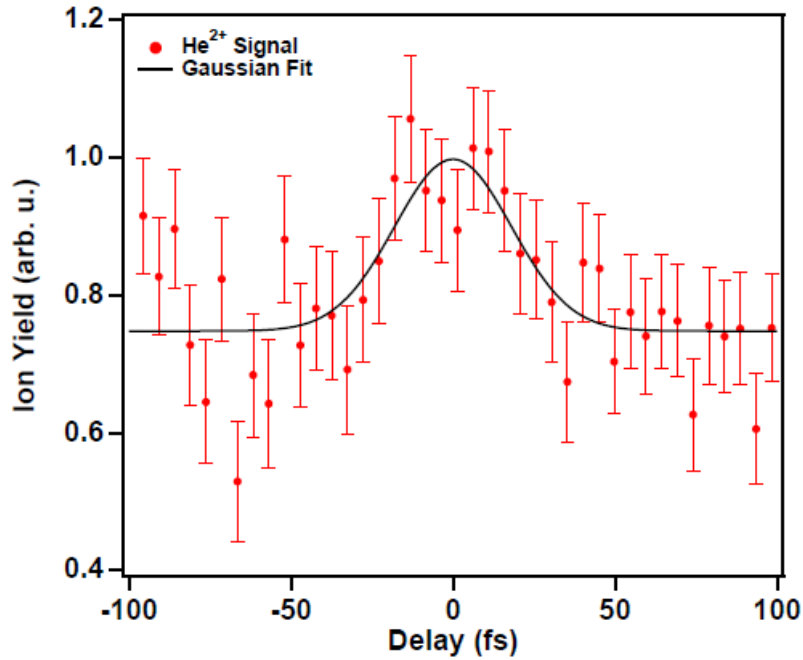


FEL split and delay

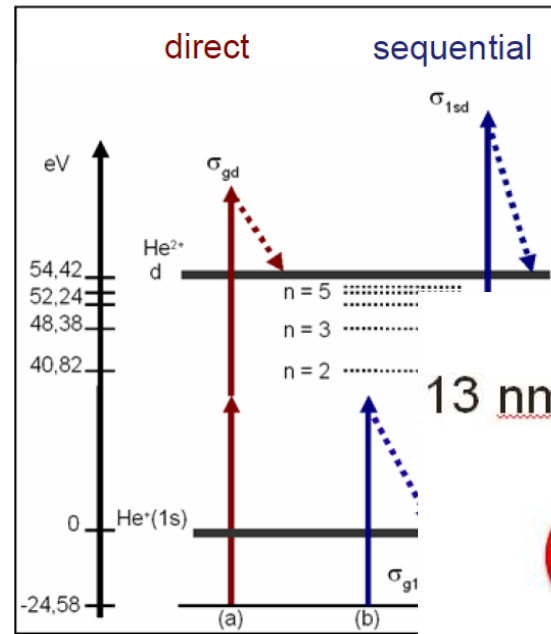


R. Mitzner, et al. Optics Express 16, 19909 (2008);
F. Sorgenfrei, et al, Rev. Sci. Instrum. 81, 043107 (2010)

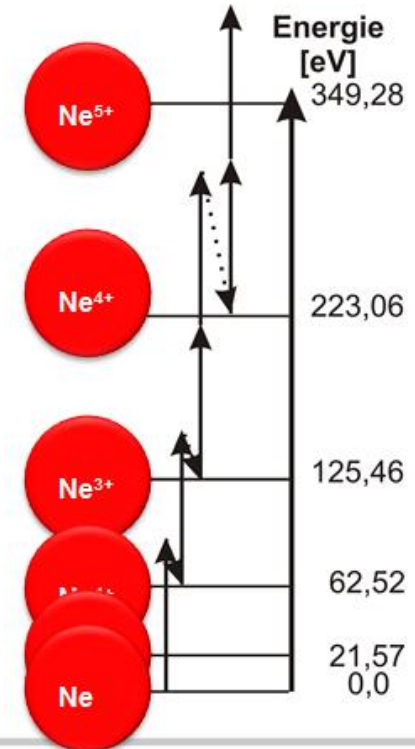
Direct PHOTON methods: auto correlation



Pathways to He²⁺ at 24 nm



13 nm (~92 eV)



Pro

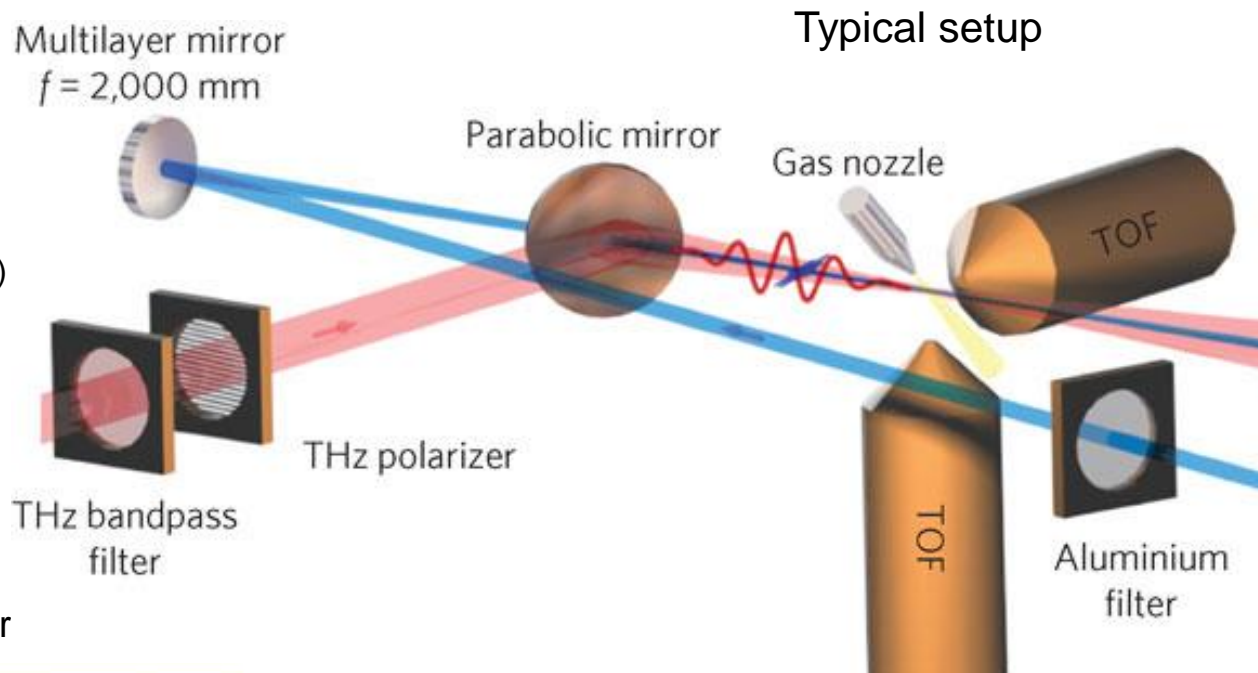
- “direct” measurement (for known reactions)

Con

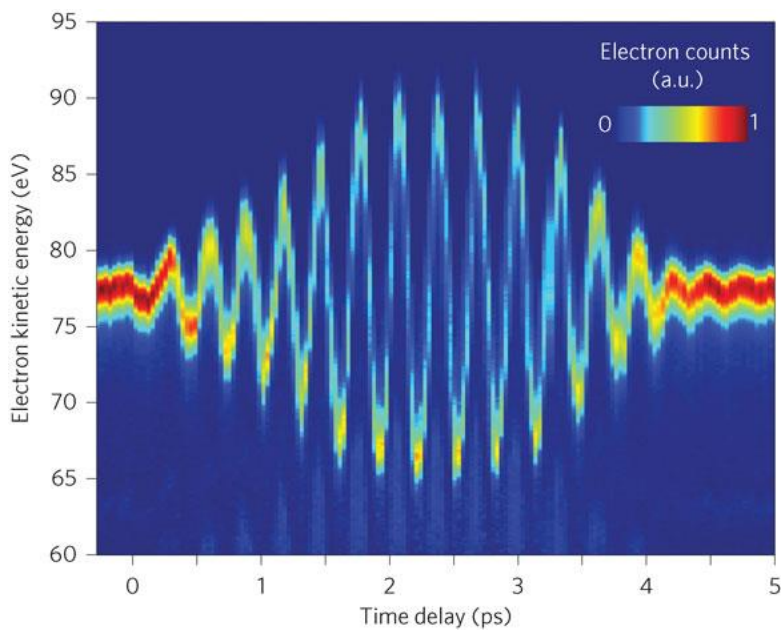
- **Experimentally challenging** (takes long time)
- (up to now) averaging technique
- **well defined for < 25 nm**
- For XUV several path lead to same ionization state -> Simulations needed

Direct PHOTON methods: THz streaking

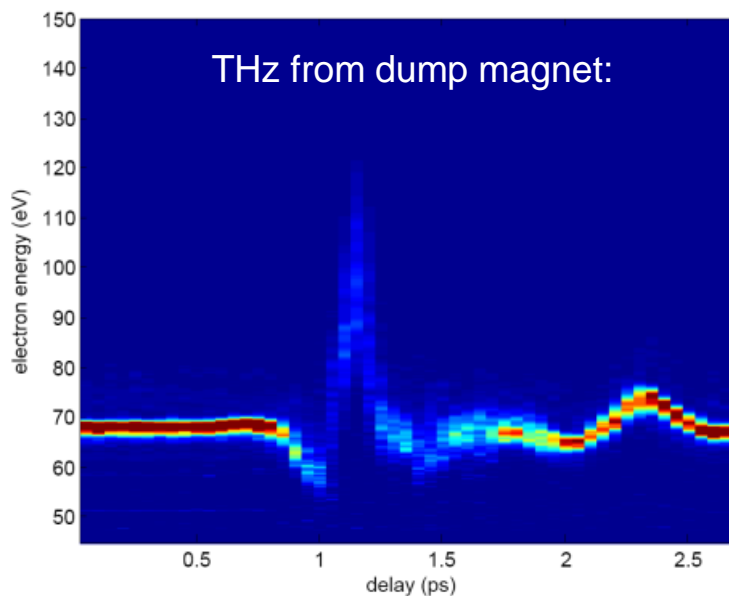
Nature Photonics **3**, 523 (2009)
Phys. Rev. Lett. **108**, 253003 (2012)



THz from undulator



THz from dump magnet:



XUV photon diagnostics: pulse duration

The most difficult one

Extensive study to different techniques: Düsterer et al, PRSTAB 17, 120702 (2014)

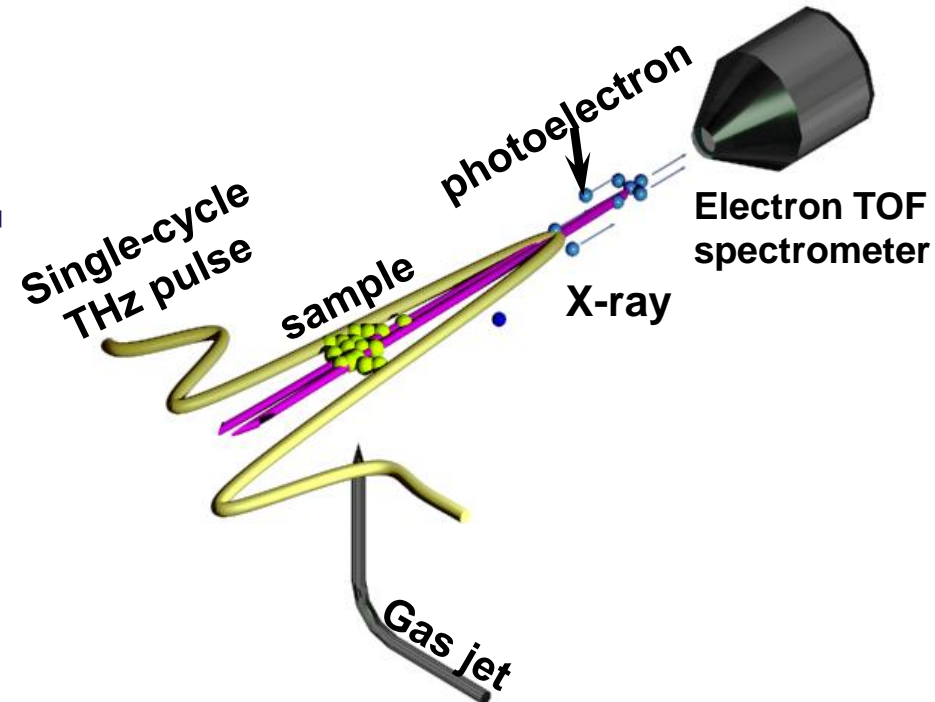
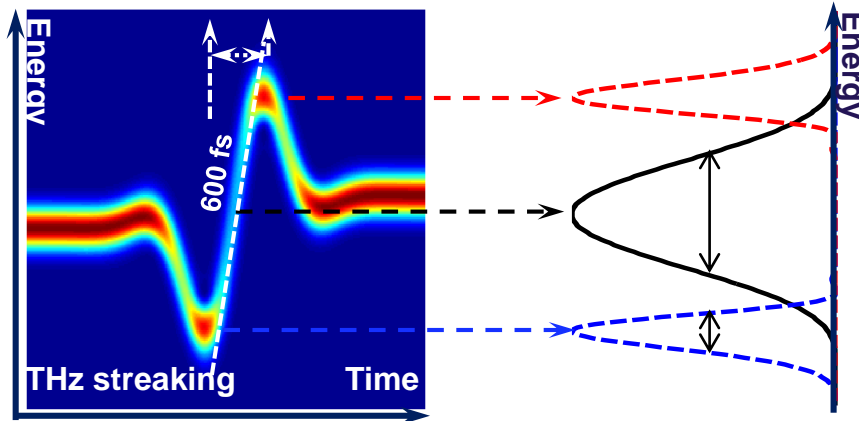
THz Streaking

measuring the pulse duration and arrival time by means of a single cycle THz streaking field:

- online monitoring
- single bunch resolved measurement
- (almost) non-invasive
- High rep. rate possible

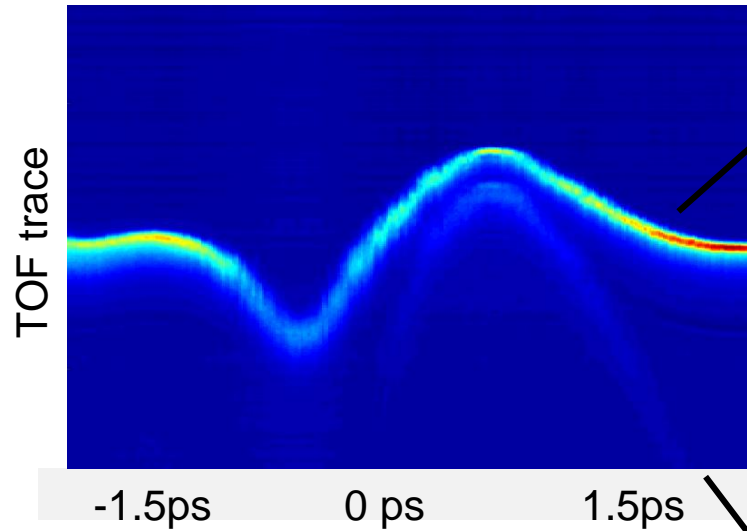
(usual) Parameter range

- 20 - 200 fs
- 4 - 40 nm
- 1 - 500 μJ

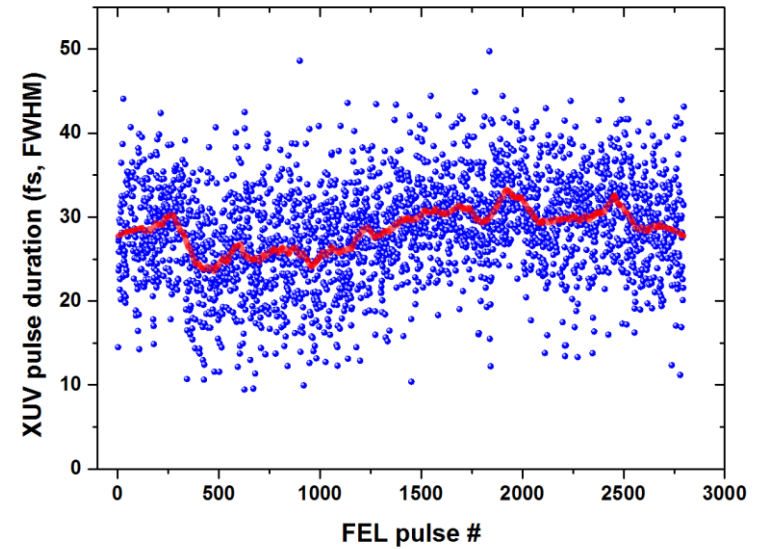


THz Streaking -> observables

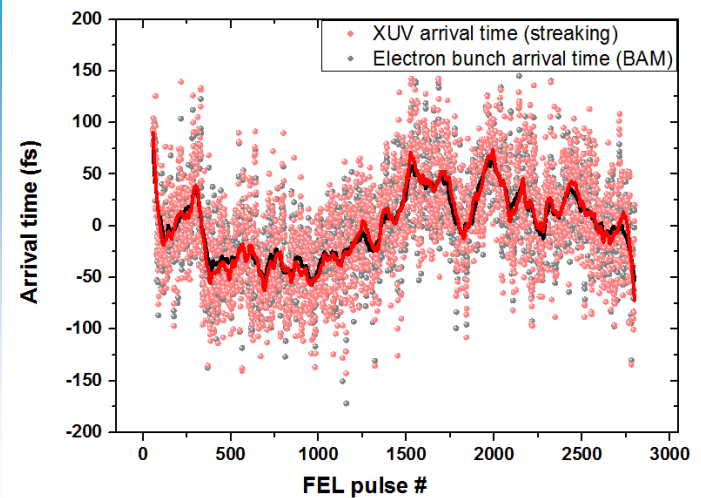
Streaking raw data
(delay scan)



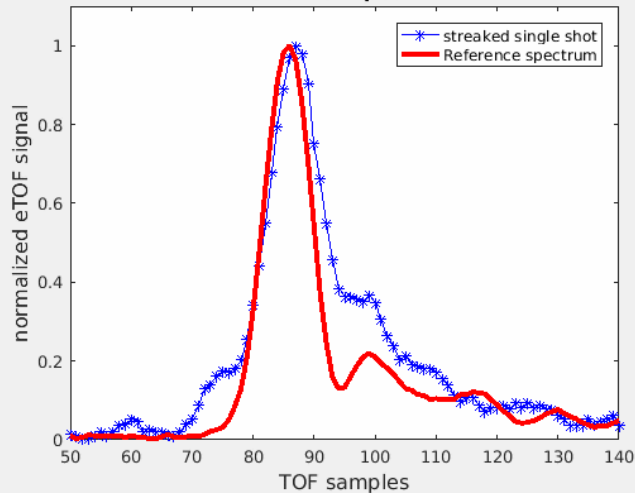
Single shot XUV pulse duration



Arrival time

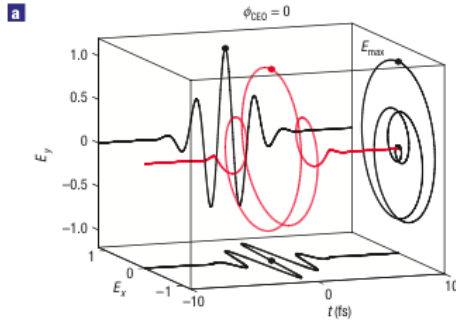


Streaked spectra



Towards attosecond pulses

Principle of angle-resolved streaking



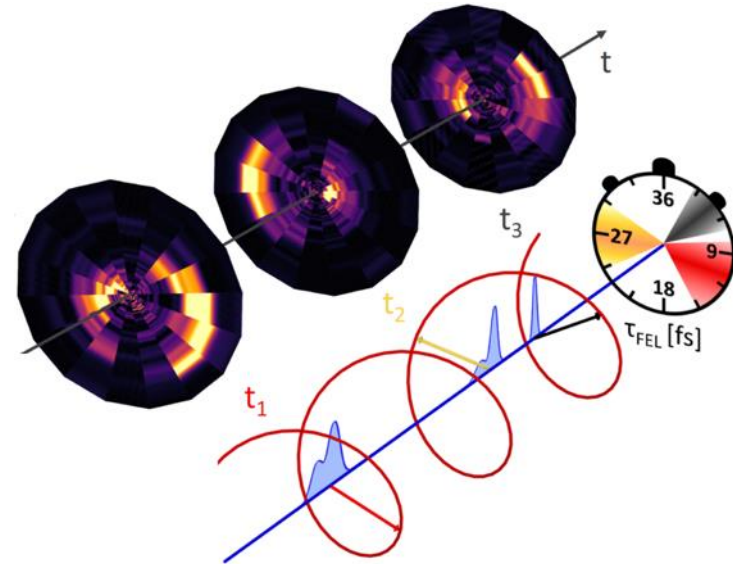
Principle:

E. Constant, P. Corkum, *Phys. Rev. A* **56** (1997)

Experiment & figure (IR ionization):

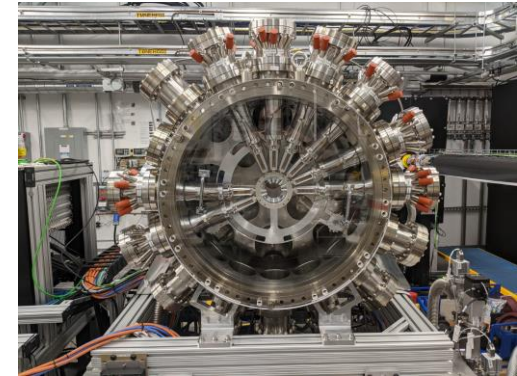
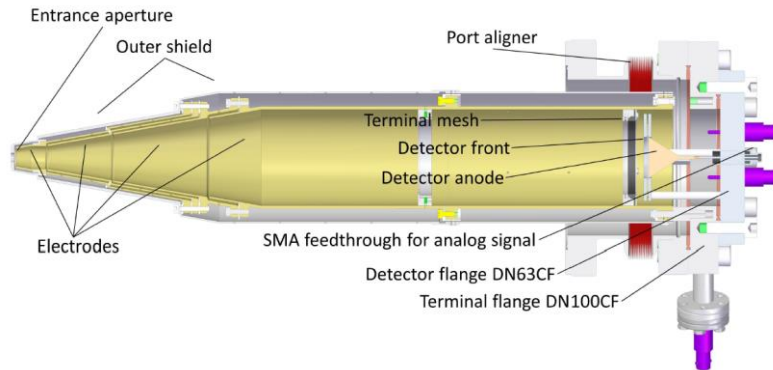
P. Eckle, U. Keller, *Nat. Phys.* **4** (2008)

- Circularly polarized laser field introduces time-dependent shift in streaking angle
- Angle-resolved detection of the photoelectron energy acts like the hands of a stop watch



