FLASH strategy and FLASH 2020+ project

DESY WA

Markus Gühr for the whole FLASH team

HELMHOLTZ
RESEARCH FOR GRAND CHALLENGES

FLASH provides unique opportunities for science

Users at FLASH study electronic processes on femtosecond timescales in many different system

SASE source

5000 bunches/second, up to 1mJ per bunch

1-400 eV photon energy in fundamental

Up to 730 eV in third harmonic

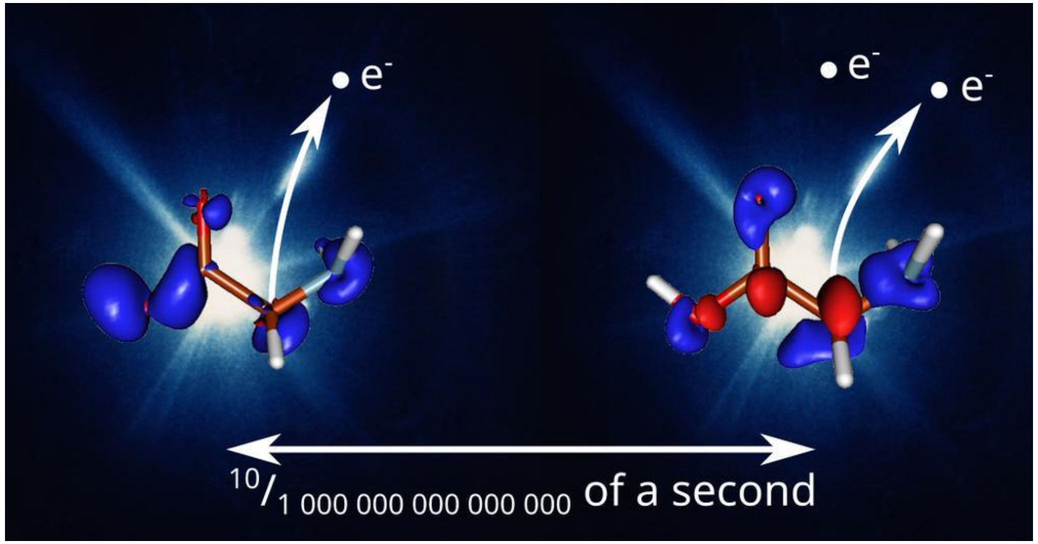
Optical lasers

X-ray split and delay

X-ray diagnosics

FLASH Science

Electronic dynamics in glycine

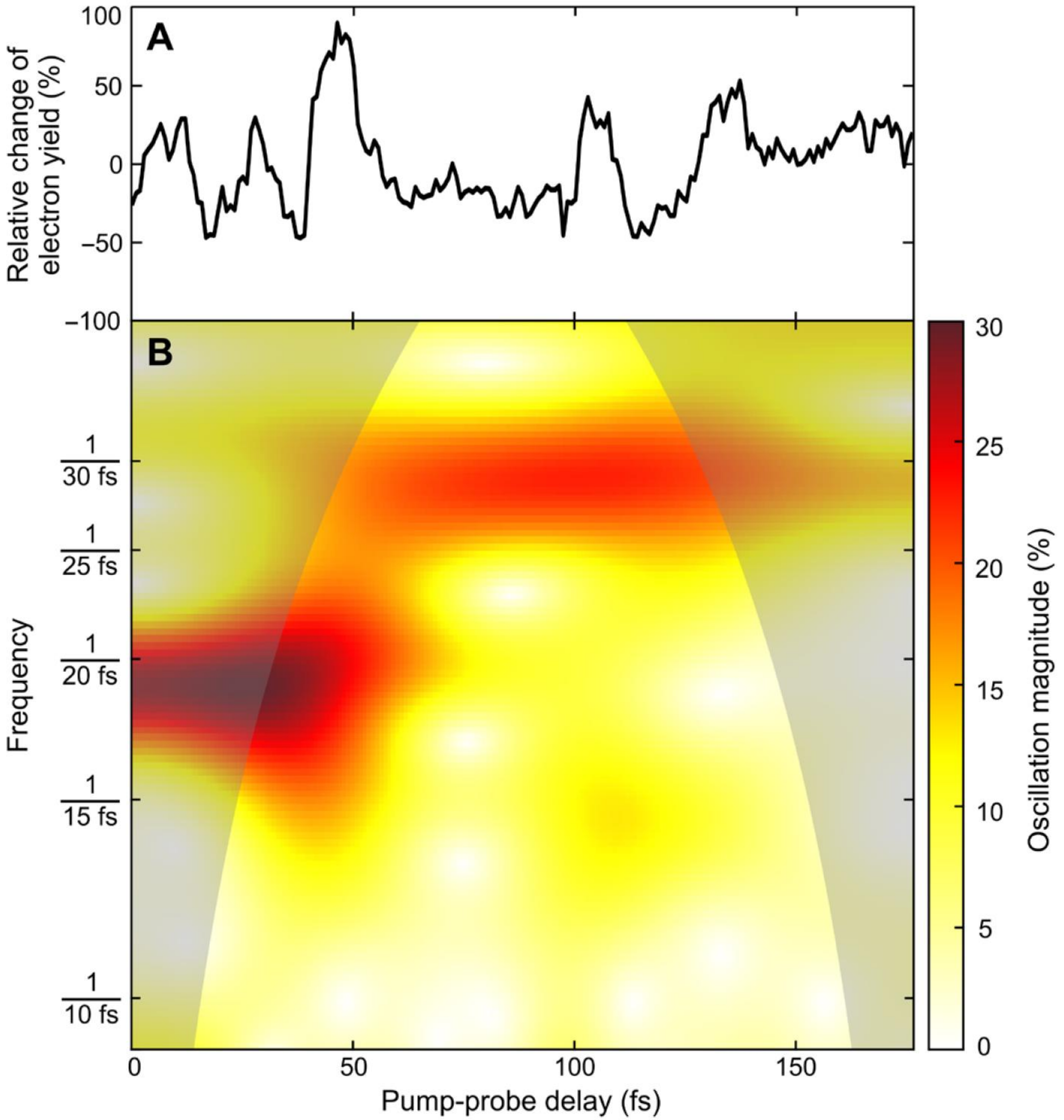


D. Schwickert, …, T. Laarmann, Science Advances, 2022

X-ray pump probe study on an aminoacid

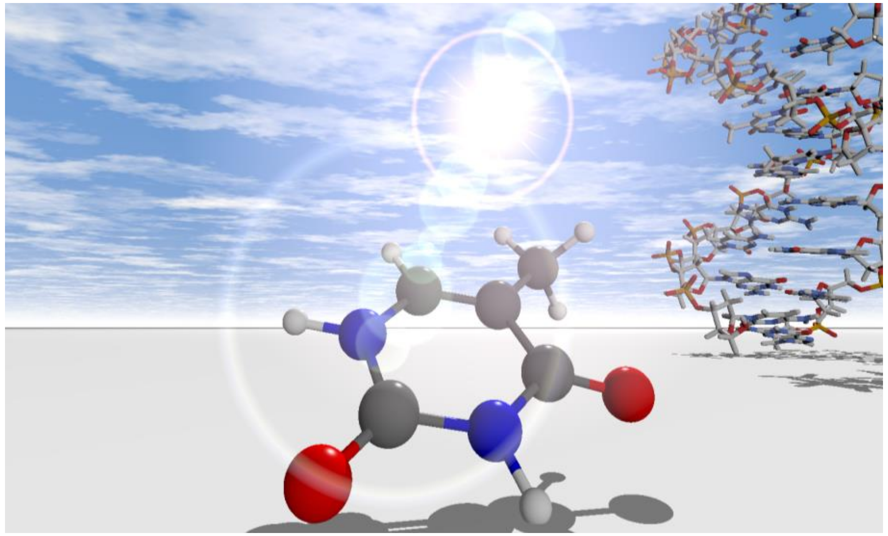
Excitation pulse leaves molecule in an electronic superposition