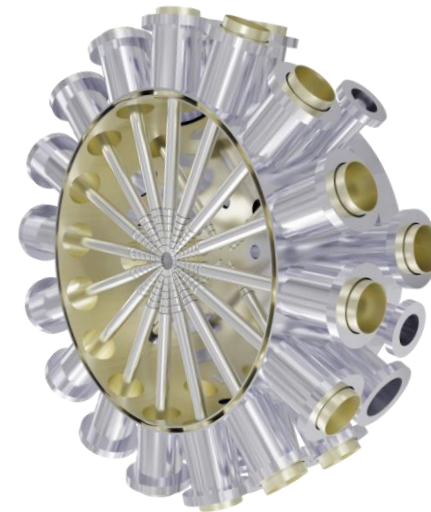
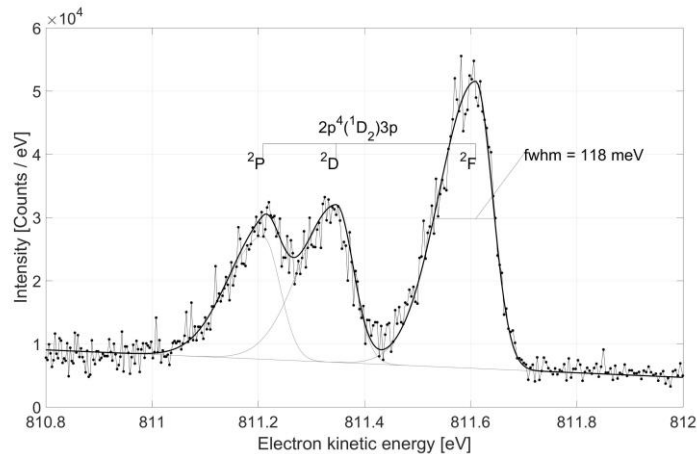
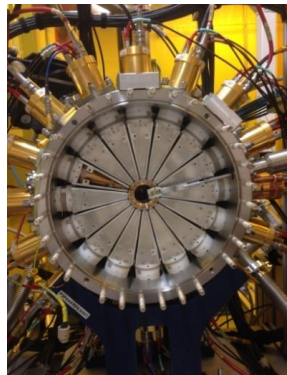
Instrumentation upgrades and advances

From diagnostic achievements to chirality science at the attosecond frontier in gas and liquid phase



P. Walter et al.
J. Synchrotron Rad. **28**, 1364 (2021)

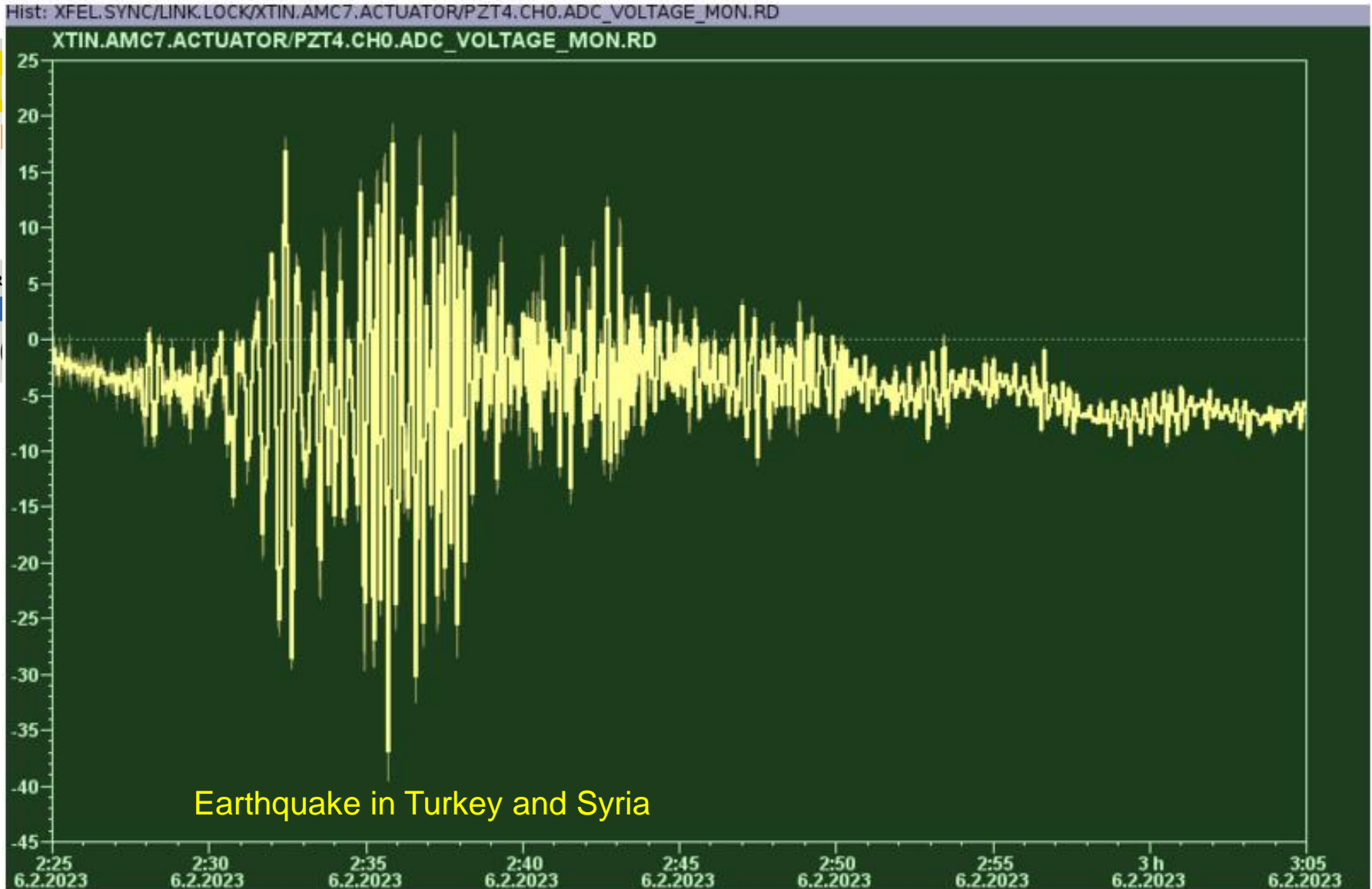
A. DeFanis et al. J. Synchrotron Rad. **29**, 755–764 (2022)



SPAR Project
for atto-streaking
W. Helm et al.

Synchronization

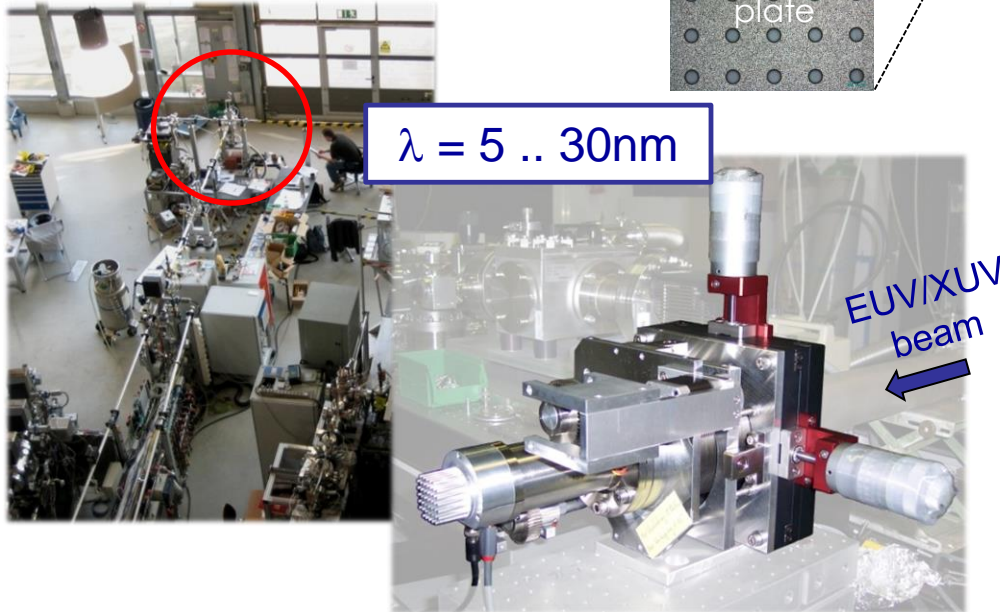
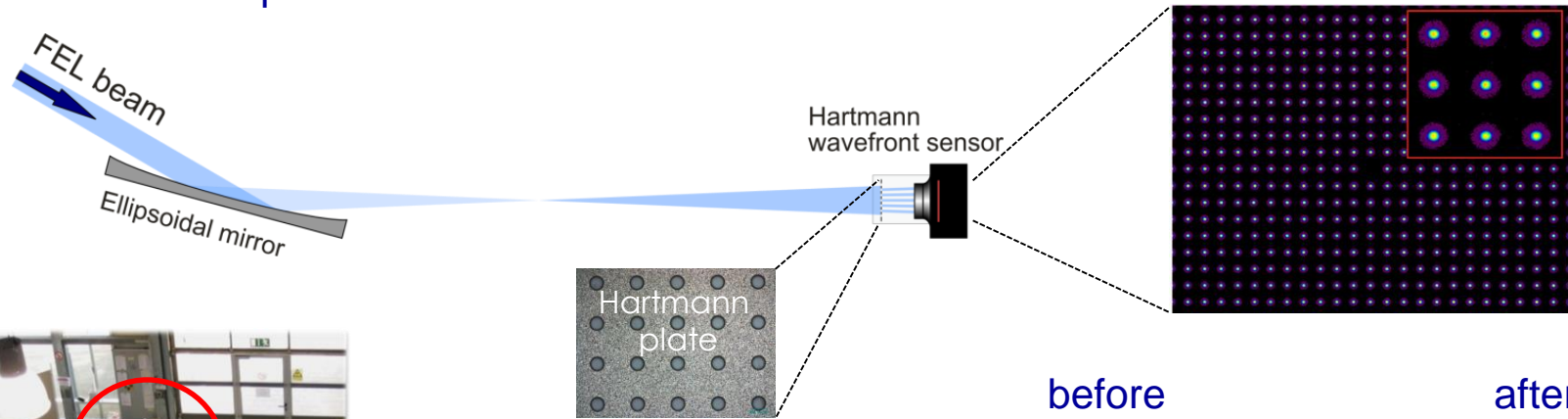
Femtosecond Optical Synchronization Systems at FLASH and the European XFEL



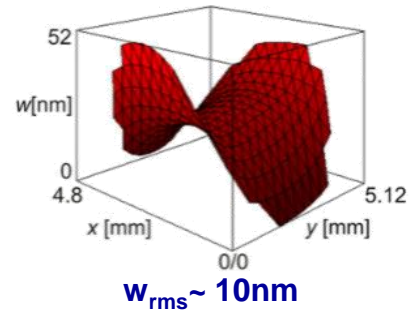
Wigner distribution measurement of the spatial coherence properties of FLASH

Credits to: Tobias Mey (Laserlaboratorium Göttingen e.V.)

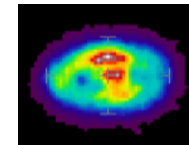
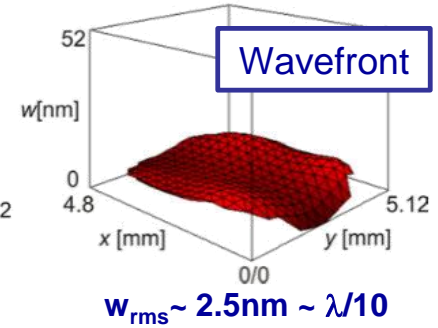
Experimental setup at BL2



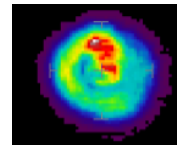
before adjustment



after adjustment



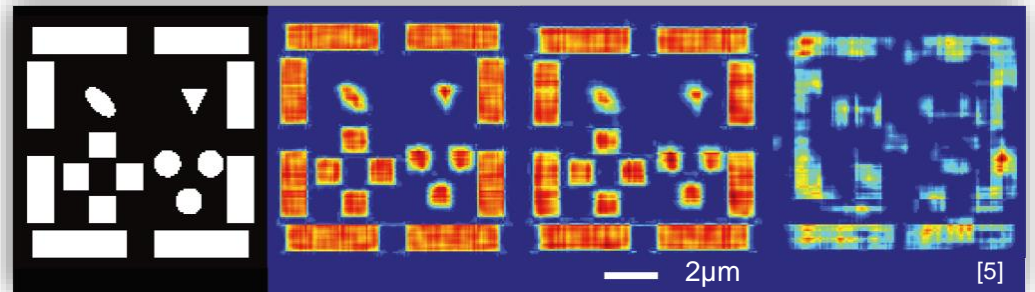
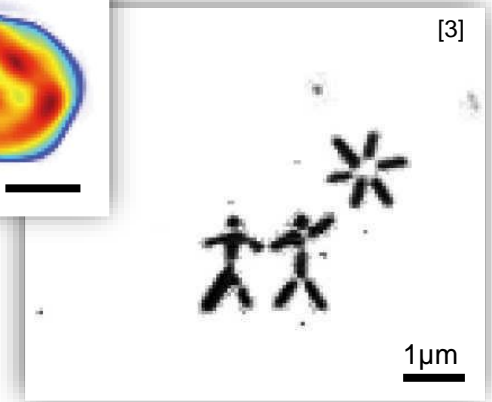
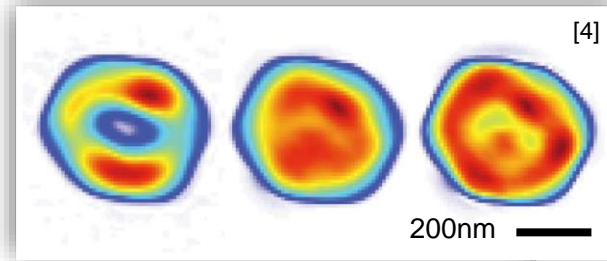
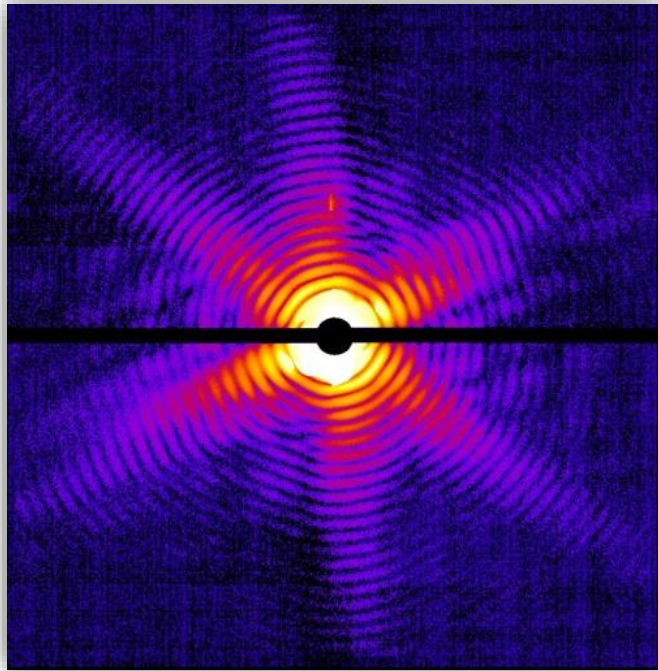
Intensity



[2] B. Flöter et al, EUV Hartmann sensor for wavefront measurements at the Free-electron LASer in Hamburg, New J. Phys. 12 (2010) 083015

Motivation

Coherent diffractive imaging



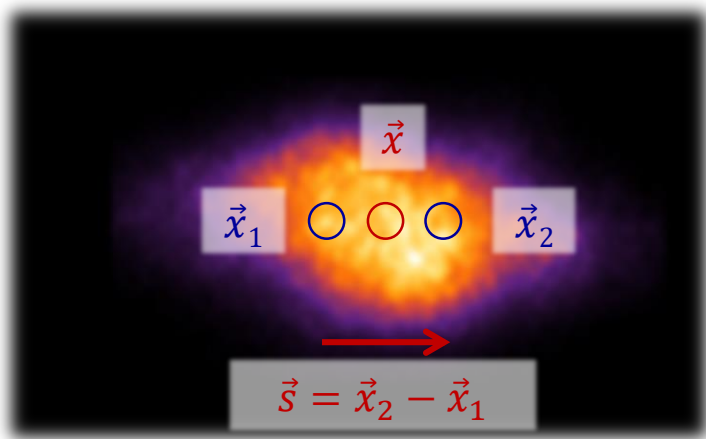
Decreasing coherence →

[3] H. N. Chapman *et al.*, "Femtosecond diffractive imaging with a soft-X-ray free-electron laser," *Nature Phys.* **2**, 839-843 (2006)

[4] M. M. Seibert *et al.*, "Single mimivirus particles intercepted and imaged with an X-ray laser," *Nature* **470**, 78-82 (2011)

[5] B. Chen *et al.*, "Diffraction imaging: The limits of partial coherence," *Phys. Rev. B* **86**, 235401 (2012)

Coherence



Mutual coherence function

$$\begin{aligned}\Gamma(\vec{x}, \vec{s}) &= \langle E(\vec{x}_1, t) \cdot E^*(\vec{x}_2, t) \rangle \\ &= \langle E(\vec{x} - \vec{s}/2, t) \cdot E^*(\vec{x} + \vec{s}/2, t) \rangle\end{aligned}$$

Local degree of coherence

$$\gamma(\vec{x}, \vec{s}) = \frac{\Gamma(\vec{x}, \vec{s})}{\sqrt{I(\vec{x} - \vec{s}/2) \cdot I(\vec{x} + \vec{s}/2)}}$$

Global degree of coherence

$$K = \frac{\iint \Gamma(\vec{x}, \vec{s})^2 d\vec{x} d\vec{s}}{(\iint \Gamma(\vec{x}, 0) d\vec{x})^2}$$

→ required for interference effects

[6] M. Born and B. Wolf, *Principles of Optics*, Cambridge University Press (1980)

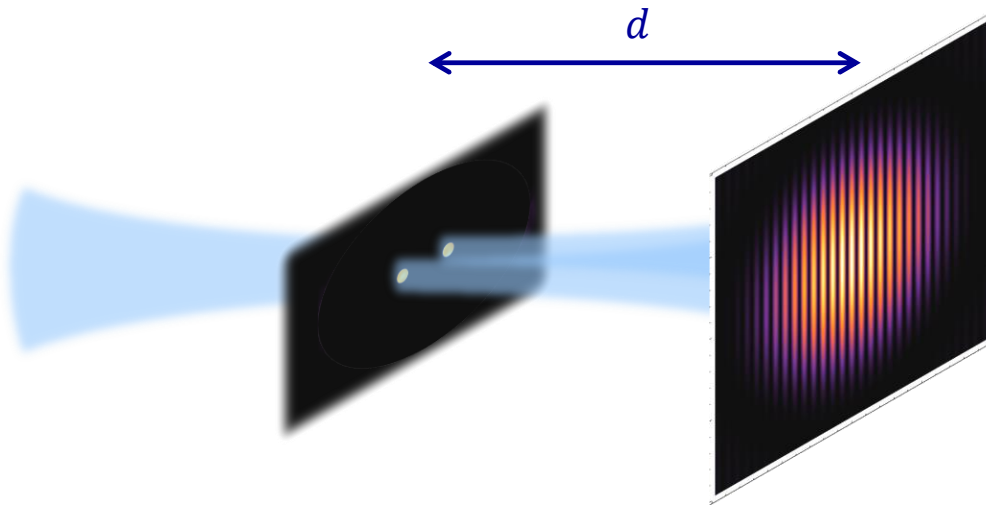
Coherence

Interference of elementary waves $\rightarrow \gamma(\vec{x}, \vec{s})$

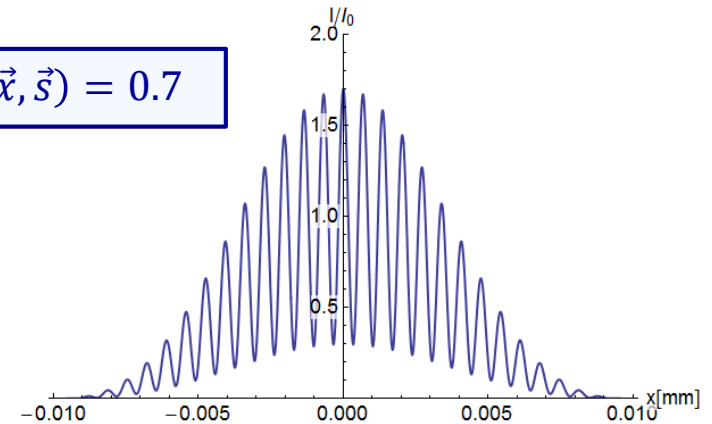
$$\gamma(\vec{x}, \vec{s}) = 0.7$$

$$I(x, y) = I_0 \cdot \left(\frac{J_1\left(\frac{2\pi ar}{\lambda d}\right)}{\frac{2\pi ar}{\lambda d}} \right)^2 \cdot \left[1 + \gamma(\vec{x}, \vec{s}) \cdot \cos\left(\frac{2\pi s}{\lambda d} x\right) \right] \quad [6]$$

$$r = \sqrt{x^2 + y^2}$$

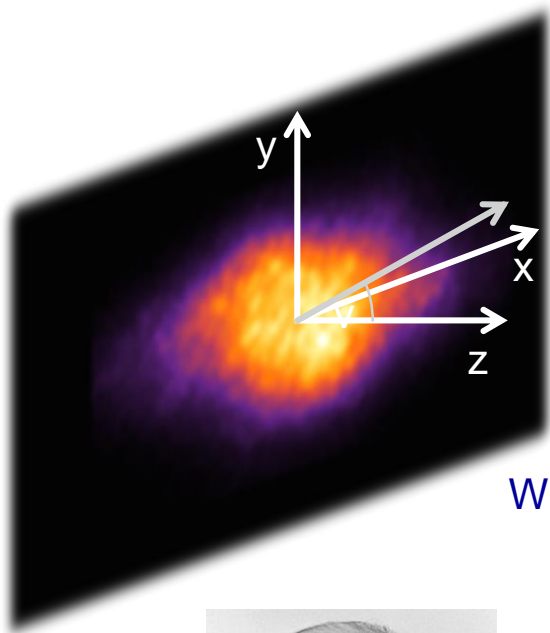


$$\gamma(\vec{x}, \vec{s}) = 0.7$$



[6] M. Born and B. Wolf, *Principles of Optics*, Cambridge University Press (1980)

Wigner distribution function



Spatial coordinate $\vec{x} = \begin{pmatrix} x \\ y \end{pmatrix}$

Mutual coherence function

$$h(\vec{x}, \vec{u}) = \left(\frac{k}{2\pi}\right)^2 \cdot \iint \Gamma(\vec{x}, \vec{s}) \cdot e^{ik\vec{u} \cdot \vec{s}} d^2s$$

Wigner distribution

Radiation angle $\vec{u} = \begin{pmatrix} u \\ v \end{pmatrix}$



Eugene Paul Wigner
Nobel price 1963

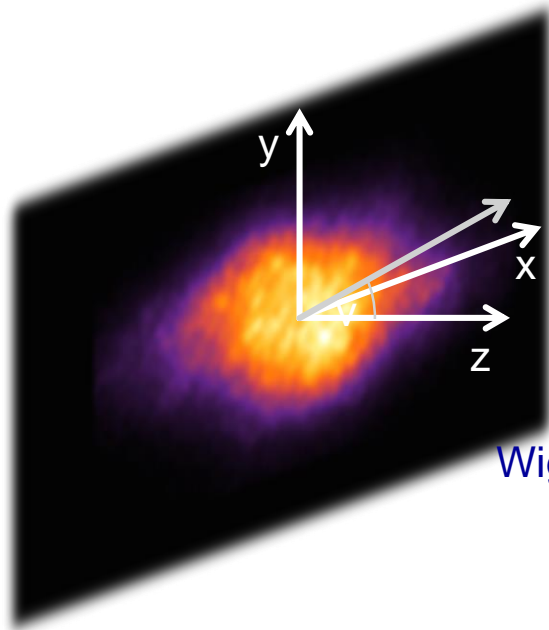
(with J. H. D. Jensen and
M. Goeppert-Mayer)

Wilhelm-Weber-Straße 22,
Göttingen ▶



[8] M. J. Bastiaans, "Application of the Wigner distribution function to partially coherent light," J. Opt. Soc. Am. A 3, 1227-1238 (1986)

Wigner distribution function



Spatial coordinate $\vec{x} = \begin{pmatrix} x \\ y \end{pmatrix}$

Mutual coherence function

$$h(\vec{x}, \vec{u}) = \left(\frac{k}{2\pi}\right)^2 \cdot \iint \Gamma(\vec{x}, \vec{s}) \cdot e^{ik\vec{u} \cdot \vec{s}} d^2s$$

Wigner distribution

Radiation angle $\vec{u} = \begin{pmatrix} u \\ v \end{pmatrix}$

Irradiance

$$I(\vec{x}) = \iint h(\vec{x}, \vec{u}) du dv$$

→ Near field

Radiance

$$\hat{I}(\vec{u}) = (2\pi)^{-2} \iint h(\vec{x}, \vec{u}) dx dy$$

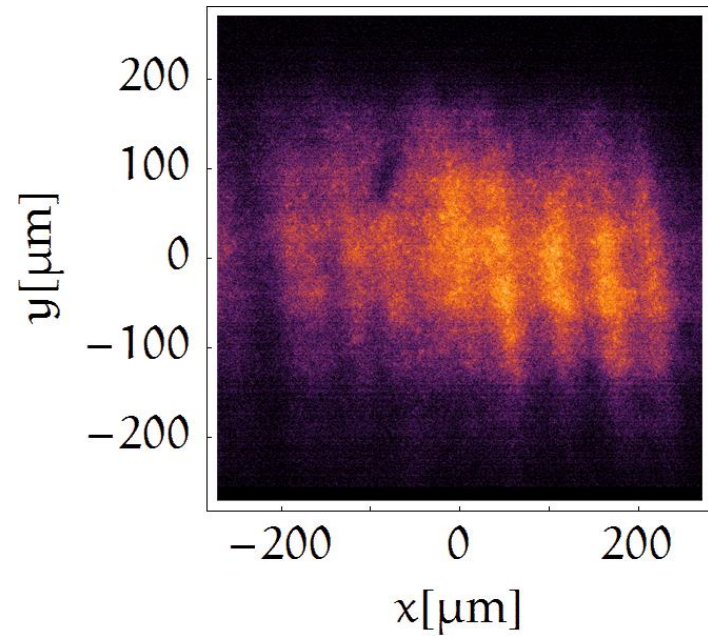
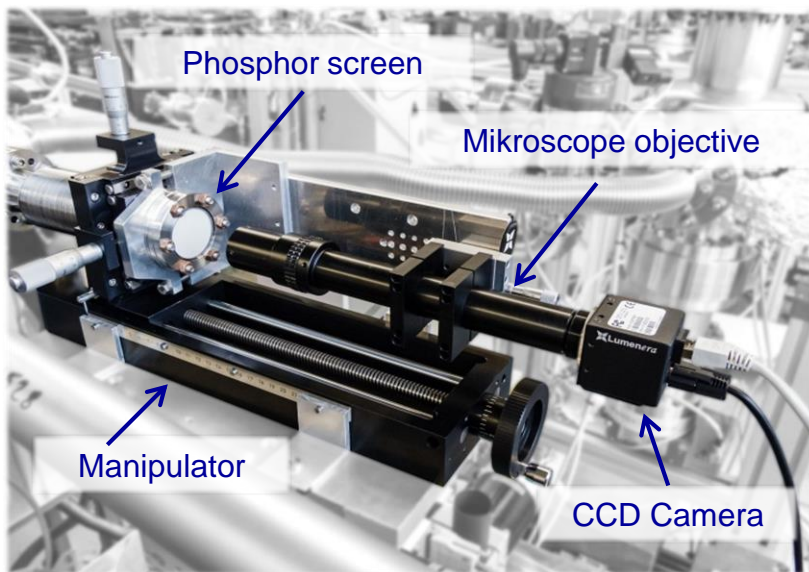
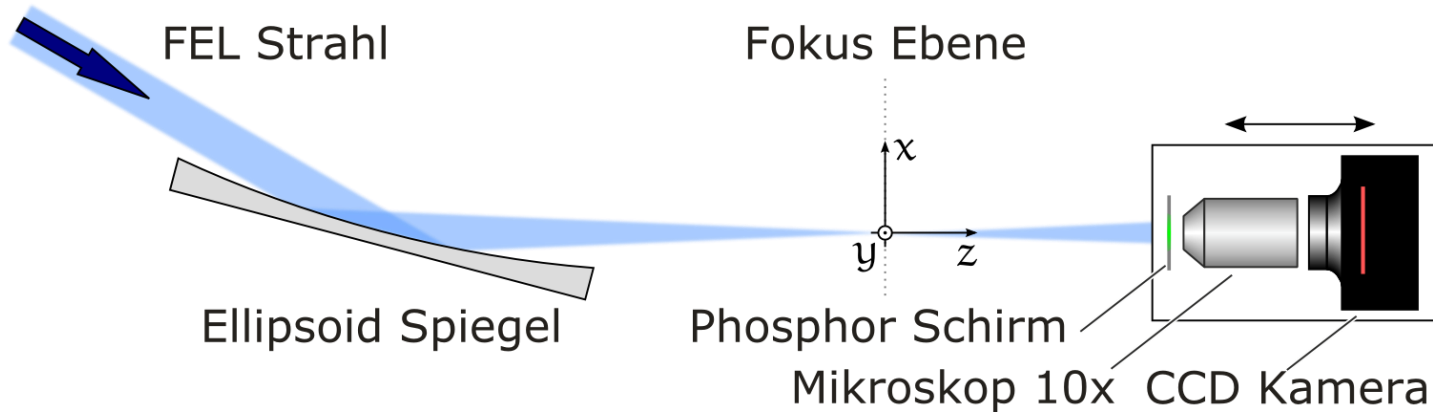
→ Far field

Global degree of coherence

$$K = \lambda^2 \frac{\iint h(\vec{x}, \vec{u})^2 dx^2 du^2}{\iint h(\vec{x}, \vec{u}) dx^2 du^2}$$

[8] M. J. Bastiaans, "Application of the Wigner distribution function to partially coherent light," J. Opt. Soc. Am. A **3**, 1227-1238 (1986)

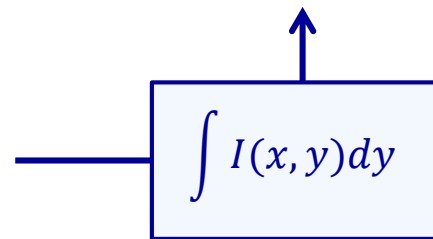
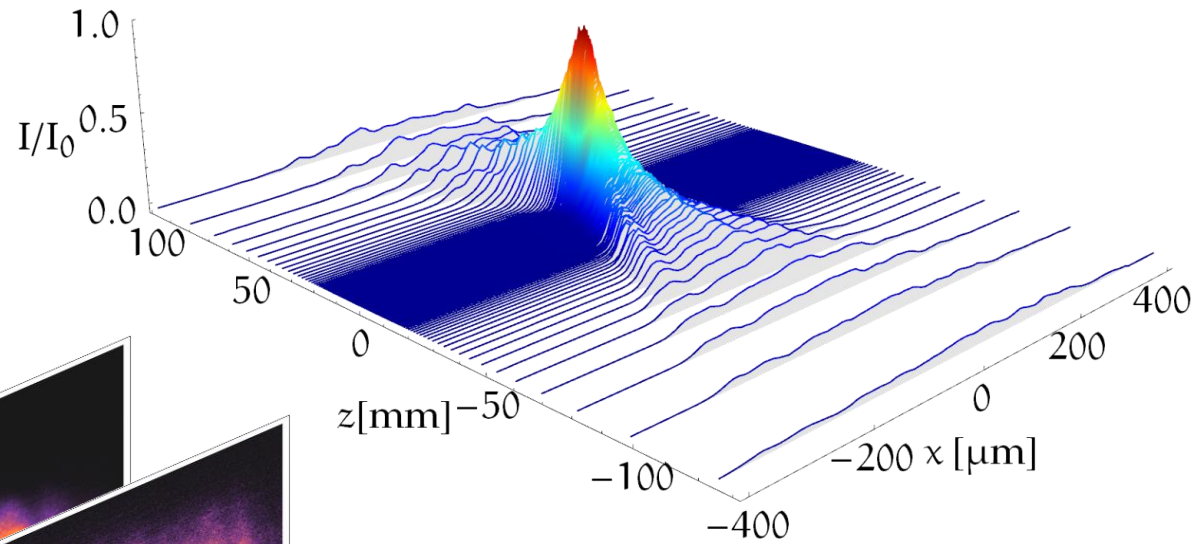
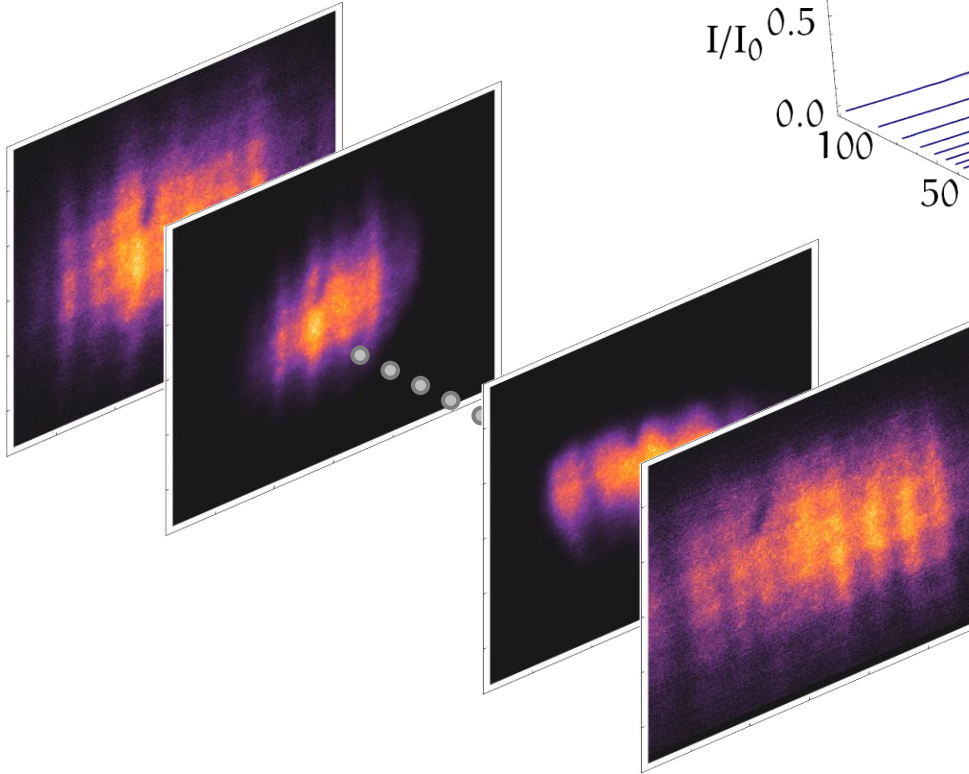
Caustic scan



Wigner distribution

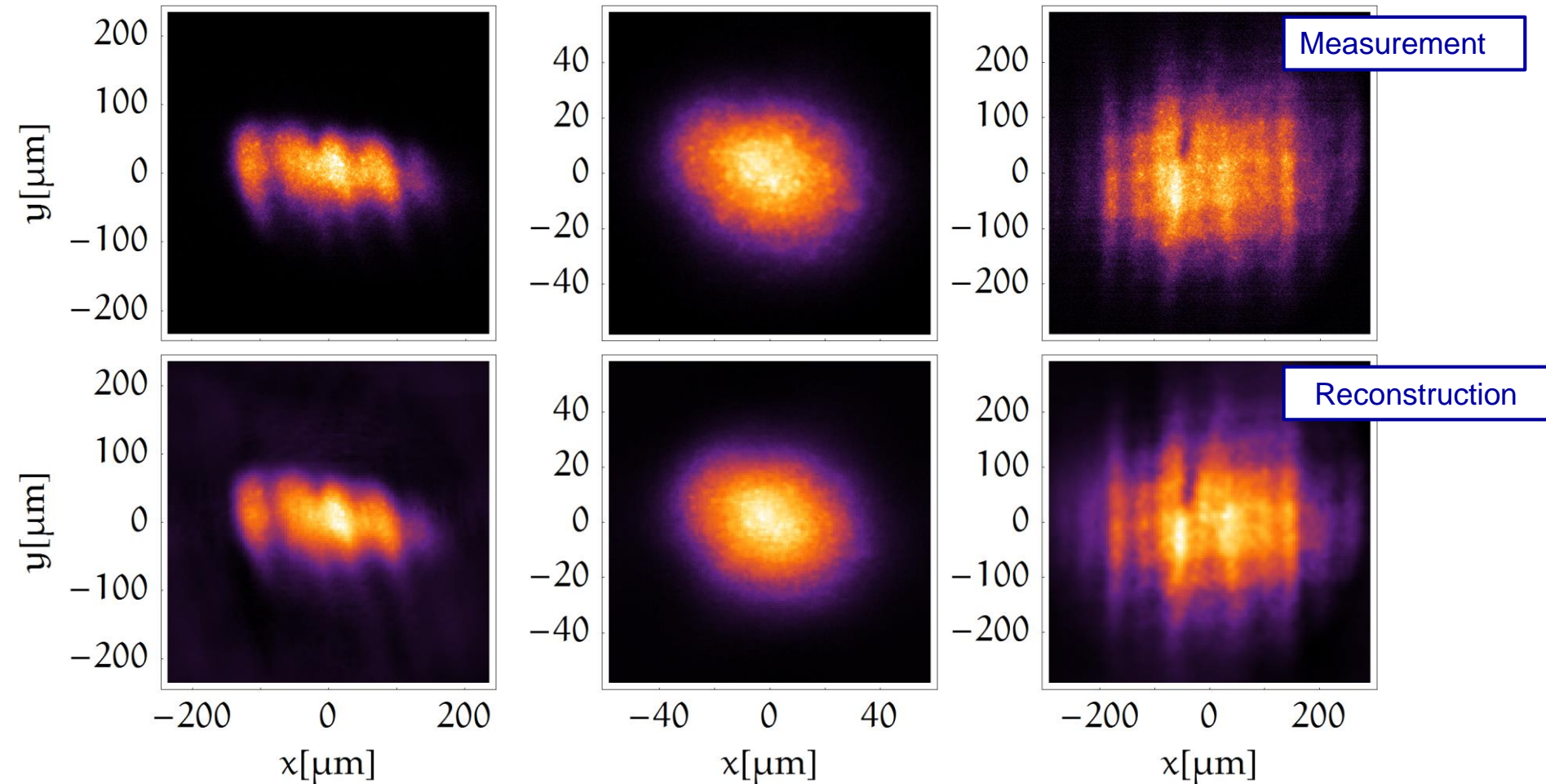
Projection-slice theorem [9]

$$\tilde{h}(q_x, z \cdot q_x) = \tilde{I}_z(q_x)$$

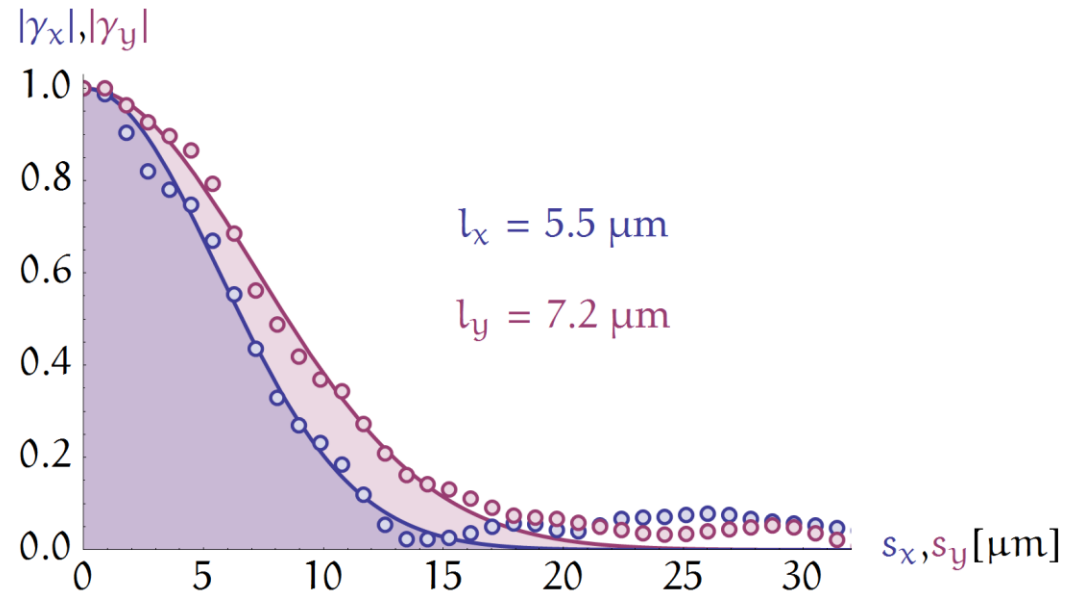
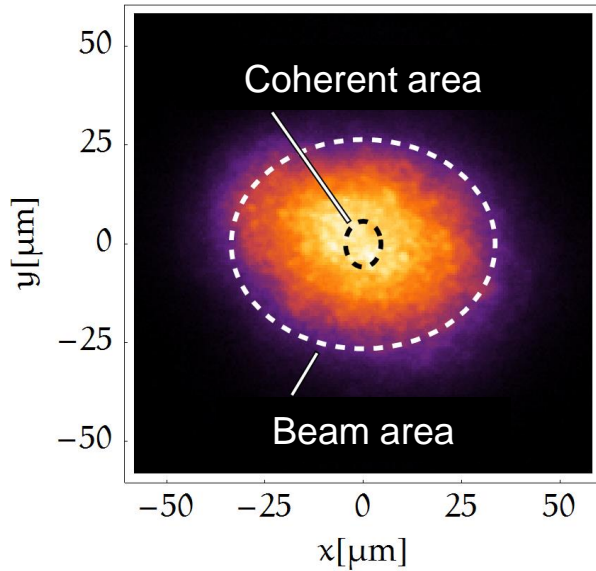


[9] A. Torre, *Linear ray and wave optics in phase space*, Elsevier B.V. Netherlands (2005)

Wigner distribution



Coherence properties



	Wavelength λ [nm]	Beam diameter d_x / d_y [μm]	Coherence length l_x / l_y [μm]	Global degree of coherence K
Wigner [10]	24.7	67 / 53	5.5 / 7.2	0.032
Double pinhole [7]	8.0	17 / 17	6.2 / 8.7	0.42

- [7] A. Singer *et al.*, "Spatial and temporal coherence properties of single free-electron laser pulses," Opt. Expr. **20**, 17480-17495 (2012)
 [10] T. Mey *et al.*, "Wigner distribution measurements of the spatial coherence properties of the free-electron laser FLASH," Opt. Expr. **22**, 16571-16584 (2014)

Thank you for your attention



Plasma Timing Tool

M. Harmand, R. Coffee, M.R. Bionta, M. Chollet, D. French, D. Zhu, D.M. Fritz, H.T. Lemke, N. Medvedev, B. Ziaja, S. Toleikis and M. Cammarata,
“Achieving few-femtosecond time-sorting at hard X-ray free-electron lasers”,
Nature Photon. 7, 215 (2013)

R. Riedel, A. Al-Shemmary, M. Gensch, T. Golz, M. Harmand, N. Medvedev, M.J. Prandolini, K. Sokolowski-Tinten, S. Toleikis, U. Wegner, B. Ziaja, N. Stojanovic and F. Tavella,
“Single-shot pulse duration monitor for extreme ultraviolet and X-ray free-electron lasers”,
Nature Comm. 4, 1731 (2013)

Experimental setup of spatial encoding tool

Ultrafast plasma switch (a)

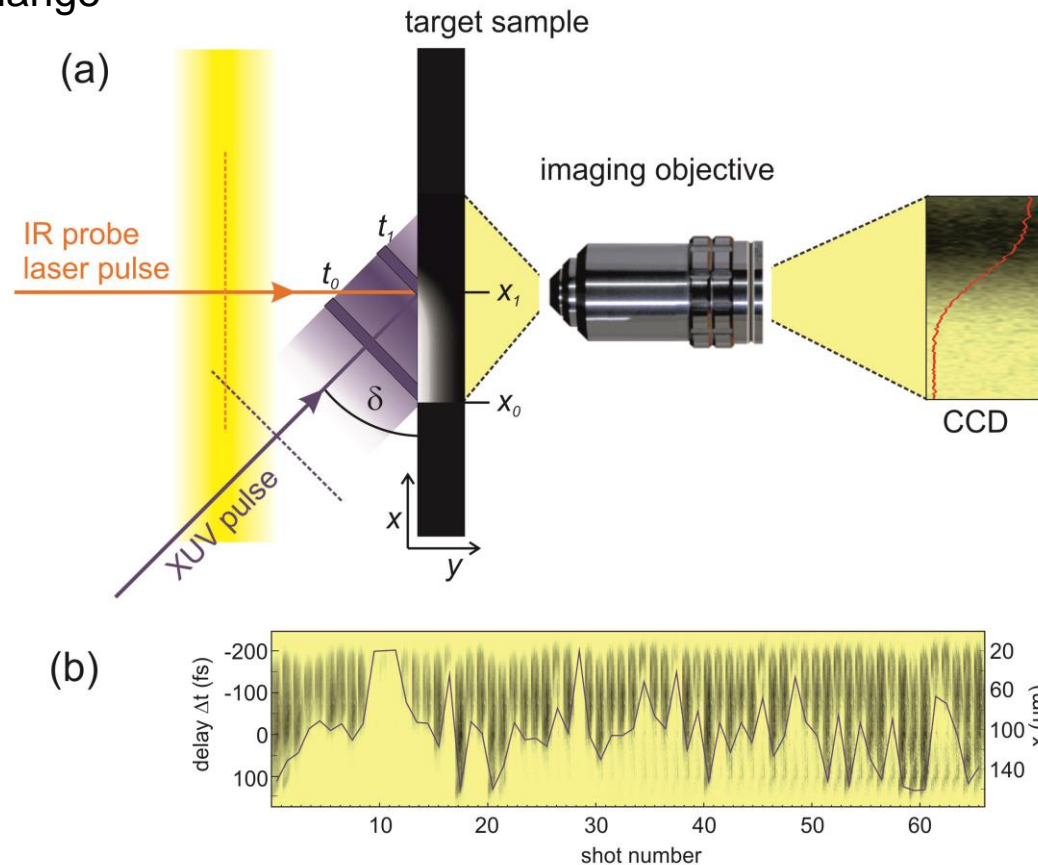
- FEL triggers reflectivity/transmission change
- oblique angle of incidence δ
- different parts of FEL wavefront reach sample at different positions
- time axis spatially resolved:

$$\Delta t = \Delta_x \cdot \cos(\delta) / c$$

(Δ_x : imaged pixel size, c : vacuum speed of light)

→ extract arrival time (b)

→ extract pulse duration



Experimental set-up at XPP (LCLS)