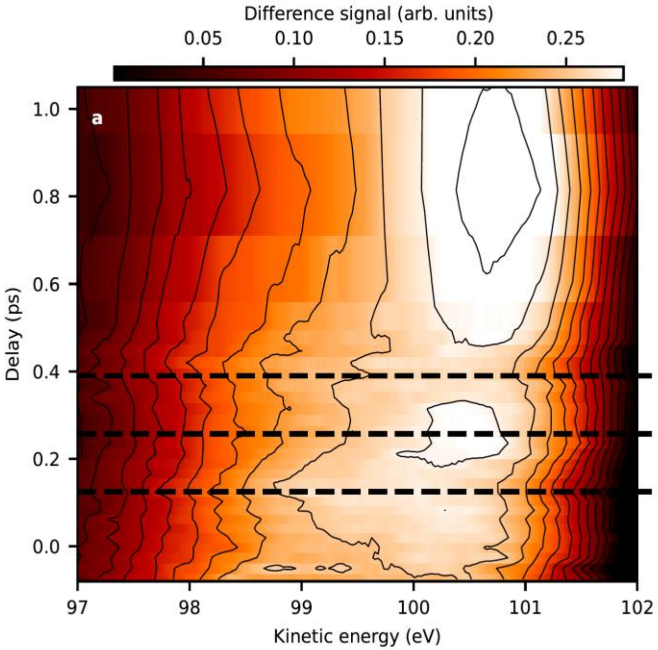
Local probing at C K-edge shows the charge migration and its coupling to nuclear vibrations

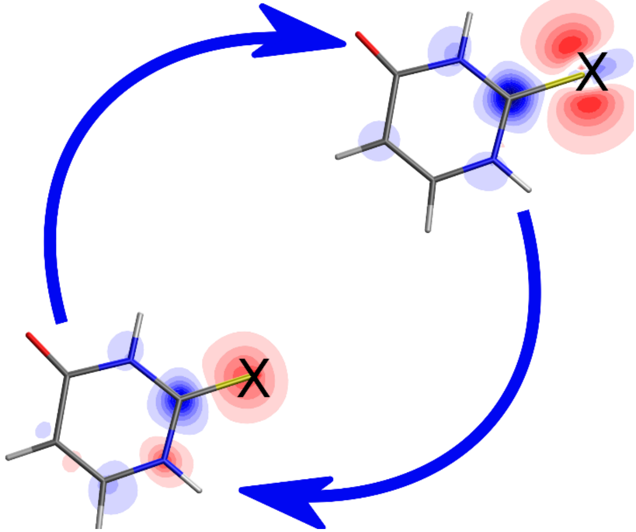


FLASH Science

Molecular photoenergy conversion – the electronic molecular movie







Mayer, Lever, Picconi, …Gühr, Nat. Comm. **13** 198, (2022)