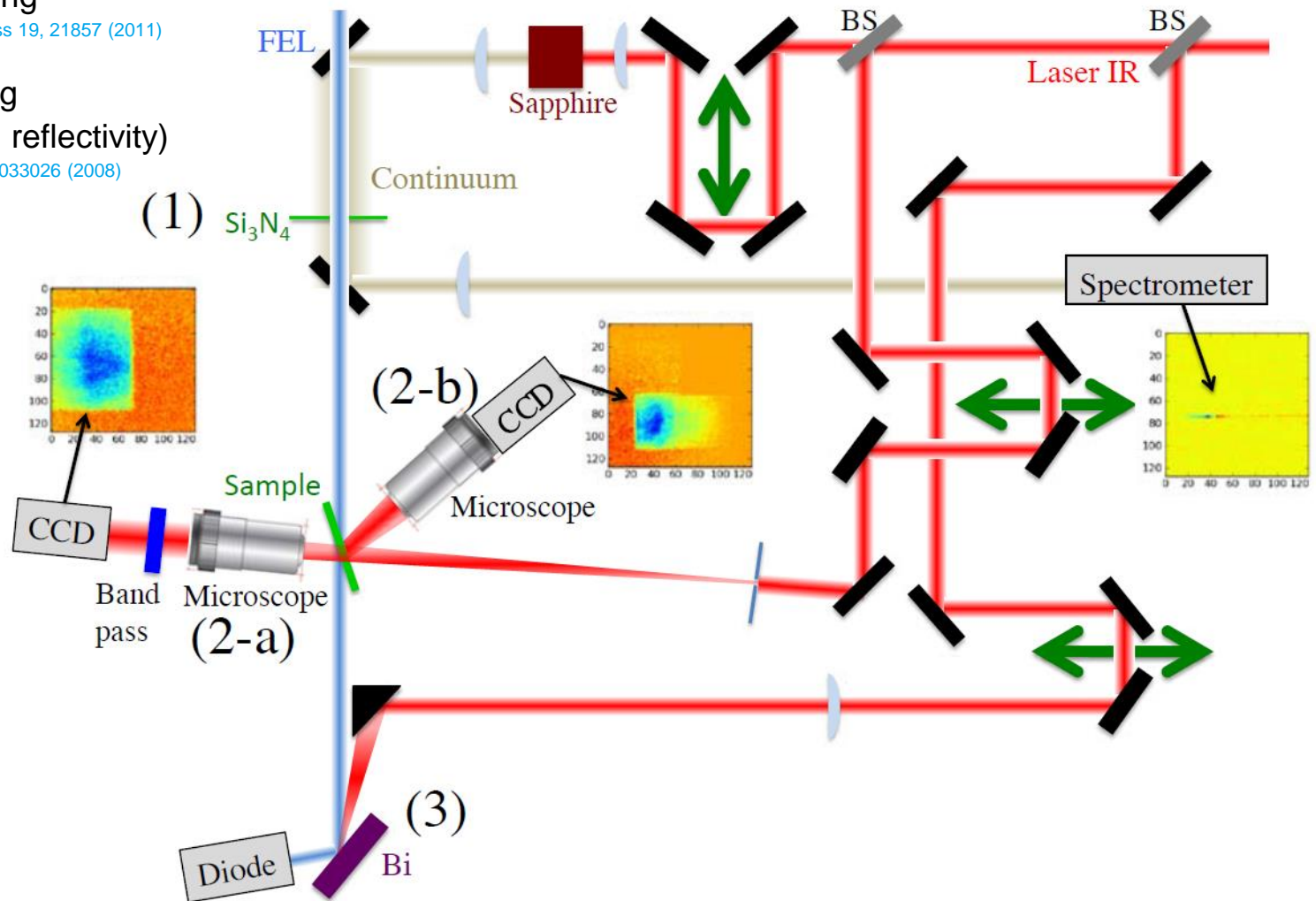
- (1) Spectral encoding

M.R. Bionta et al., *Optical Express* 19, 21857 (2011)

- (2) Spatial encoding

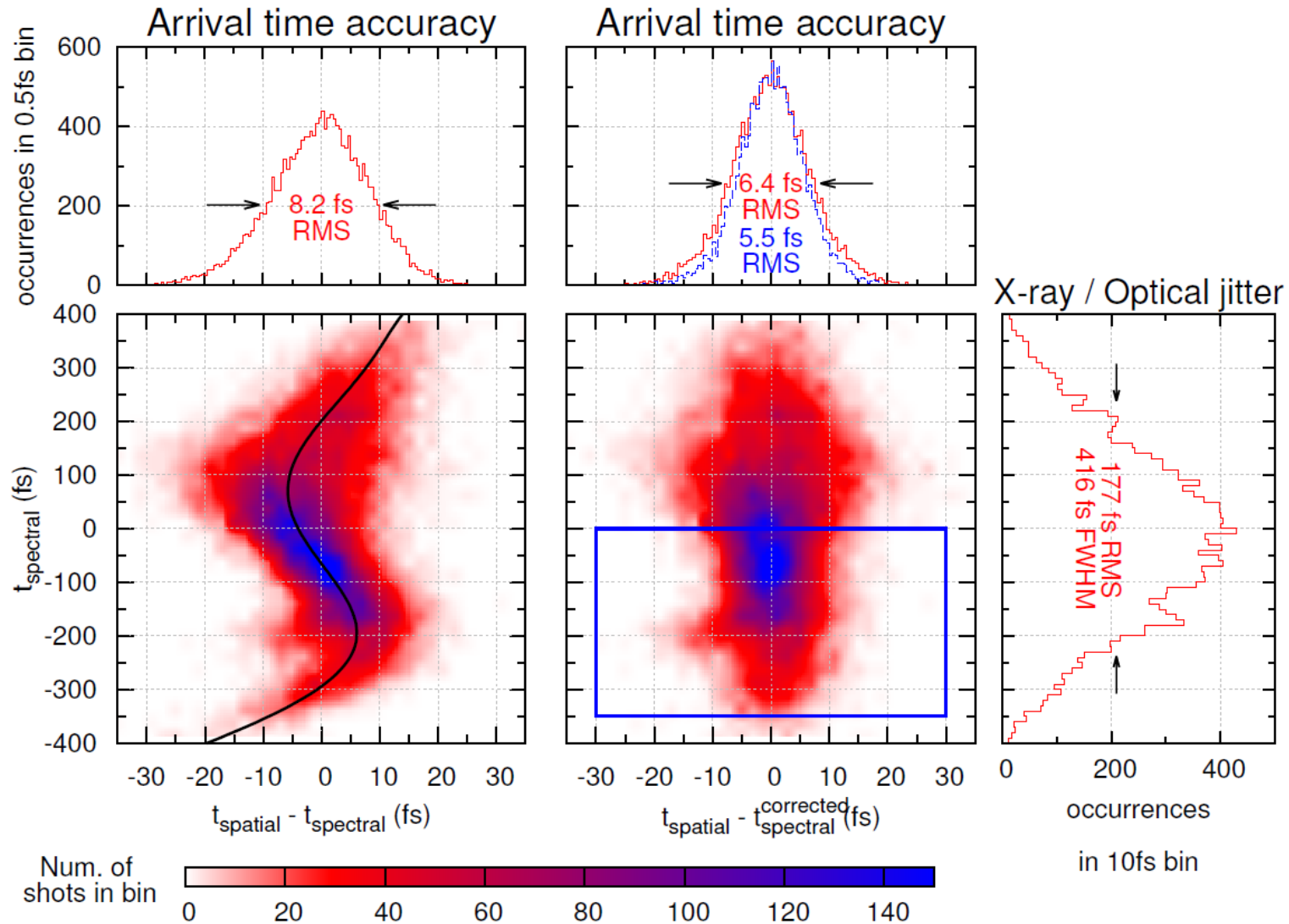
(a: transmission, b: reflectivity)

T. Maltezopoulos et al., *NJP* 10, 033026 (2008)

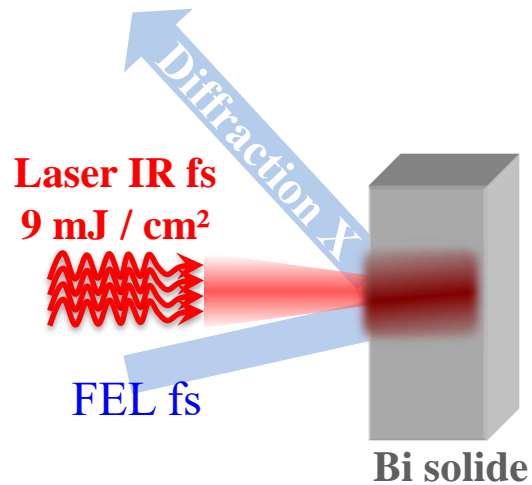


Diagnostic correlation of LCLS experiment at XPP

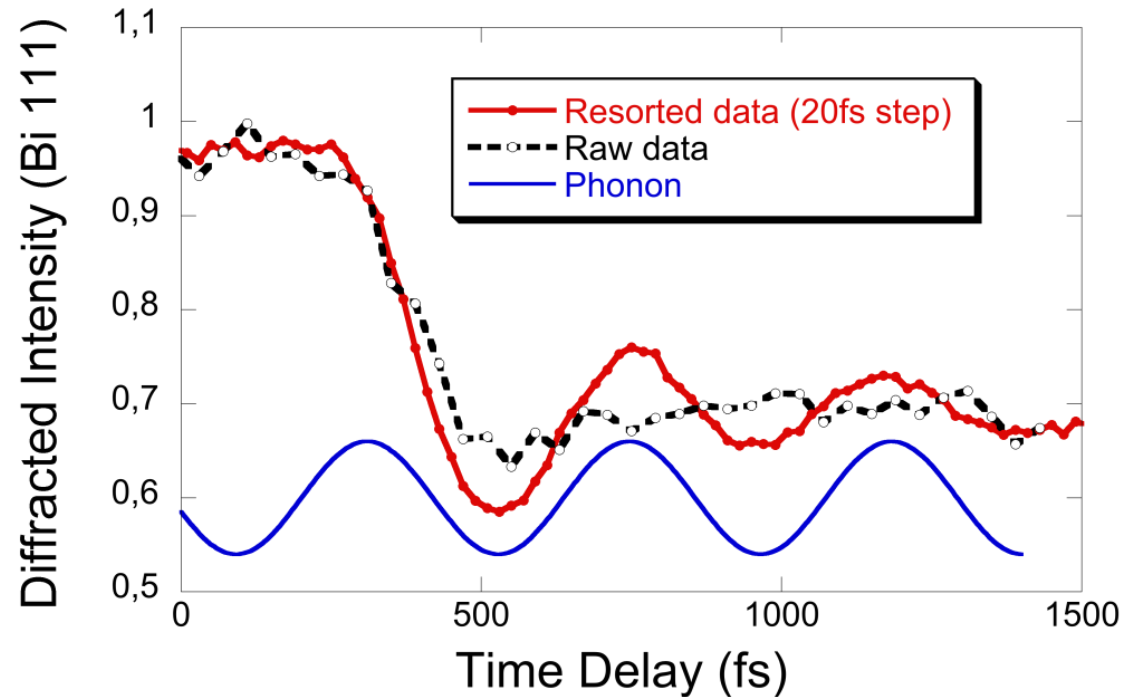
Spectral vs. Spatial (transmission)



Experimental test on Bi



fs atomic displacements generated in Bi samples, in response to ultrafast photoexcitation by an optical laser



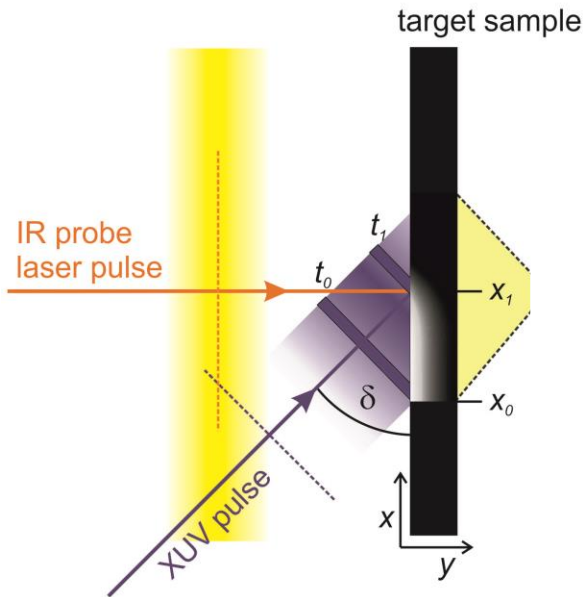
- by resorting data according to their arrival time ultrafast processes can be resolved

D.M Fritz et al., Science 315, 633 (2007)

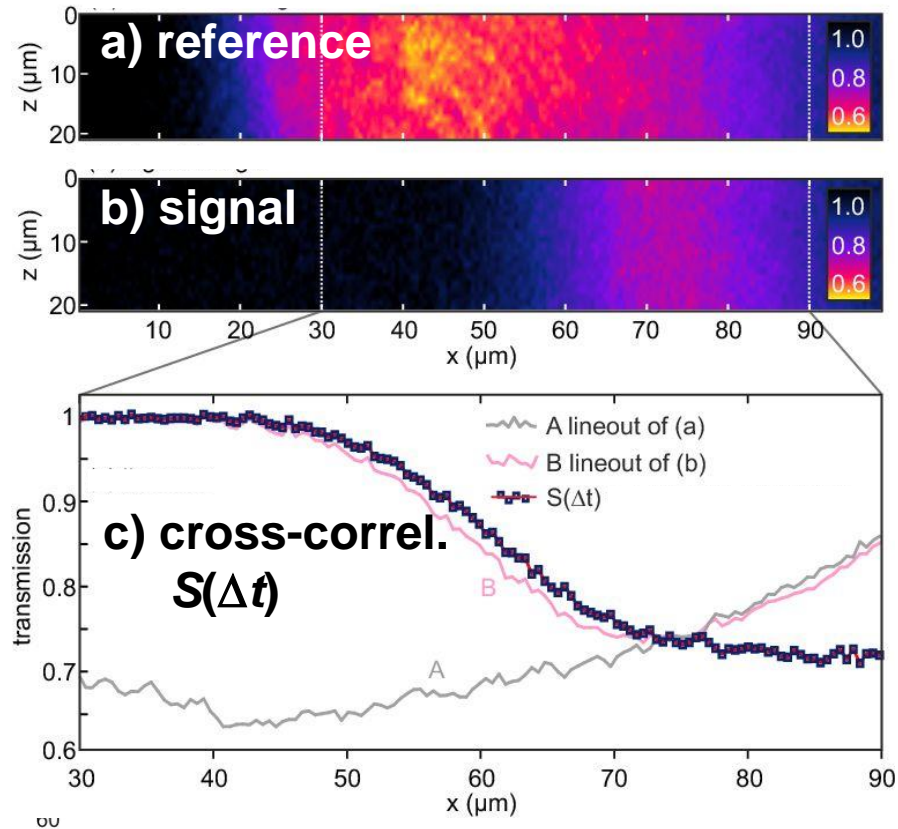
K. Sokolowski-Tinten et al., Nature Phys. 422, 287 (2003)

Signal processing for optical-FEL cross correlation

- measured transmission change



- a) probe pulse arrives after XUV pulse
- b) probe pulse arrives simultaneously
- c) cross-correlation
(corrected for FEL beam profile)



Data analysis

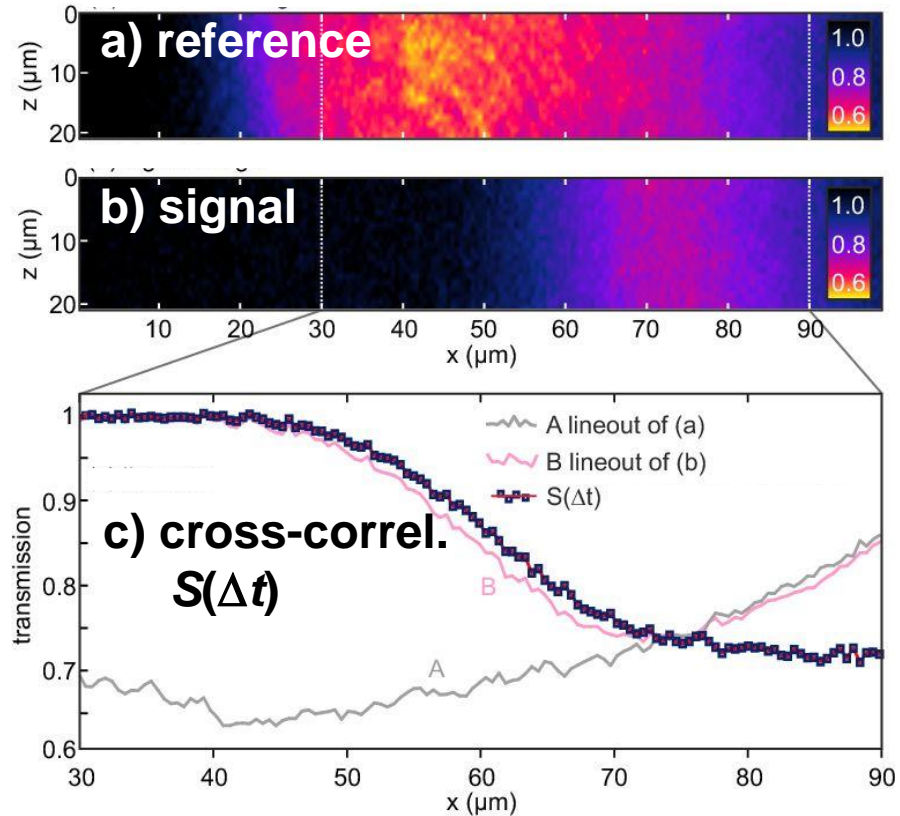
- **Cross-correlation function**

$$S(\Delta t) = \int_{-\infty}^{+\infty} I_{\text{Laser}}(t) G(t - \Delta t) dt$$

measured measured study
Gaussian (64±7) fs
at 800 nm

- arrival time directly obtained from $S(\Delta t)$
- $G(t)$ ultrafast transmission change
- contains information about FEL intensity

$$G(t) = G[I_{\text{FEL}}(t)]$$



Data analysis and results

- FLASH pulse durations at different wavelengths (fused silica sample)

FEL wavelength: 41.5 nm

FEL pulse energy: $(84 \pm 4) \mu\text{J}$

Bunch charge: 0.50 nC

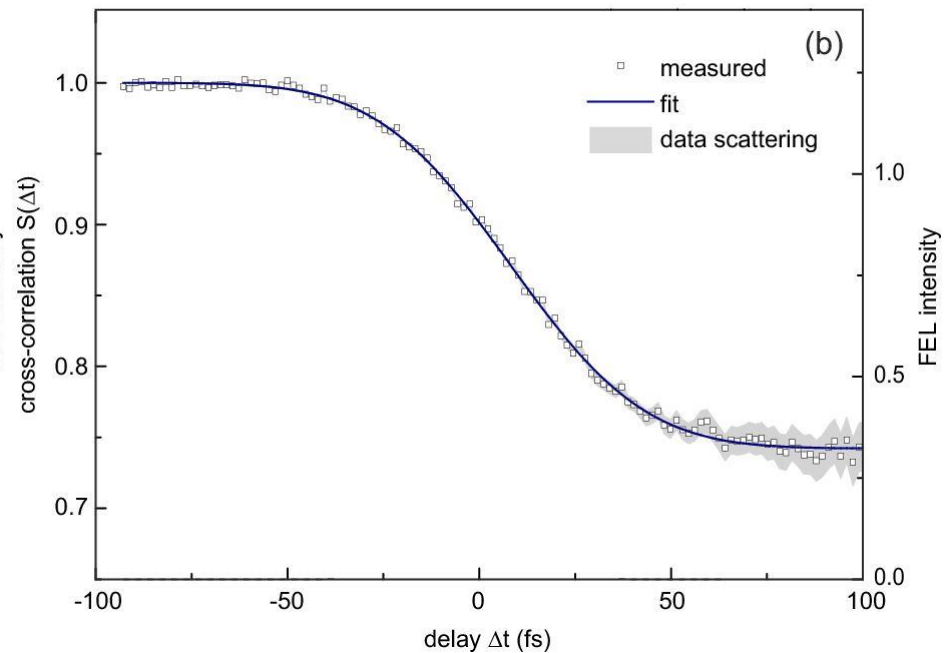
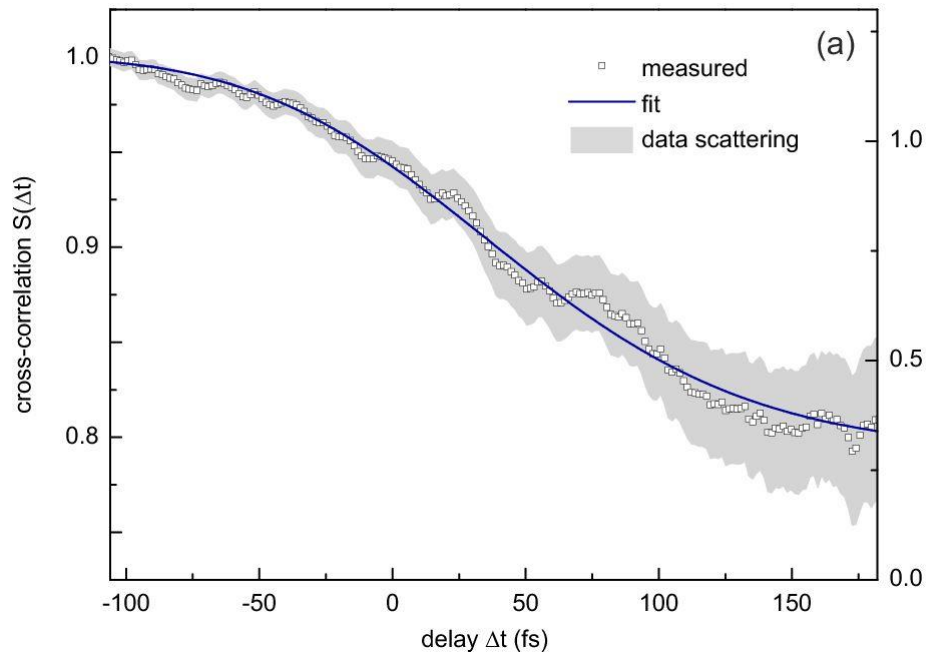
Pulse duration: $(184 \pm 14) \text{ fs}$

FEL wavelength: 5.5 nm

FEL pulse energy: $(29 \pm 10) \mu\text{J}$

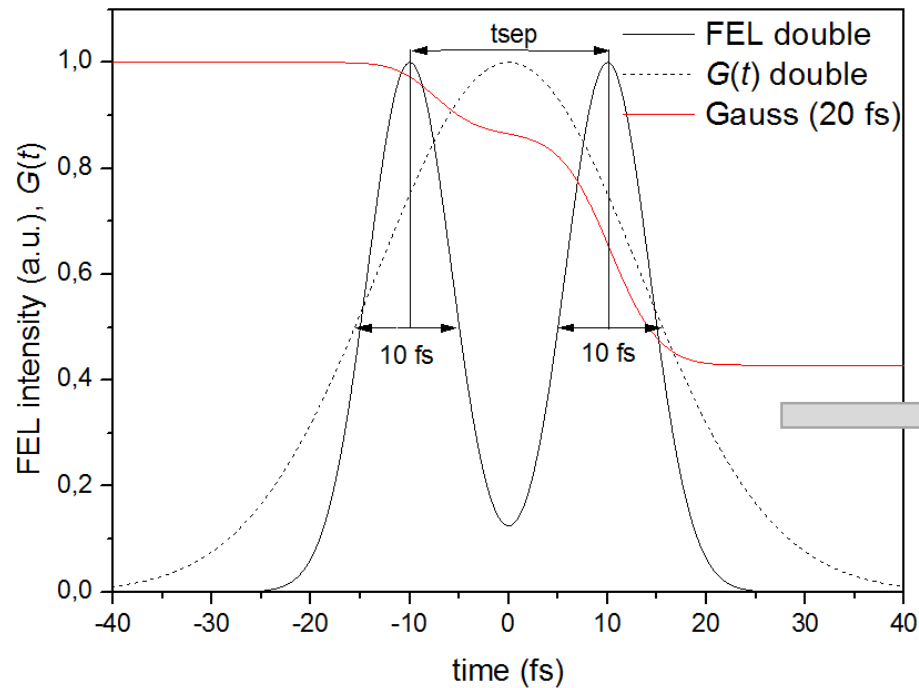
Bunch charge: 0.25 nC

Pulse duration: $(21 \pm 19) \text{ fs}$

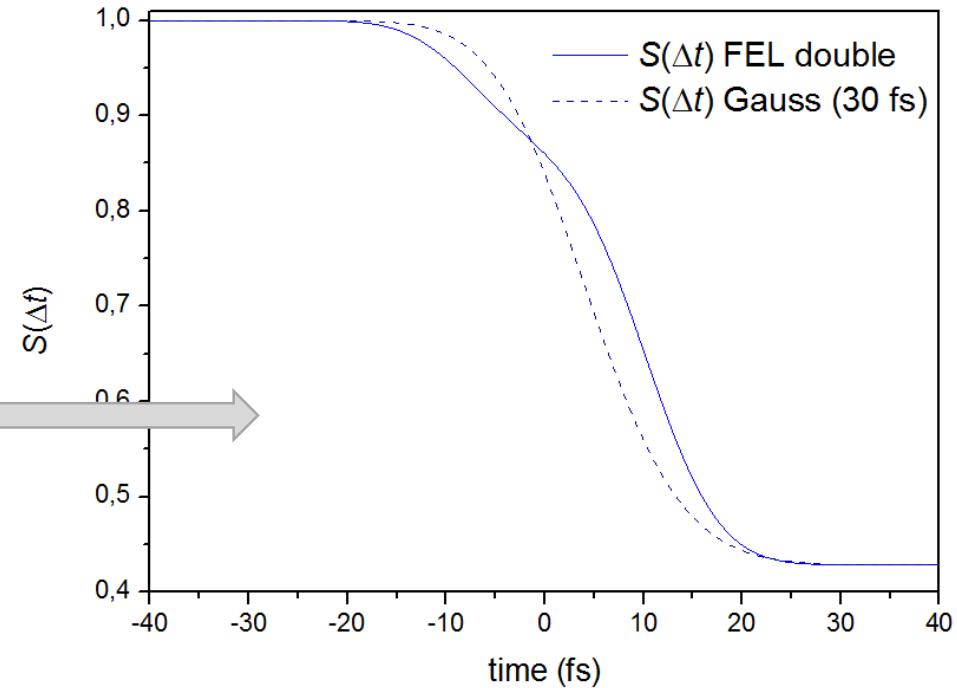


FEL double pulses

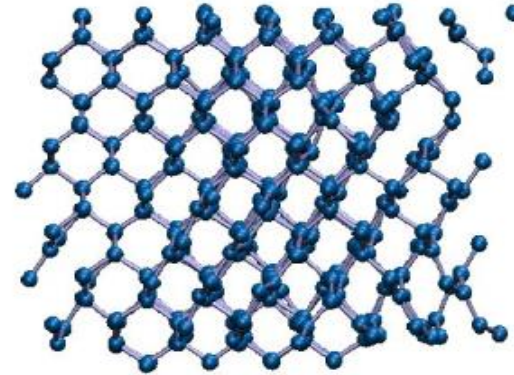
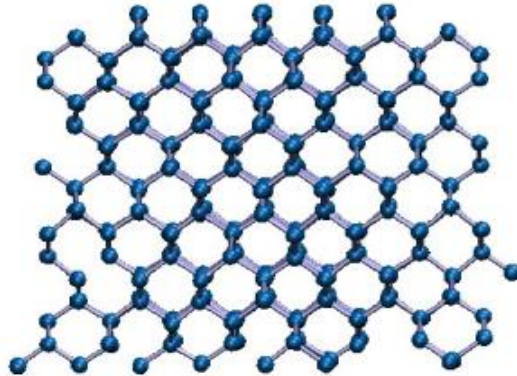
- Example: FEL double pulses (2x 10 fs)
(separated by 20 fs)



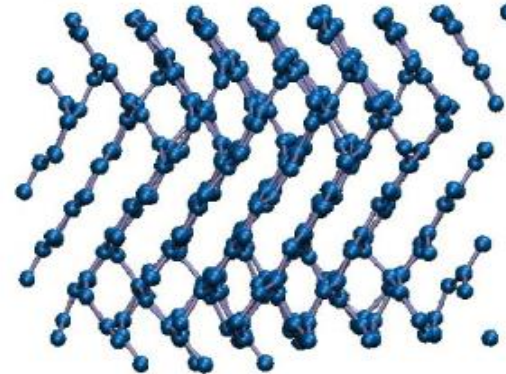
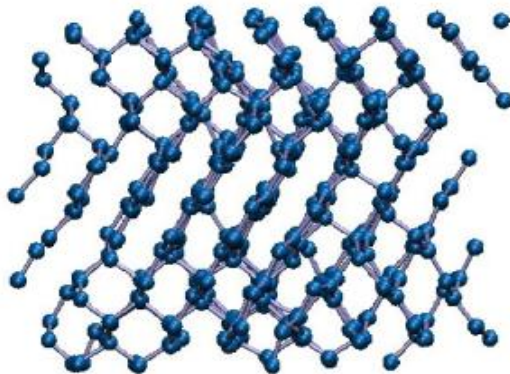
$S(\Delta t)$ with 10 fs probe laser



Ultra-fast solid to solid phase transition in diamond.



Sven Toleikis



Nonthermal graphitization of diamond induced by a femtosecond x-ray laser pulse

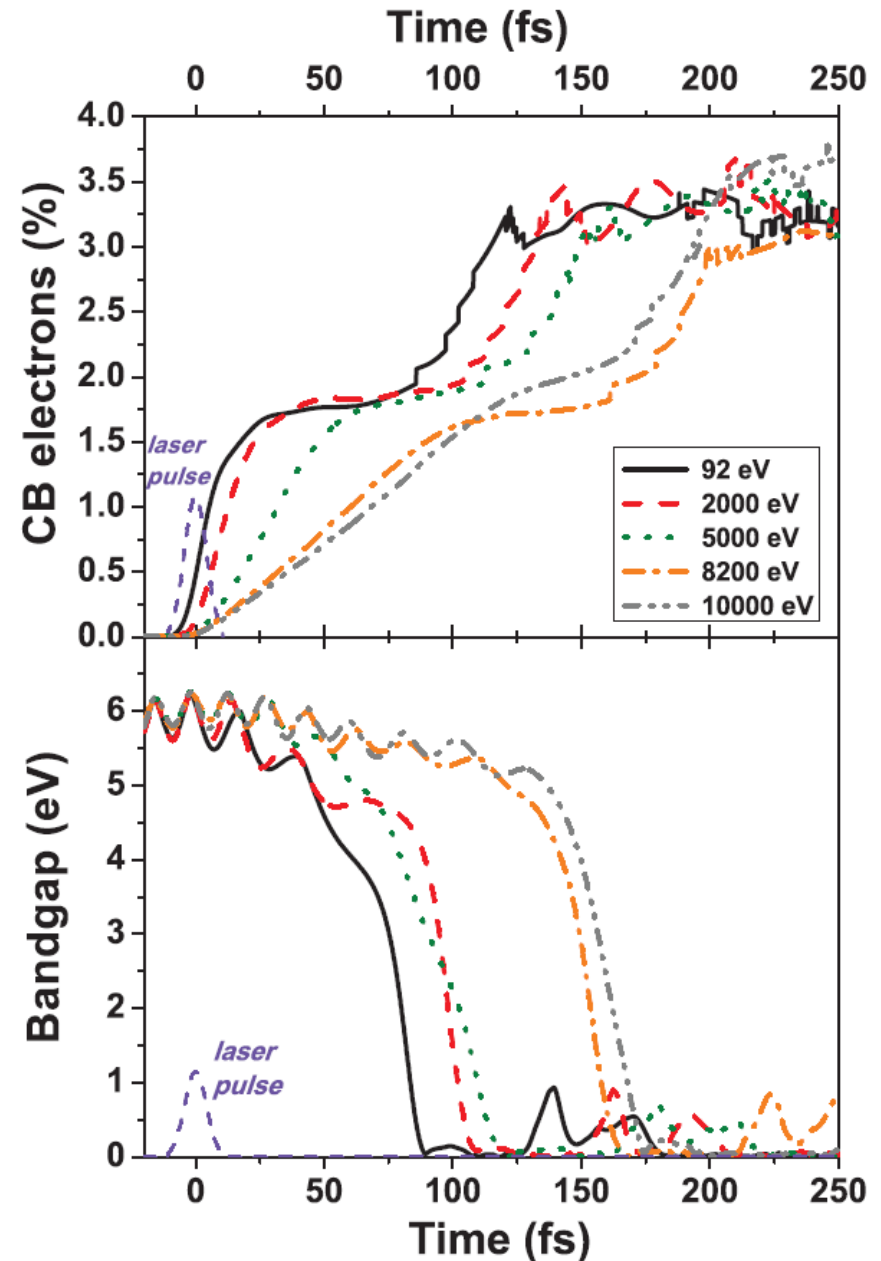
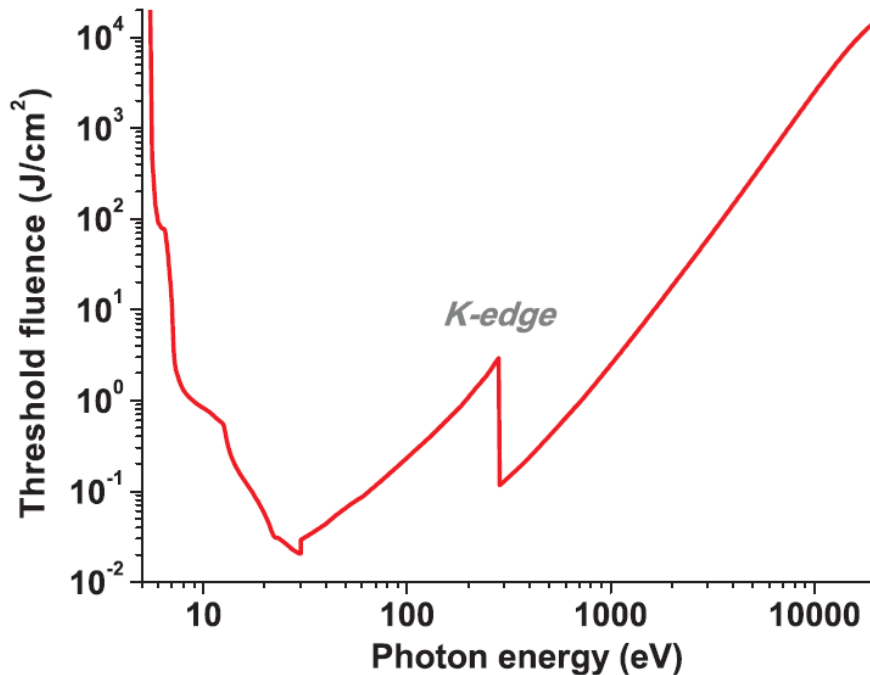
- N. Medvedev, H.O. Jeschke, and B. Ziaja; Phys. Rev. B **88**, 224304 (2013)
- Mechanism:
short x-ray pulses -> photoionization
-> photoelectrons -> impact ionization -> secondary e^-
-> K-shell / L-shell holes -> Auger decays -> impact ionization -> secondary e^-
-> further cascading
- Hybrid model described in:
N. Medvedev, H. O. Jeschke, and B. Ziaja, New J. Phys. **15**, 015016 (2013)

Combines different theoretical approaches:

- MC method to describe photoabsorption and Auger decays + transient nonequilibrium kinetics of high energy electrons and their secondary cascading
-> transient electron distribution:
low-energy Fermi-like distribution (e^- temperature equation) + high-energy electrons
- The potential energy surface, the collective forces acting on each atom, and the transient electronic band structure are calculated by diagonalizing a tight-binding (TB) Hamiltonian
- Atom dynamics is followed by classical molecular dynamics (MD)

Results / Prediction for diamond

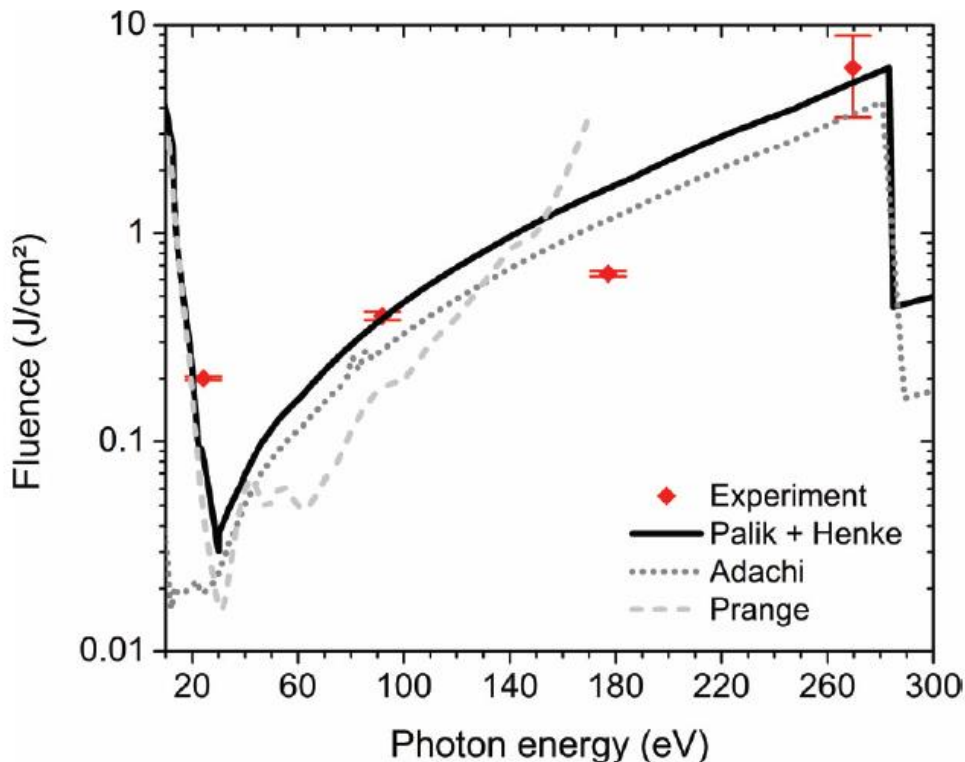
- Damage threshold fluence for a broad photon energy range is always **$\sim 0.7 \text{ eV / atom}$**
- The x-rays induces an ultrafast non-thermal phase transition:
nonthermal graphitization of diamond



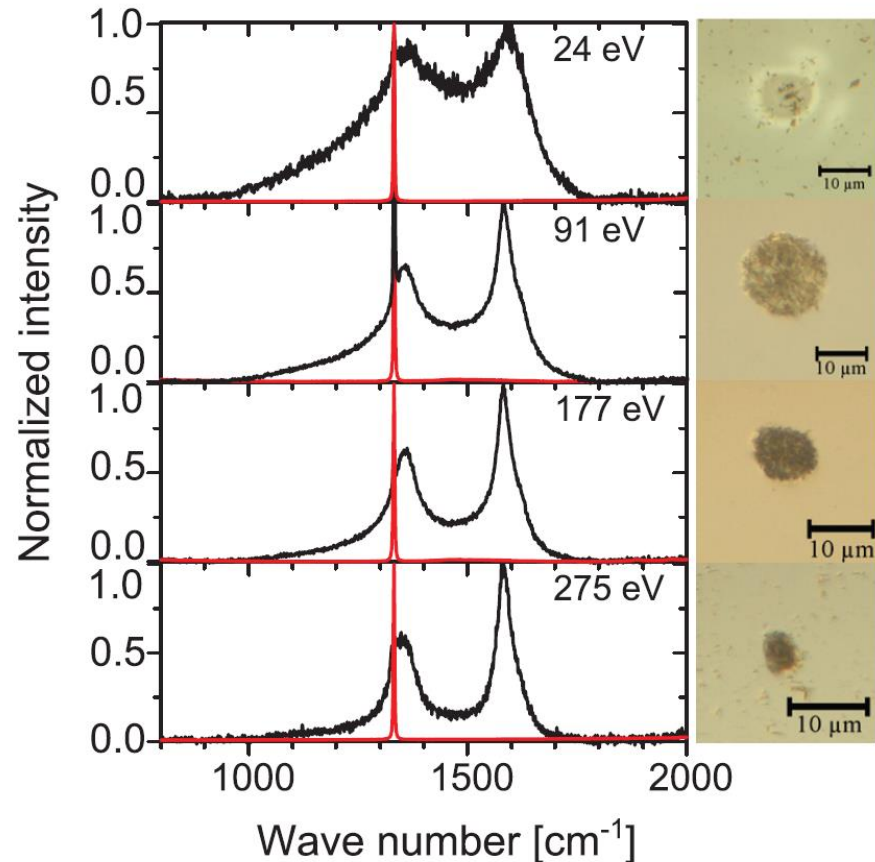
Damage experiment @ FLASH

- J. Gaudin, N. Medvedev, J. Chalupsky, T. Burian, S. Dastjani-Farahani, V. Hajkova, M. Harmand, H.O. Jeschke, L. Juha, M. Jurek, D. Klinger, J. Krzywinski, R.A. Loch, S. Moeller, M. Nagasono, C. Ozkan, K. Saksl, H. Sinn, R. Sobierajski, P. Sovak, S. Toleikis, K. Tiedtke, M. Toufarova, T. Tschentscher, V. Vorliceck, L. Vysin, H. Wabnitz, and B. Ziaja;
Phys Rev. B **88**, 060101(R) (2013)

Fluence treshold for graphitization



Post-mortem micro-Raman spectra of the diamond samples

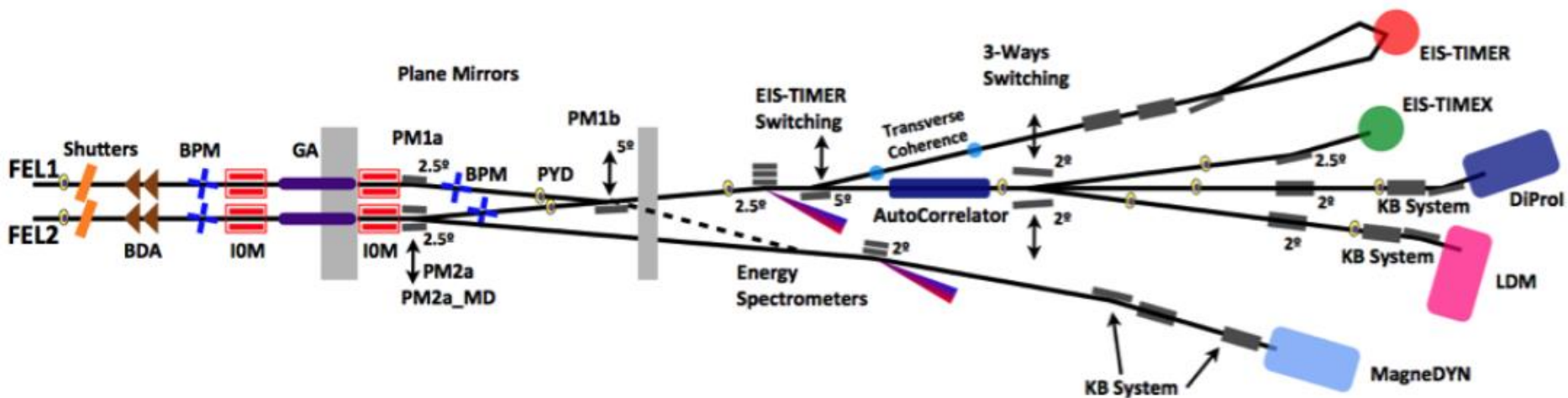


Experiment @ FERMI (a seeded XUV FEL)

F. Tavella, H. Höppner, V. Tkachenko, N. Medvedev, F. Capotondi, T. Golz, Y. Kai, M. Manfredda, E. Pedersoli, M.J. Prandolini, N. Stojanovic, T. Tanikawa, U. Teubner, S. Toleikis, and B. Ziaja,

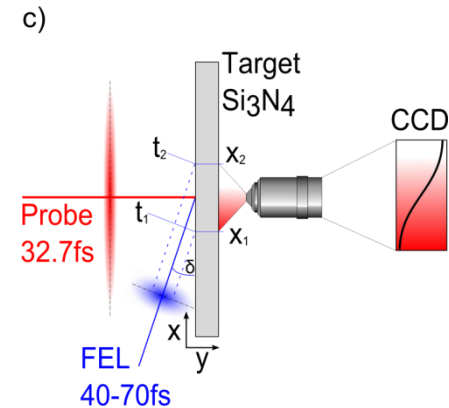
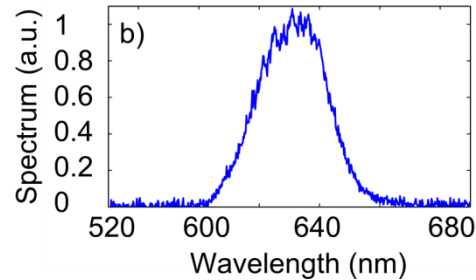
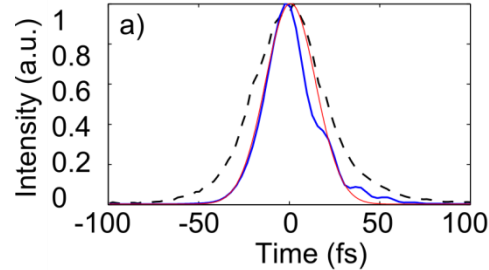
Soft x-rays induced femtosecond solid-to-solid phase transition

High Energy Density Physics 24, 22 (2017)

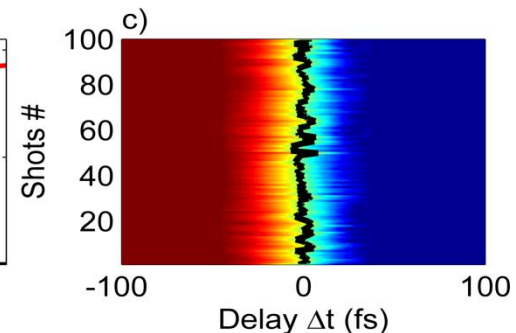
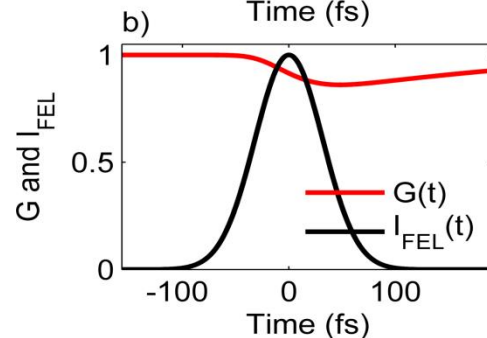
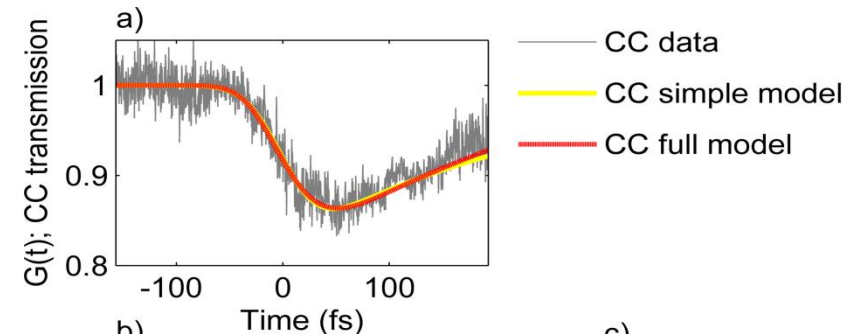


Timing tool @ FERMI

- Experimental setup @ FERMI (c)
- Using own NOPA driven by the seed laser: 32.7 fs (a), 630 nm (b), 300 nJ on target
- Spatial time resolution (of the imaging system and the angle δ): ~ 4 fs
- XUV beam spot size on target: $150 \times 30 \mu\text{m}^2$
- Non-damaging mode