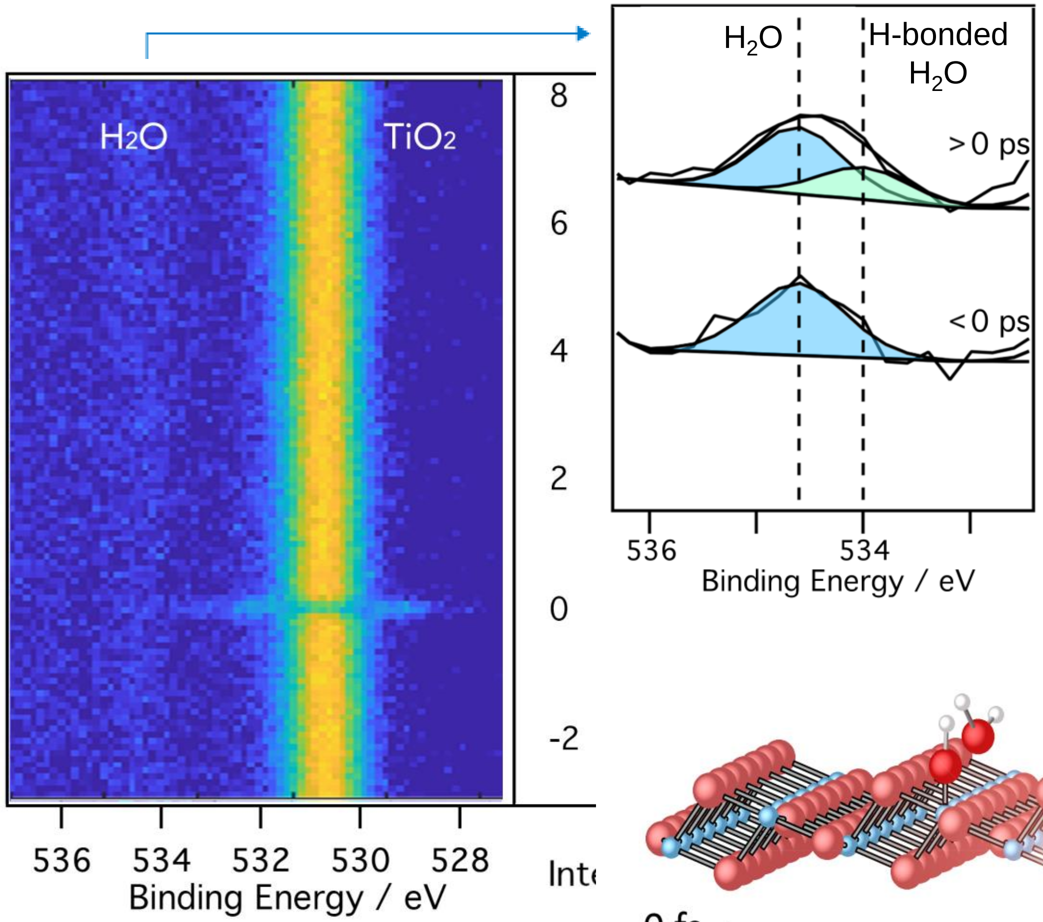
- UV absorption in nucleobases – conversion to heat
- UV absorption in *thio*nucleobases - conversion to destructive triplet states
- FLASH S 2p XPS study shows a *coherent* path towards triplet state

- Electronic movies from XPS serve to understand the molecular photoelectron conversion
- Goal: steering photochemistry by molecular design

FLASH science

Ultrafast Photoinduced Dynamics at interface of water and TiO2

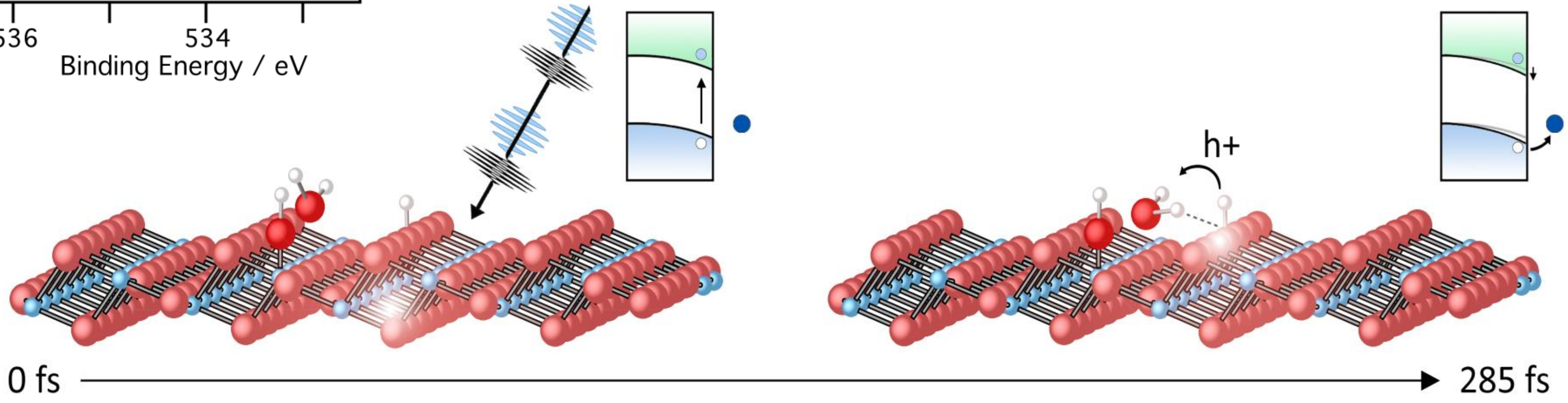
A combined femtosecond time-resolved optical pump--soft X-ray probe photoemission and Ehrenfest molecular dynamics study



Optical pulse generates electron hole pairs in water covered TiO_2.

Subsequent charge carrier dynamics were probed by monitoring changes in the O 1s core level

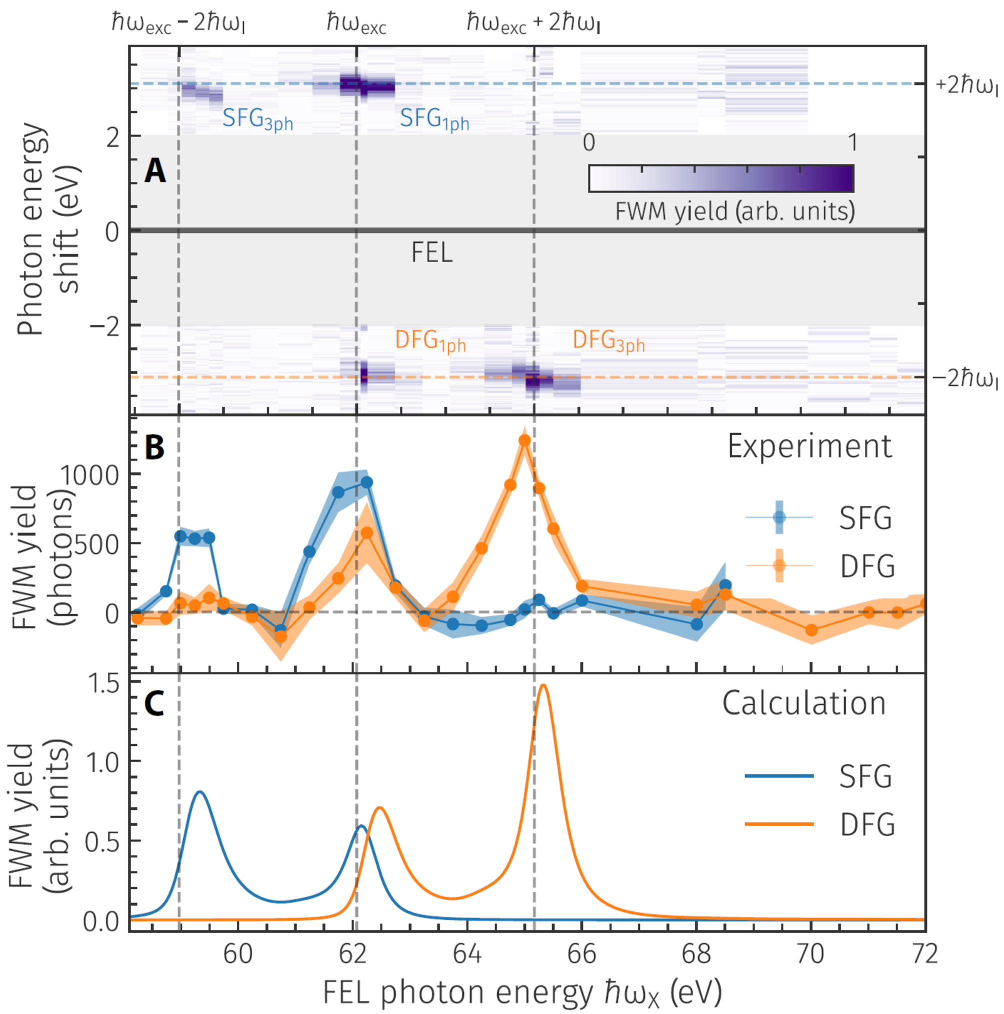
Peak shifts are due to ultrafast interfacial hole transfer within 285 fs, facilitated by new hydrogen bond formation between water and the surface