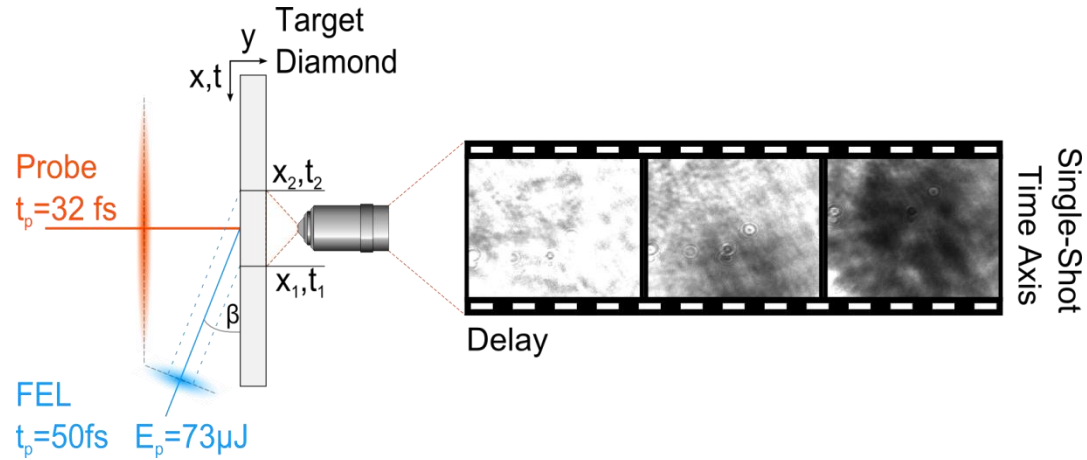


- Data analysis of a single-shot cross correlation (CC) (a)
- FEL at 26.17 nm (47.38 eV)
- Retrieved gating function $G(t)$ and single-shot pulse structure $I_{\text{FEL}}(t)$ (b):
➔ **single-shot pulse duration: 74.9 fs**
- Jitter analysis for 100 shots (c):
arrival time jitter: 2.2 fs (rms)

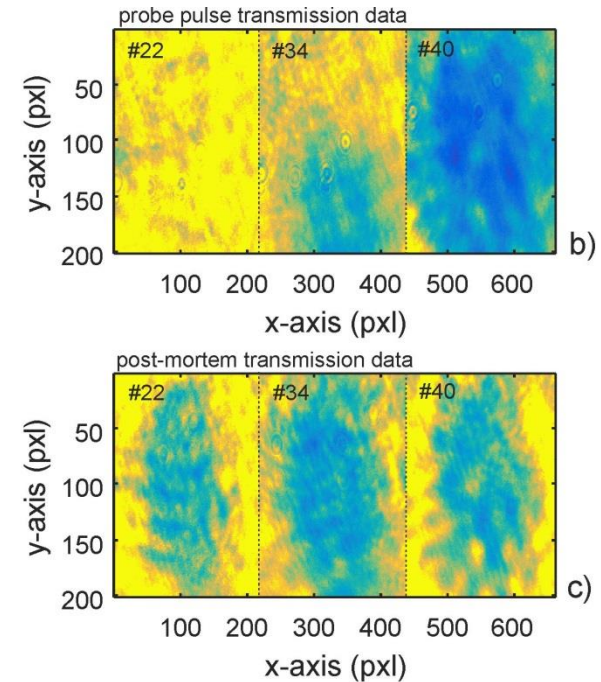
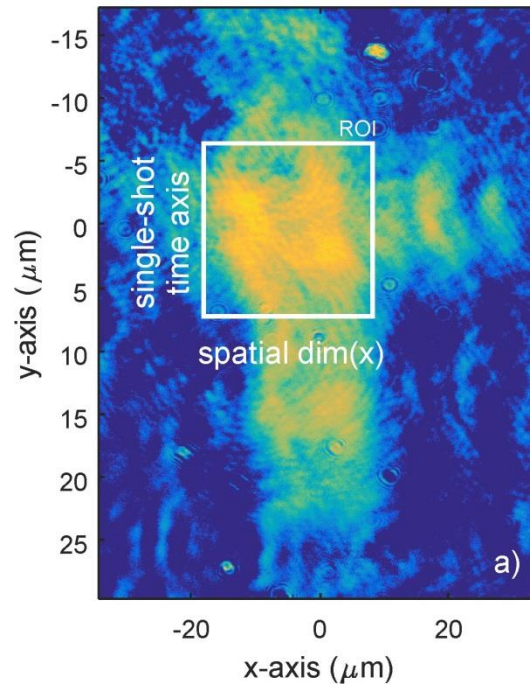


Ultra-fast solid to solid phase transition in diamond

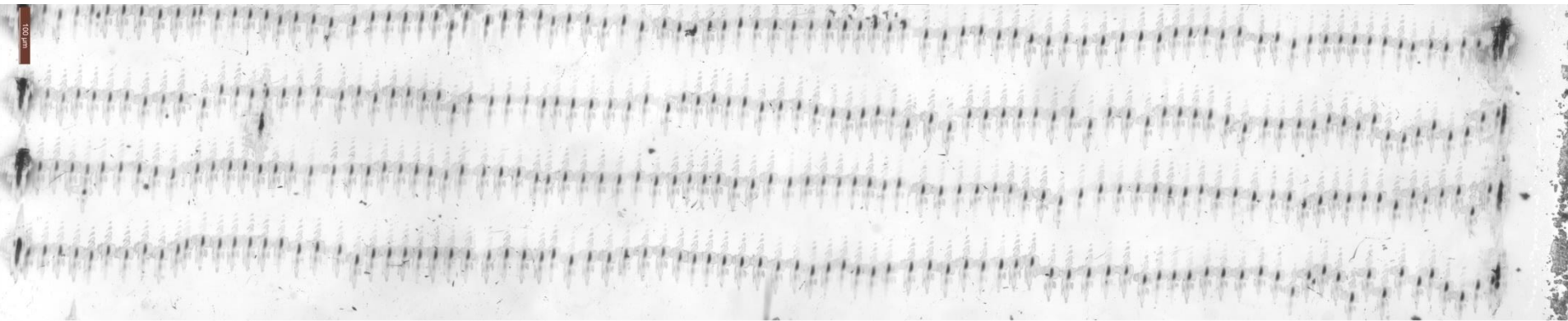
- Setup @ DiProl beamline
- Target: 300 μm thick poly-crystalline CVD diamond
- XUV beam spot size on target: $17 \times 7.5 \mu\text{m}^2$
- FEL wavelength: 26.17 nm
- FEL pulse length: 52.5(3.4) fs



- Single shot raw data example
- Scanning time delay in 10 fs steps
- Shot #22: probe pulse hits target prior to FEL pulse
- Shot #34: probe pulse hits the target at the same time as the FEL pulse
- Shot #40: probe pulse hits the target after the FEL pulse



Scanning the target and shooting the diamond target with 10 Hz

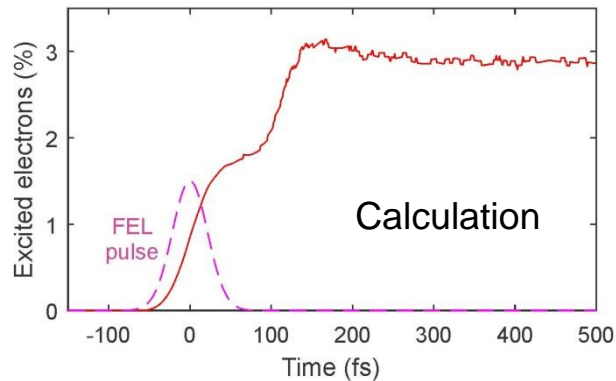


Ultra-fast solid to solid phase transition in diamond

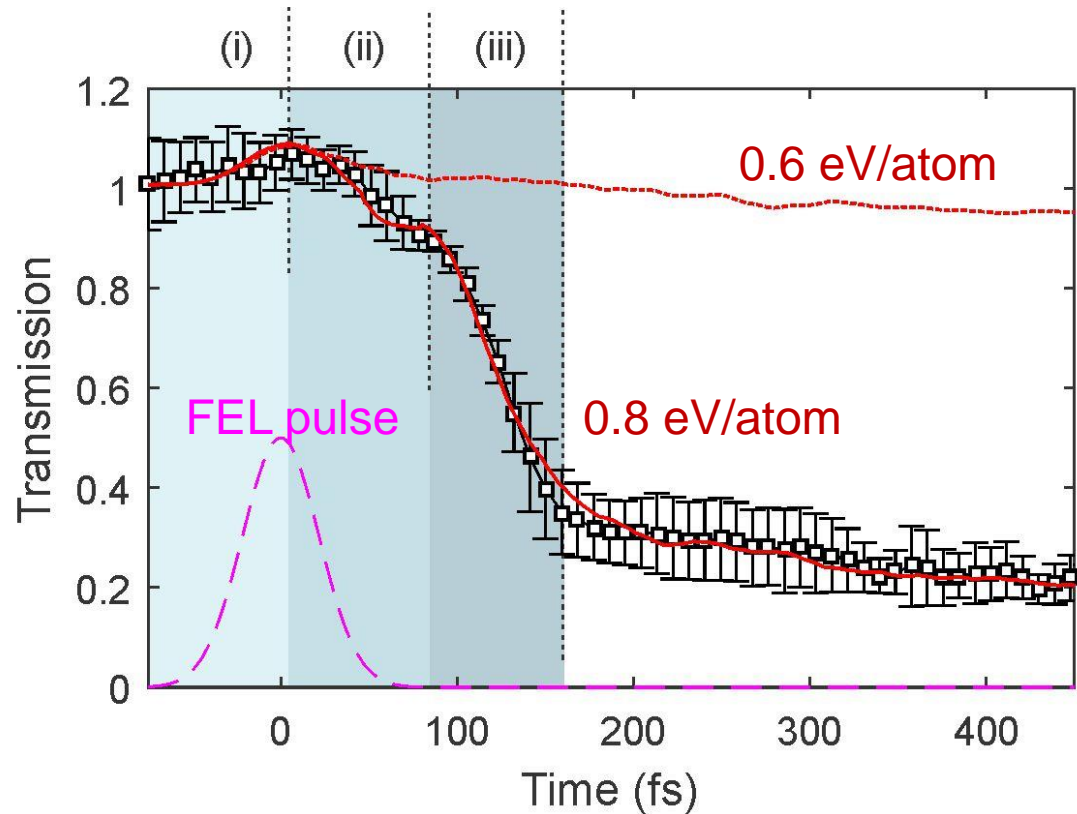
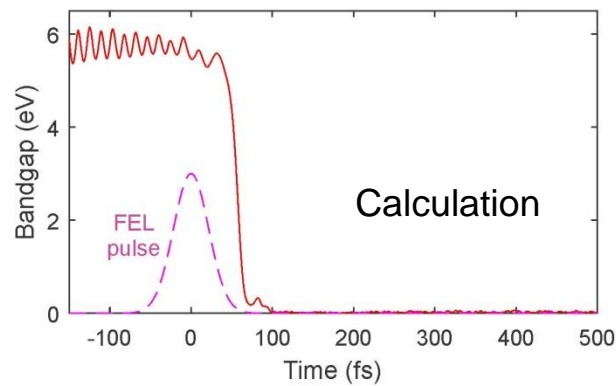
- Transient optical transmission at ~ 630 nm from a diamond sample irradiated with XUV

➤ Non-thermal graphitization in three steps:

➤ (i) initial electronic excitation to the conduction band

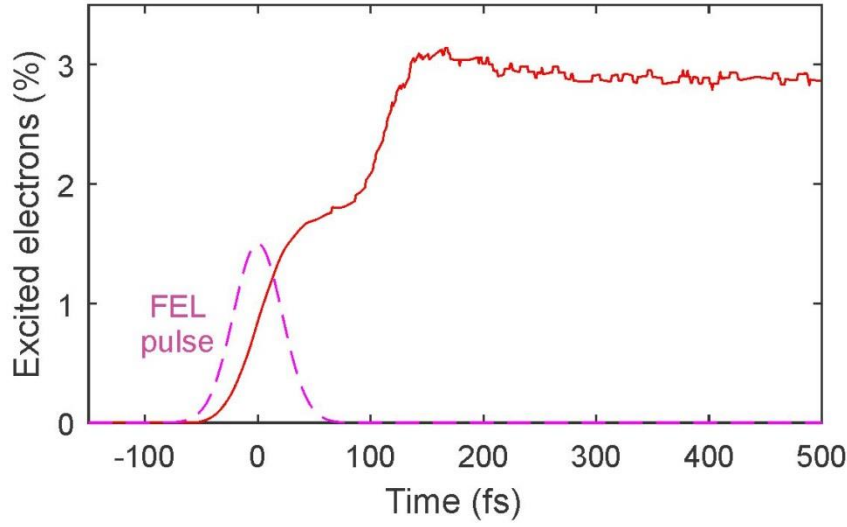
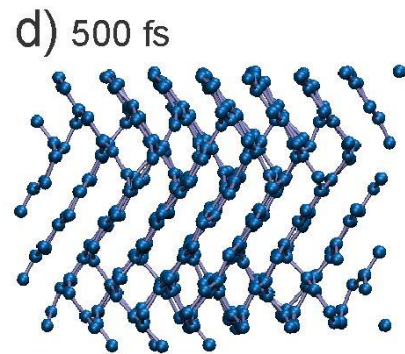
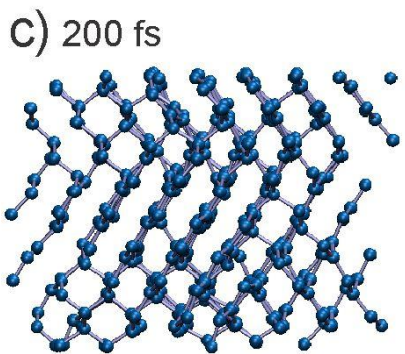
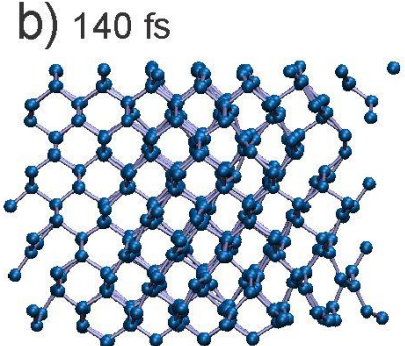
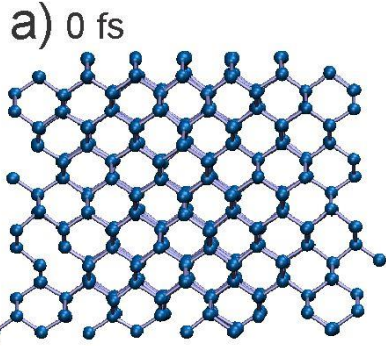
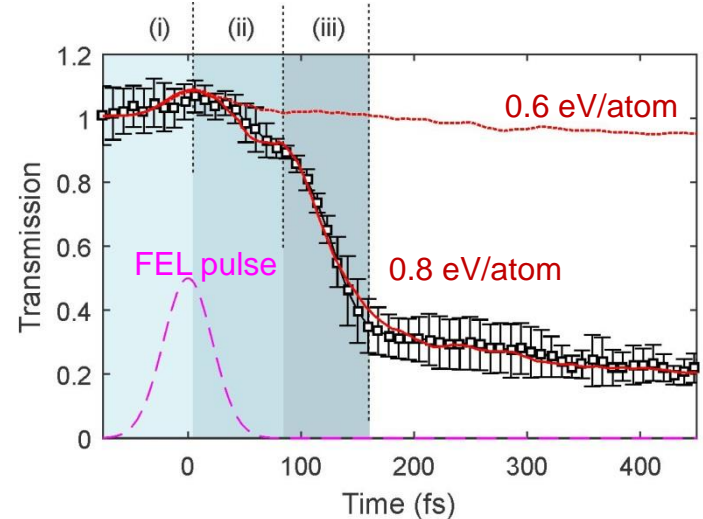


➤ (ii) electronic excitation triggers band gap collapse



Ultra-fast solid to solid phase transition in diamond

- > (iii) Significant decrease of transmission
Atomic relocation (from ~140 fs on)
Changing the material properties from insulating diamond to semi-metallic graphite
Further increase of electron density in CB
- > Final atomic relocation in overdense graphite with broken plane orientations

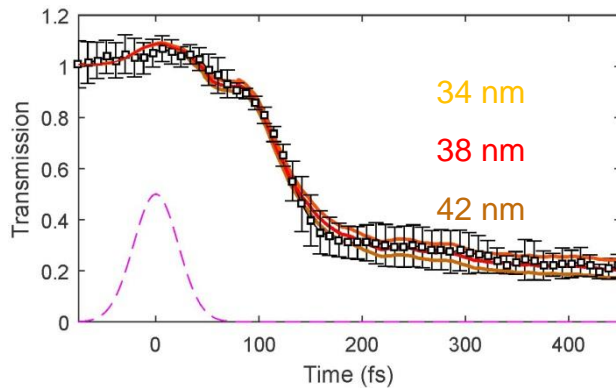
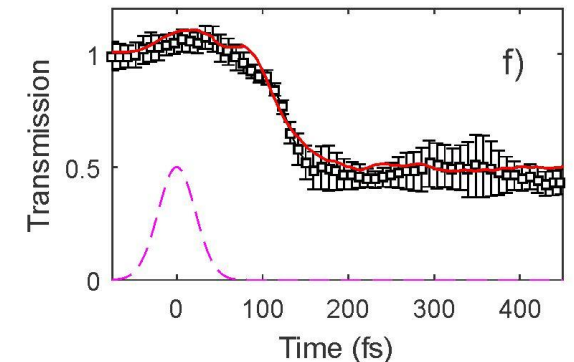
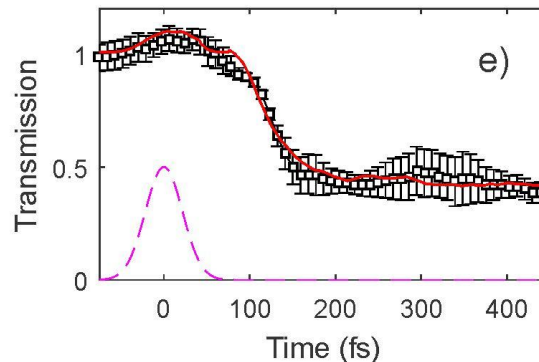
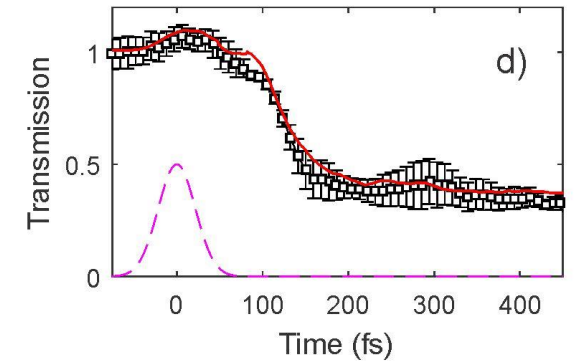
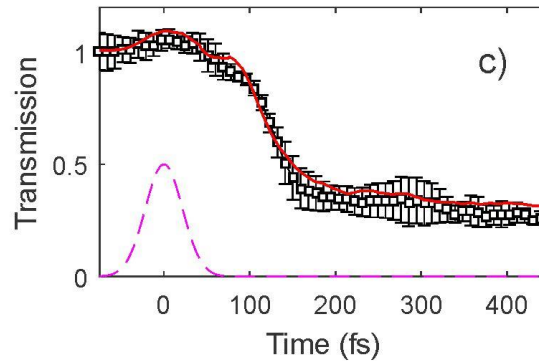
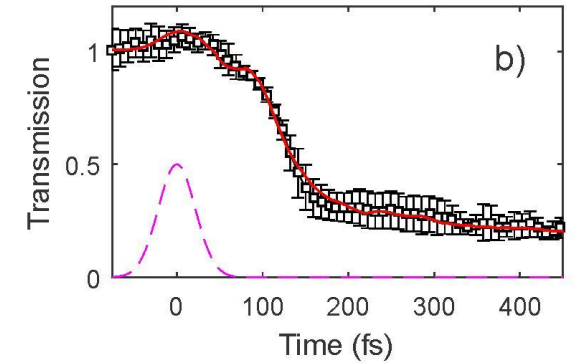
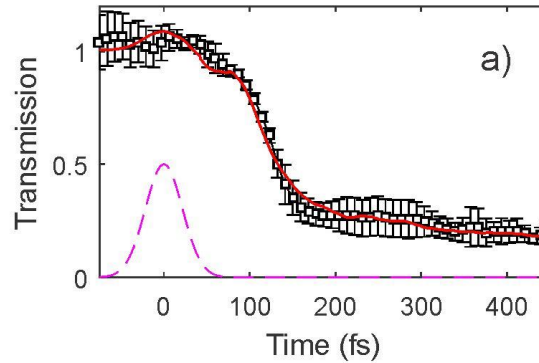


Ultra-fast solid to solid phase transition in diamond

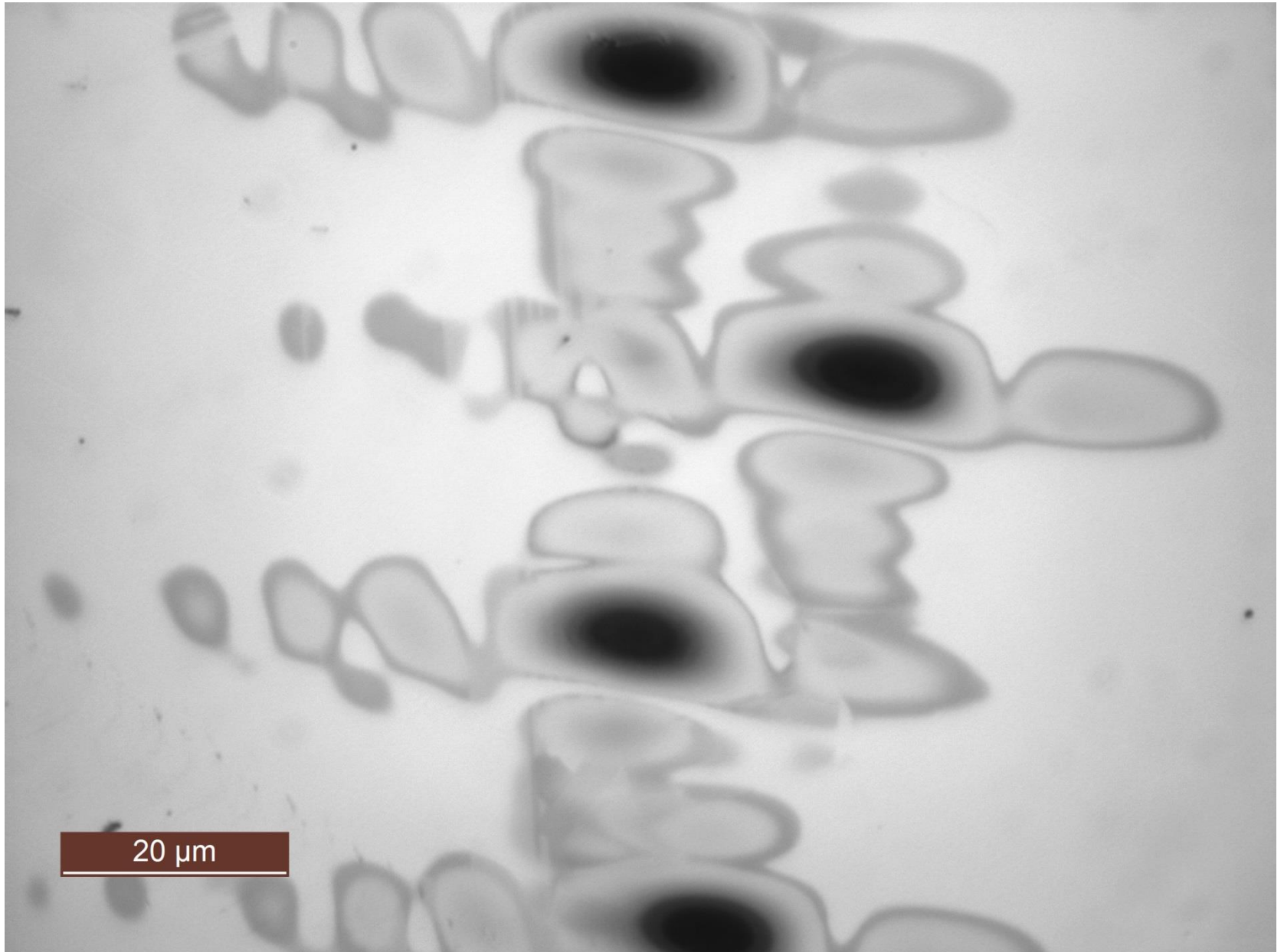
- Temporal behavior is independent from the fluence (if above 0.7 eV/atom)

➤ Transient graphite layer thickness at $t=400$ fs:

- a) 40 nm
- b) 38 nm
- c) 29 nm
- d) 25 nm
- e) 22 nm
- f) 17 nm

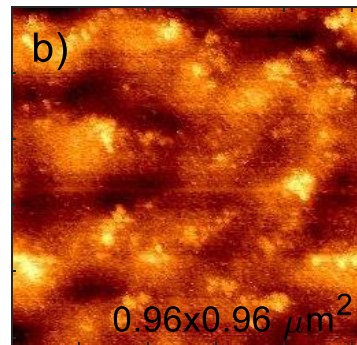
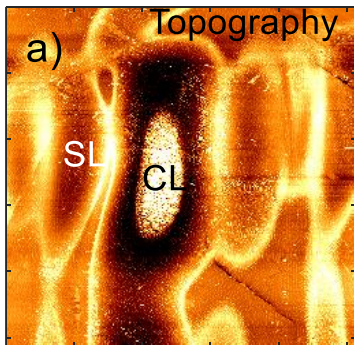
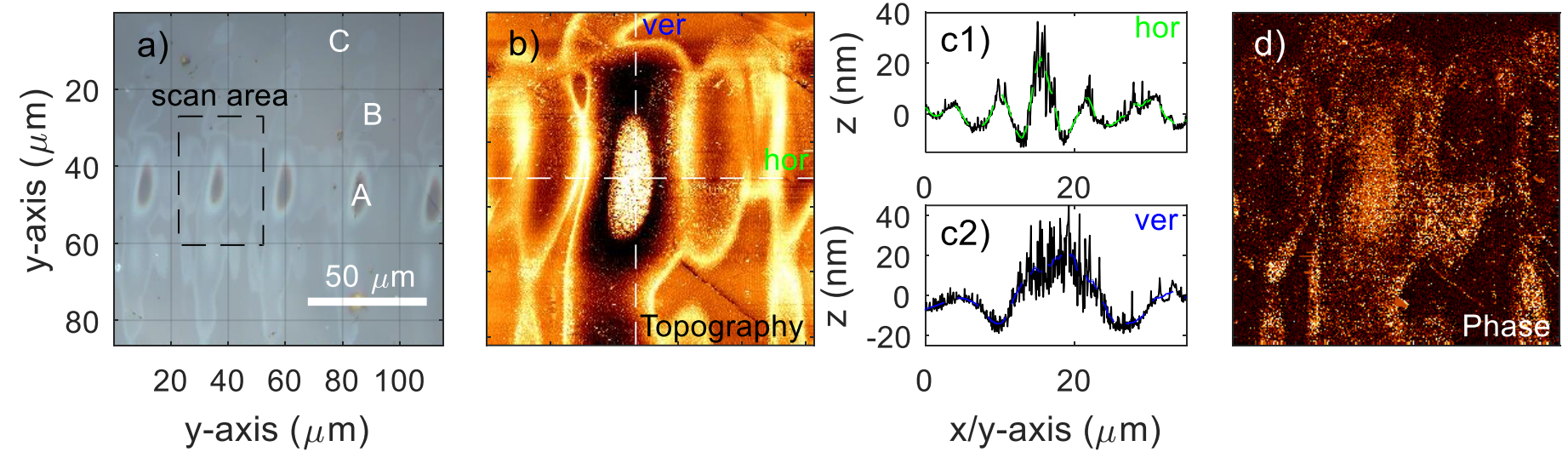


Post-mortem analysis

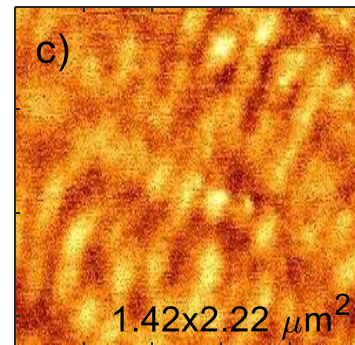


Post-mortem analysis I

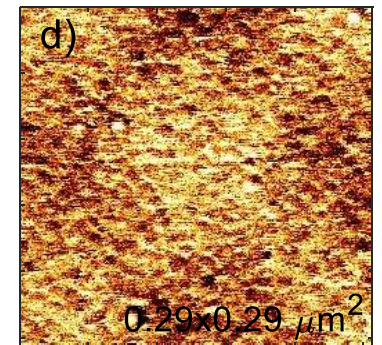
- Scanning probe microscopy (a), non-contact AFM Scan (b), line-outs (c), phase scan (d)



CL



SL



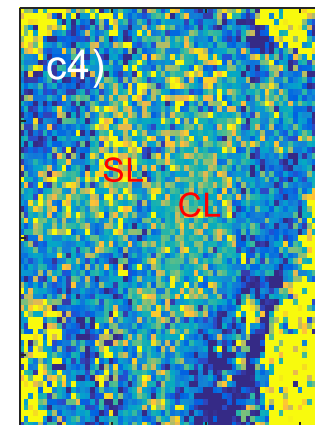
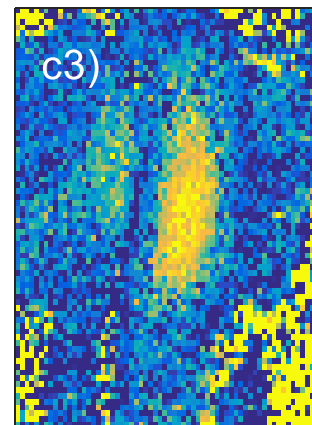
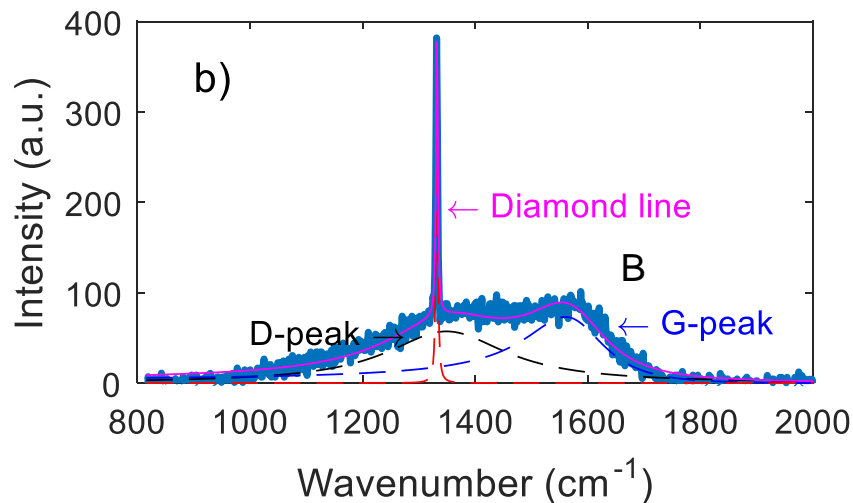
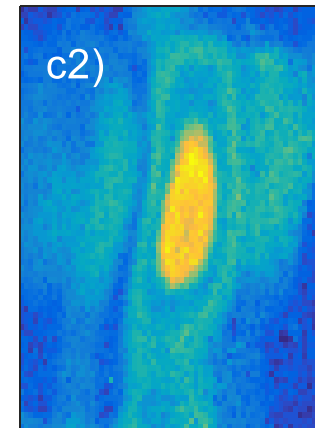
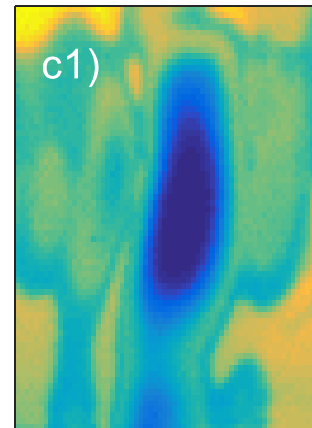
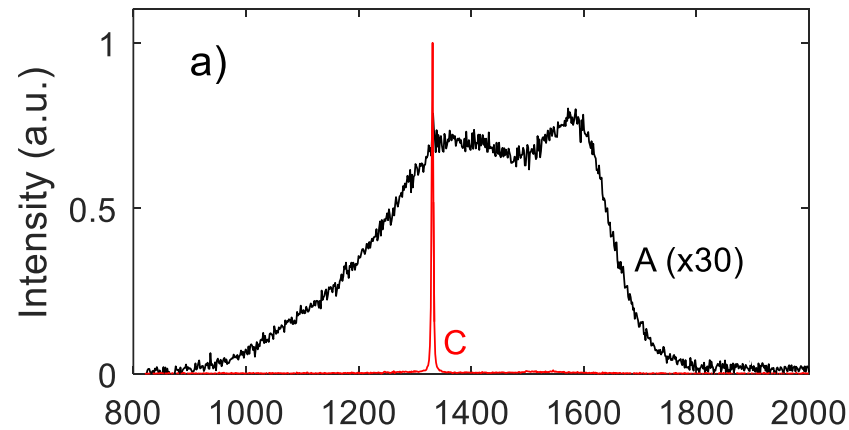
SL

Post-mortem analysis II

> Confocal Raman Micro-Spectroscopy (spot size: $\sim 1 \mu\text{m}$)

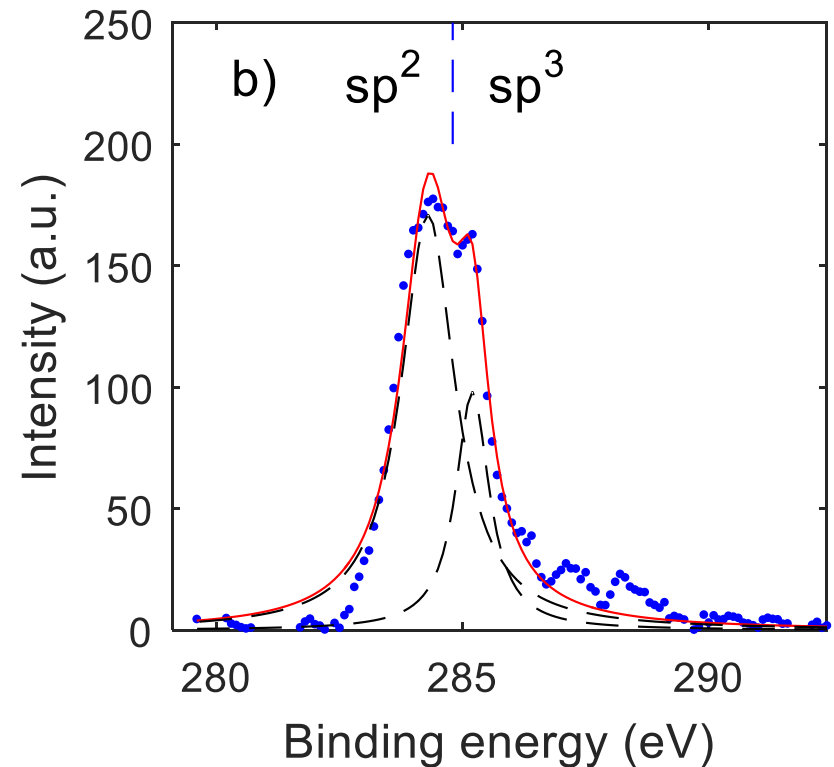
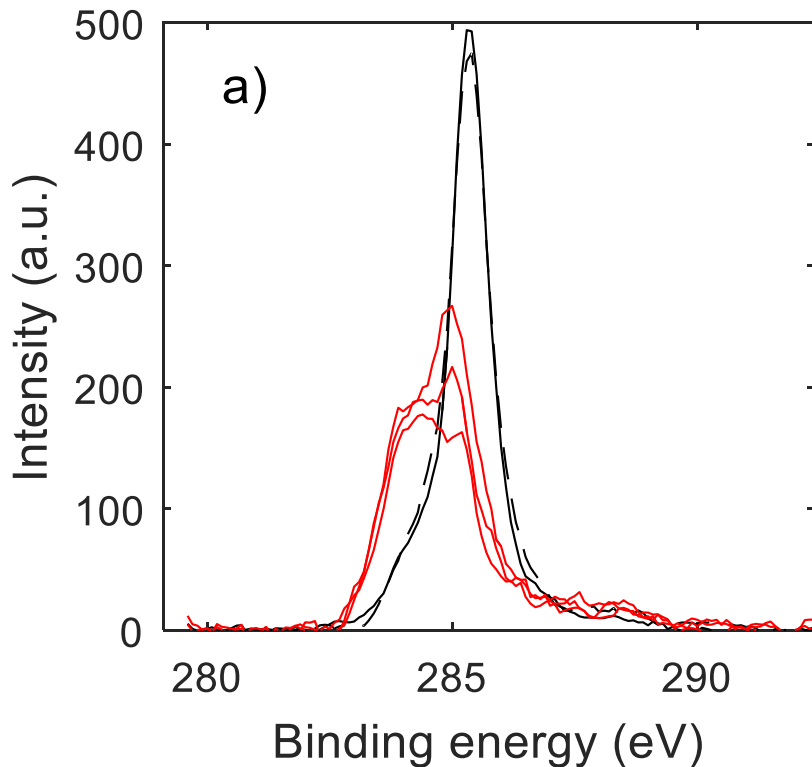
C: diamond line @ 1332 cm^{-1}

A: graphitized layer: D peak ($\sim 1350 \text{ cm}^{-1}$), and G peak ($\sim 1580 \text{ cm}^{-1}$)



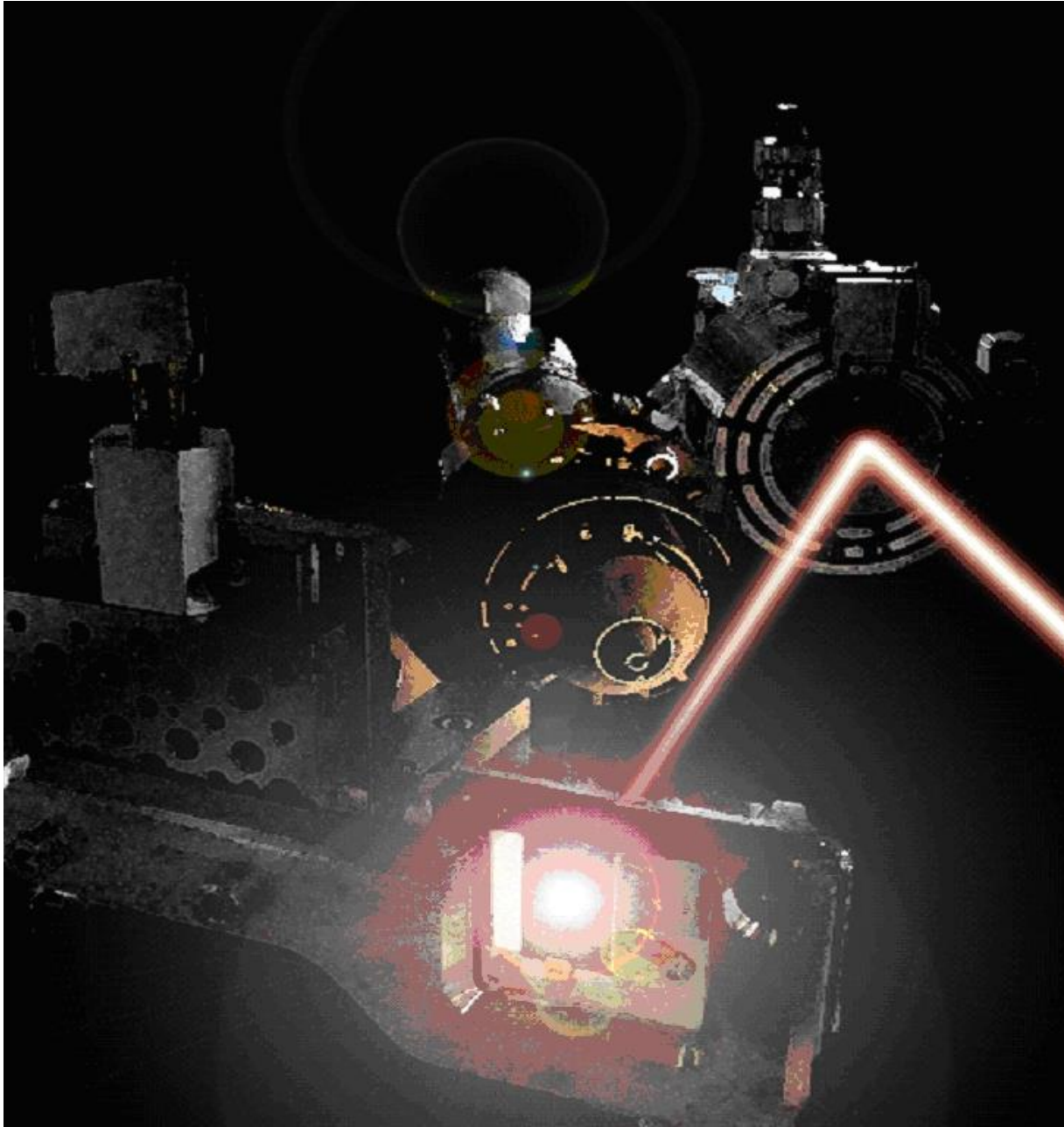
Post-mortem analysis III

- > X-ray Photoelectron Spectroscopy (XPS) with an Al (K- α) source (1486 eV):
spot size: $\sim 9 \mu\text{m}$, measurement depth: 1-5 nm
- > a) black lines: non-irradiated diamond sample, 2 different positions, diamond peak @ 285.5 eV
a) red lines: irradiated sample, 3 different positions of graphitization
- > b) determine sp^2/sp^3 ratio (sp^2 carbon ~ 284 eV and sp^3 carbon ~ 284.9 eV)
-> sp^3 hybrid carbon content in irradiated sample positions varies between 27-42 %



“Transparent” aluminium

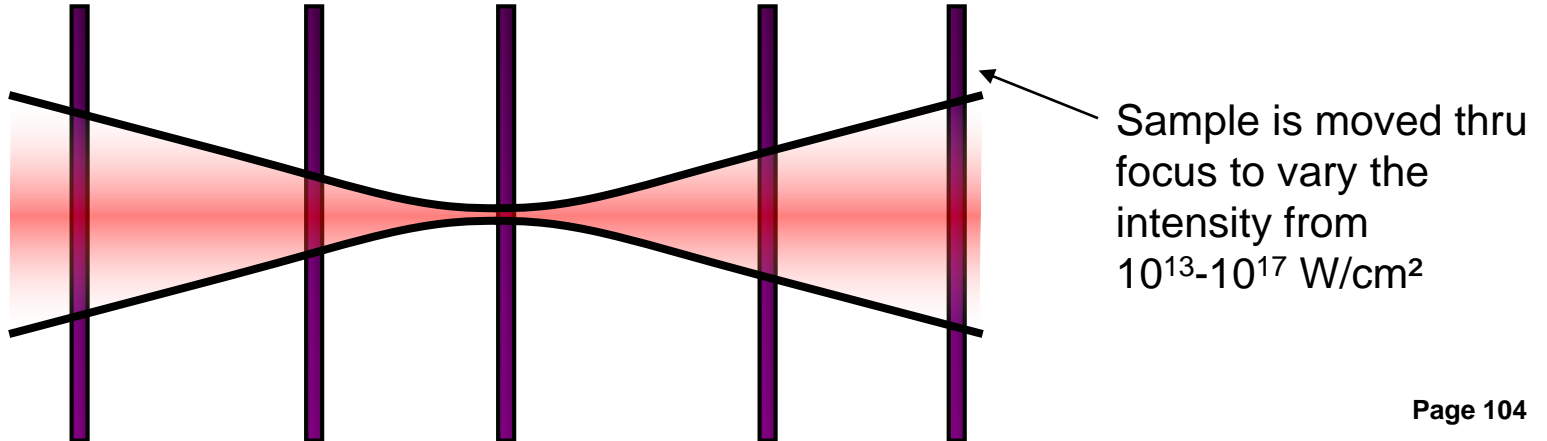
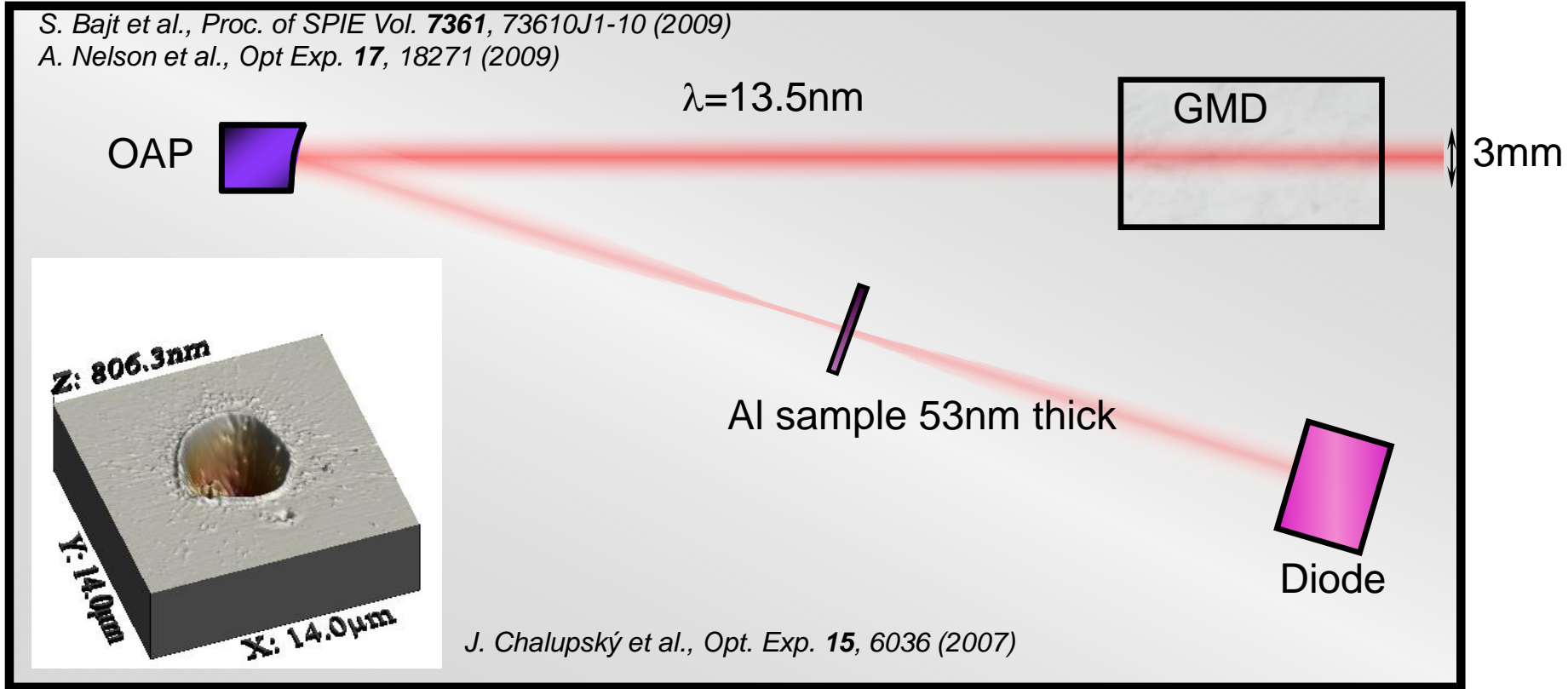
Microfocusing @ FLASH