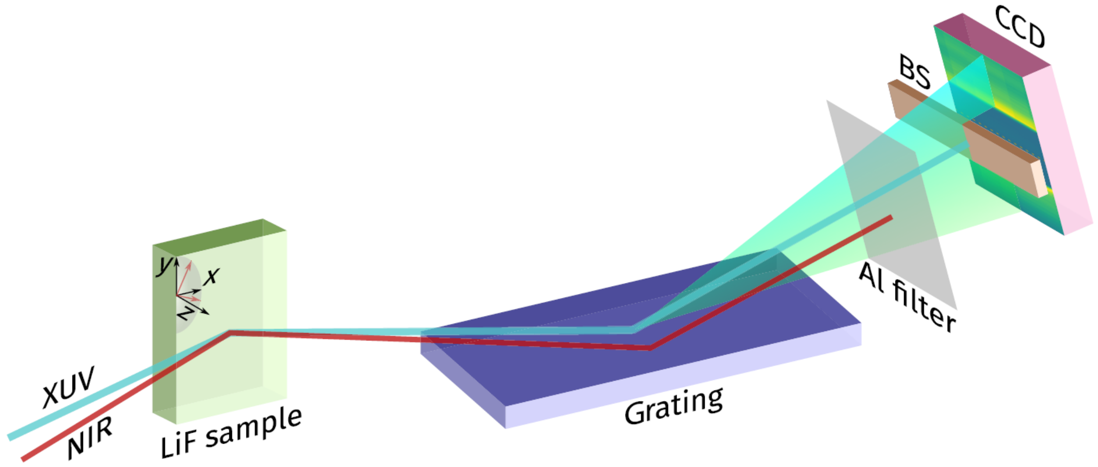


M. Wagstaffe, …, H. Noei , Physical Review Letters, 2023

FLASH science

Four wave mixing in LiF





Four wave mixing in LiF using XUV and NIR pulses

Third order susceptibility is high for colocalization of initial and final states