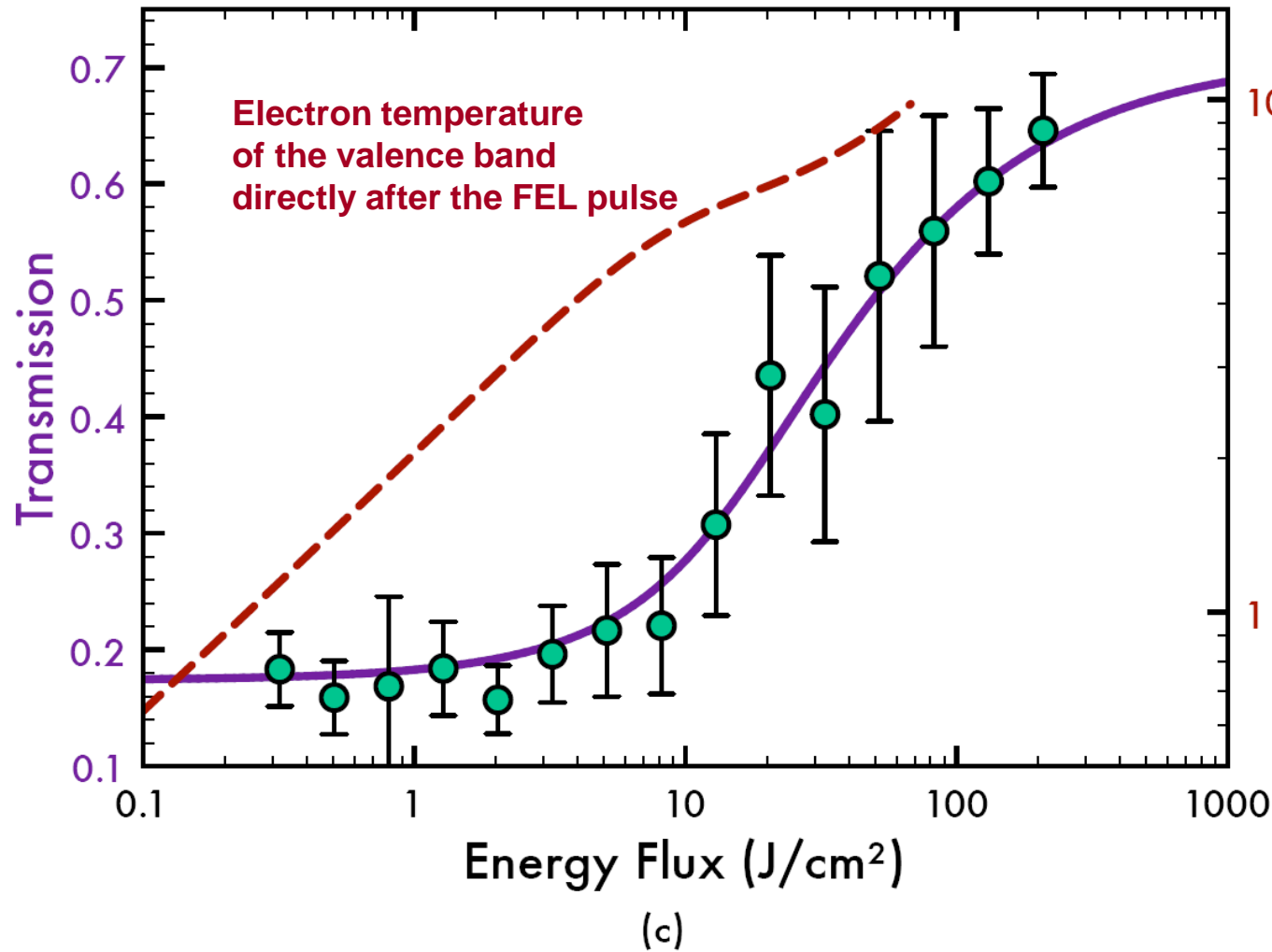
Transparent Aluminium - Experimental Setup

S. Bajt et al., Proc. of SPIE Vol. 7361, 73610J1-10 (2009)

A. Nelson et al., Opt Exp. 17, 18271 (2009)



Transmission of 92 eV photons thru 53 nm Al foil including 10 nm oxide layers

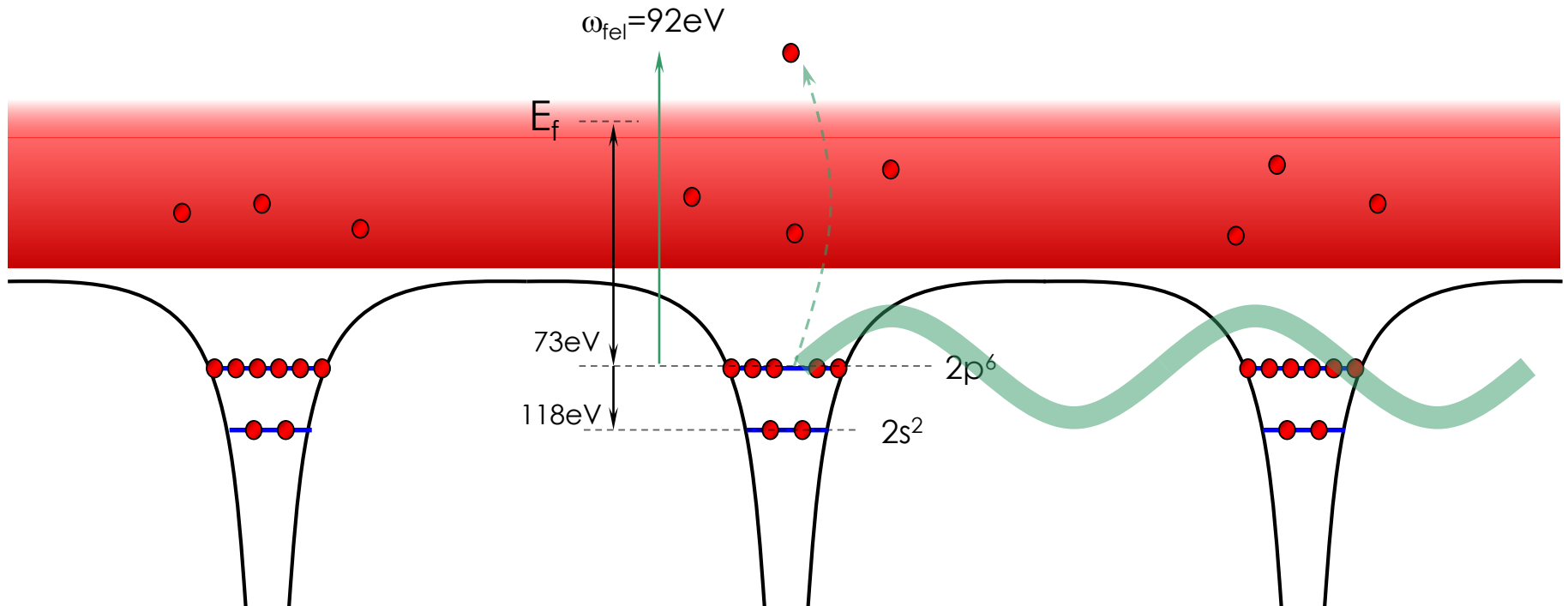


- Photoionization of L-shell electrons
- L-shell core hole state
- L-shell shift
- Quenching of bound-free absorption

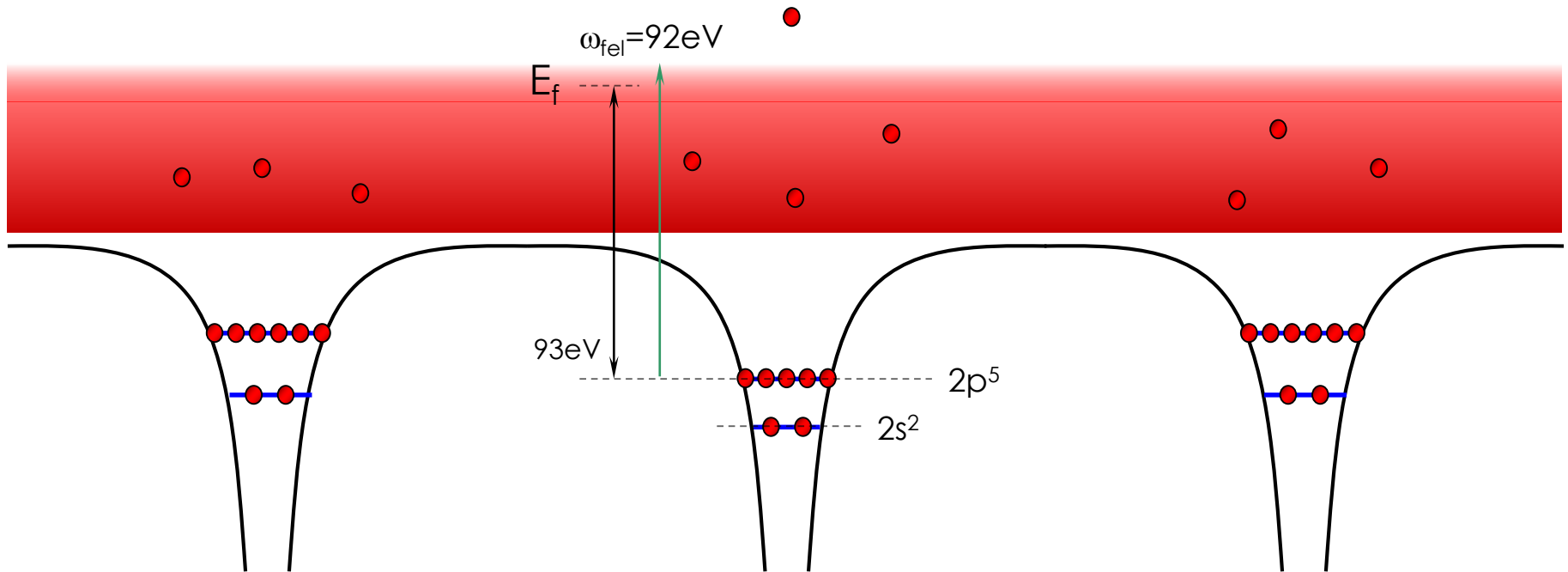
- Recombination time ~40 fs (Auger recombination)
- FEL pulse duration ~30 fs

B. Nagler et al., Nature Physics 5, 693 (2009)

Al after a single photoionization



Al L-edge shift after photoionization

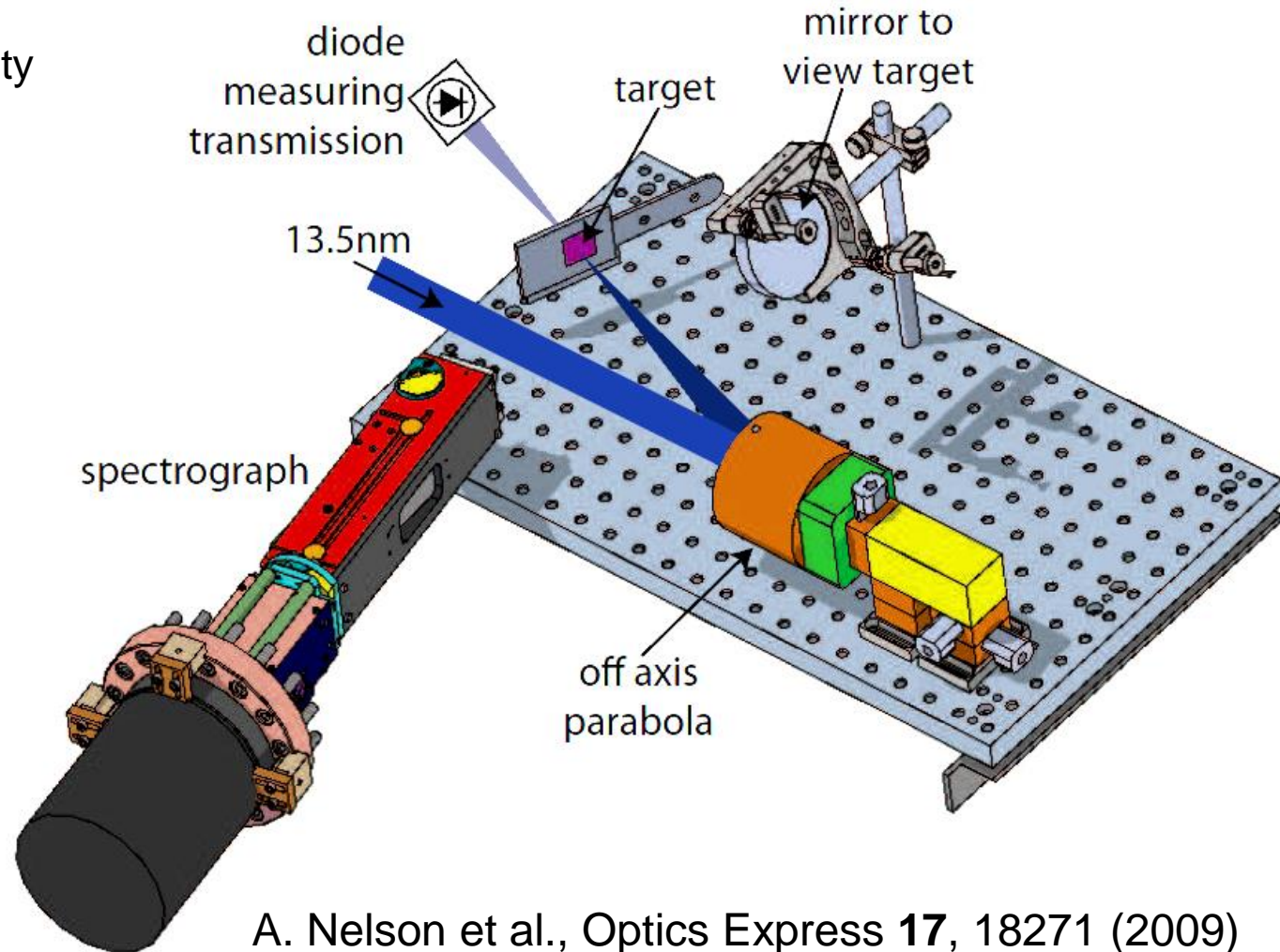
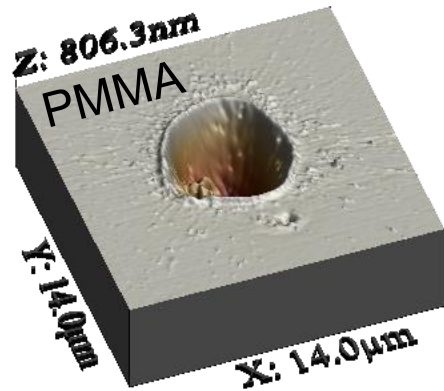


- Removal of 1st 2p electron causes shift of $n = 2$ shell due to loss of the outer screening.
- Further photons cannot ionize the L-shell
- E_f will increase as there are now four electrons in the conduction band ($\sim 2.4\text{ eV}$)

Microfocusing FLASH

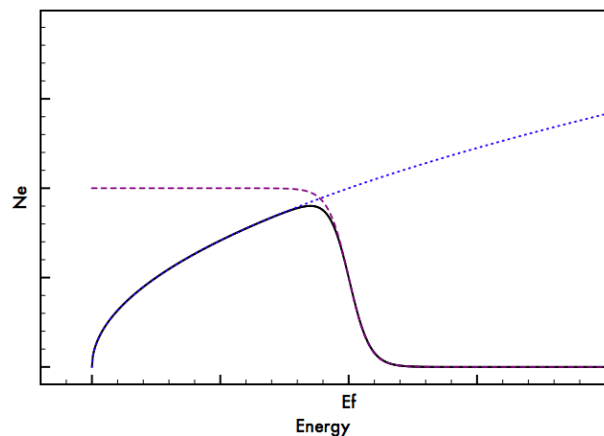
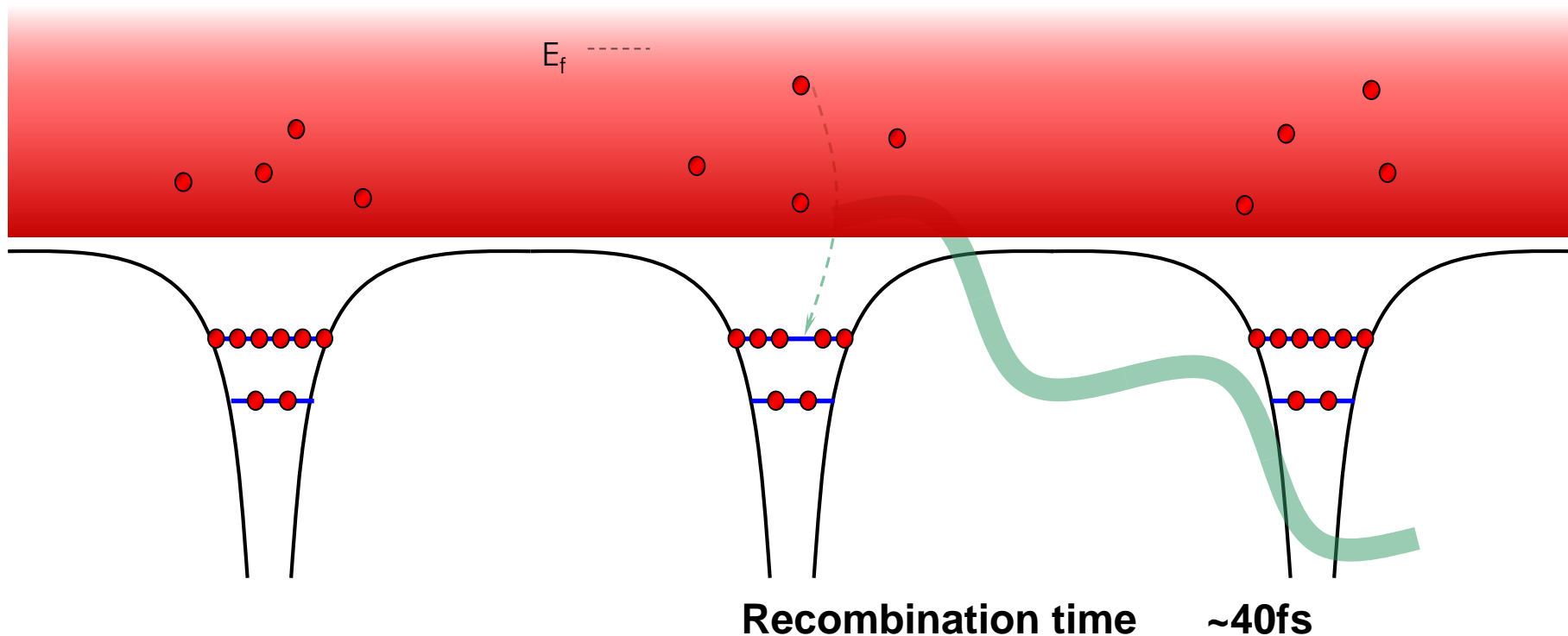
→ High Energy Density

- 10^{17} W/cm²
- Focus ~ 0.7 μ m



A. Nelson et al., Optics Express **17**, 18271 (2009)

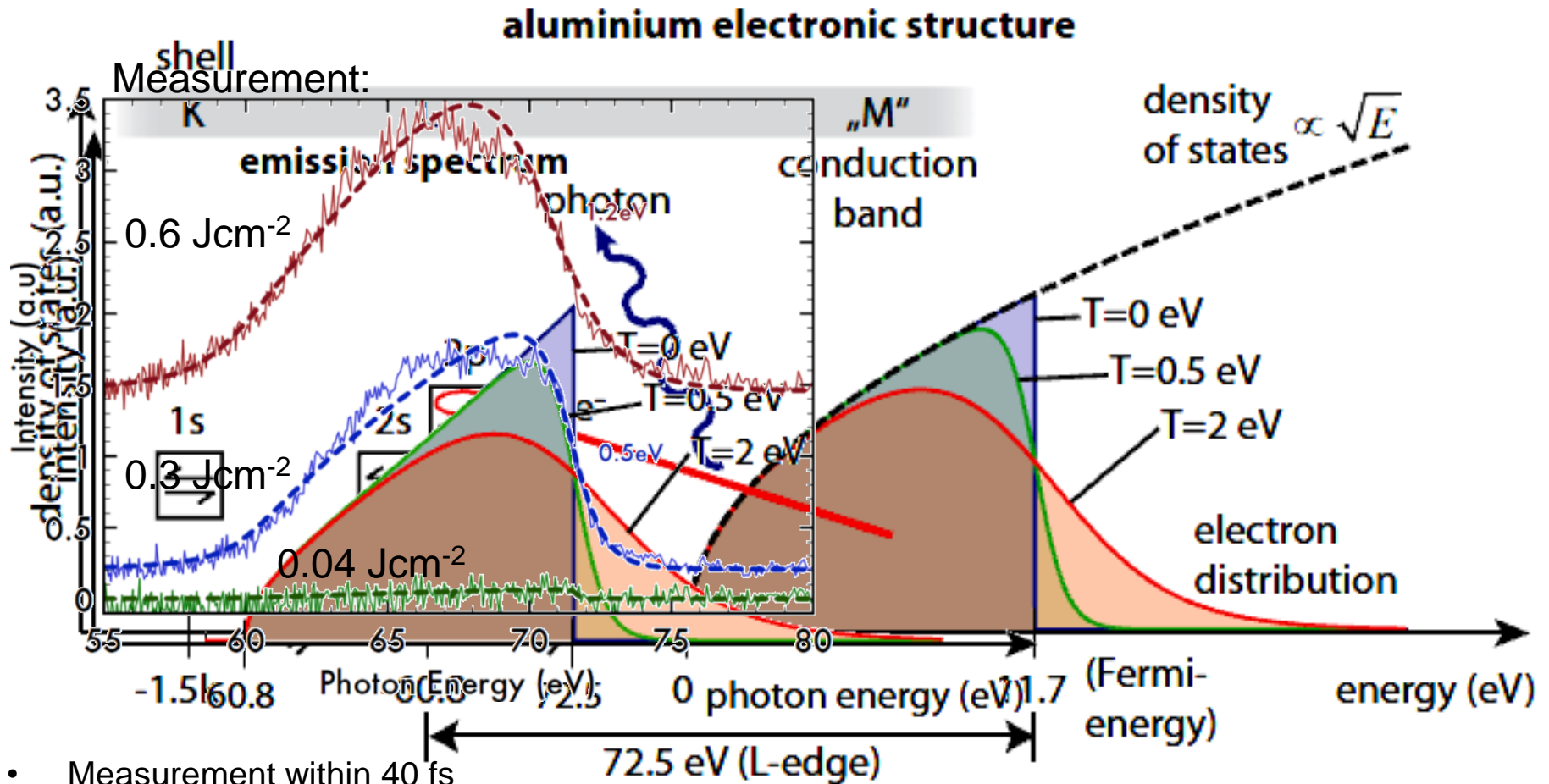
Recombination Time



**Should see fluorescence before
lattice moves.**

Fluorescence proportional to $\omega^3 g(E)f(E, T)$
(better model under development)

Fluorescence Maps Valence Band



- Measurement within 40 fs
- Time limited by Auger decay

S. Vinko et al., PRL 104, 225001 (2010)

Thank you for your attention