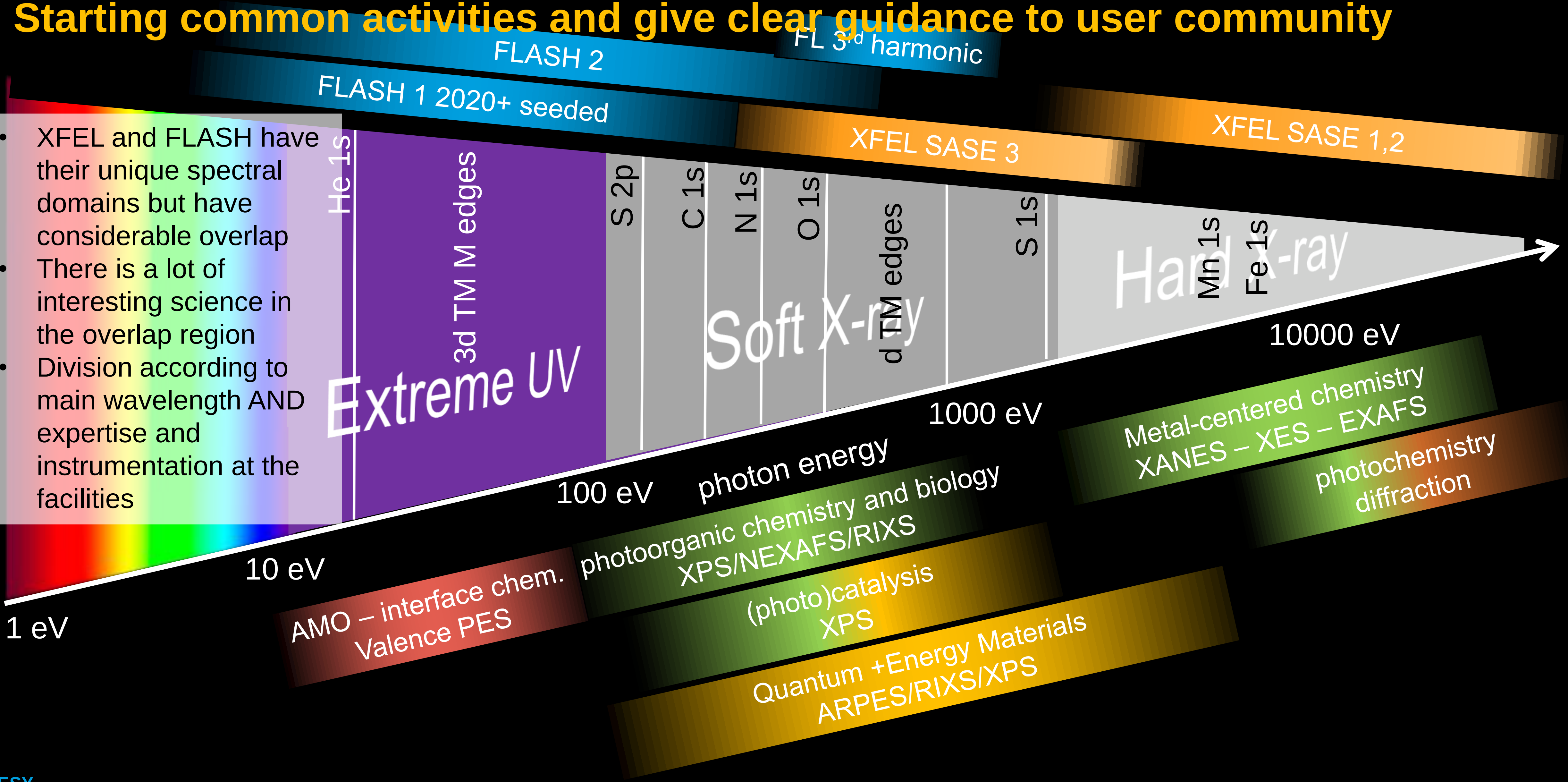
Access to optically dark states at the targetted site

H. Rottke, R. Engel. D.Schick …,M. Beye, S. Eisebitt, Science Advances, 2022

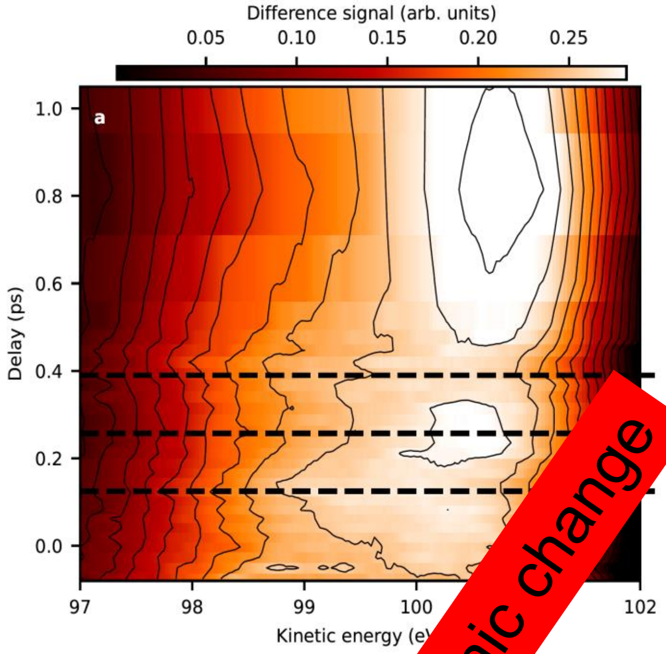
FLASH – XFEL synergies

Starting common activities and give clear guidance to user community

- XFEL and FLASH have their unique spectral domains but have considerable overlap
- There is a lot of interesting science in the overlap region
- Division according to main wavelength AND expertise and instrumentation at the facilities

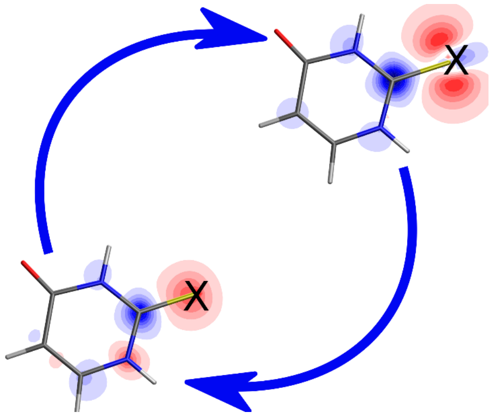


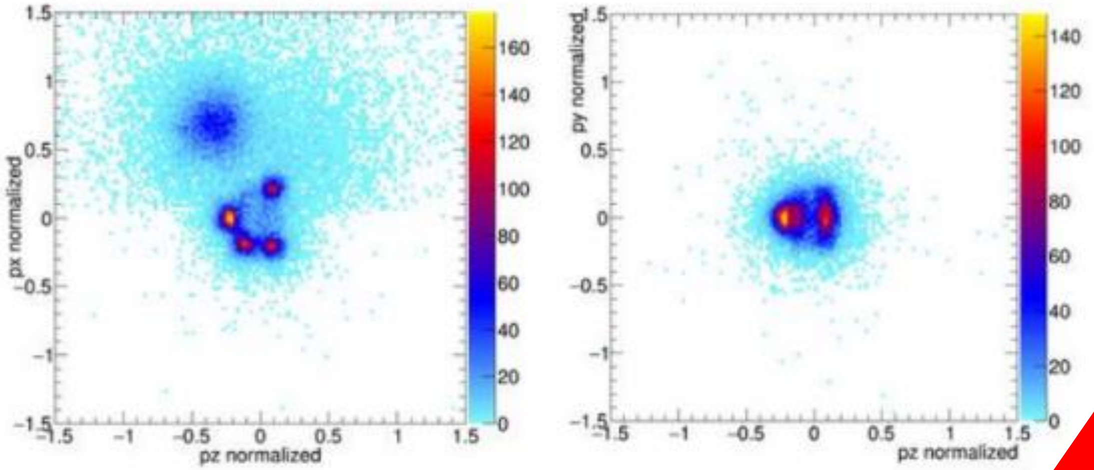
FLASH – XFEL synergies

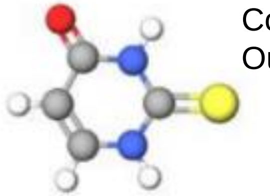


Mayer...Gühr, Nat. C... 13 198, (2022)

Electronic change

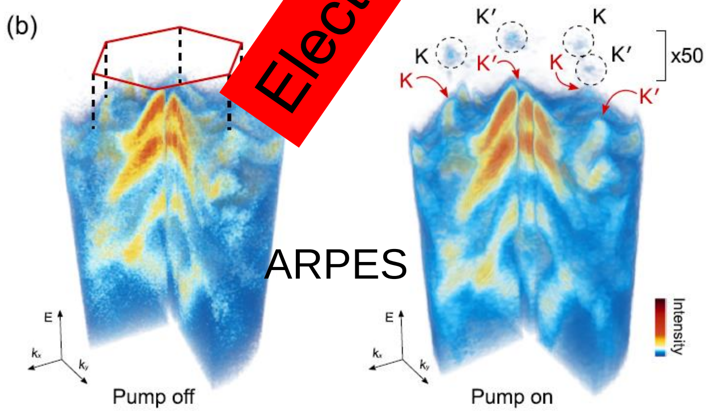






Courtesy: Till Jahnke
Our Beamtime Nov 15-20th, SQS

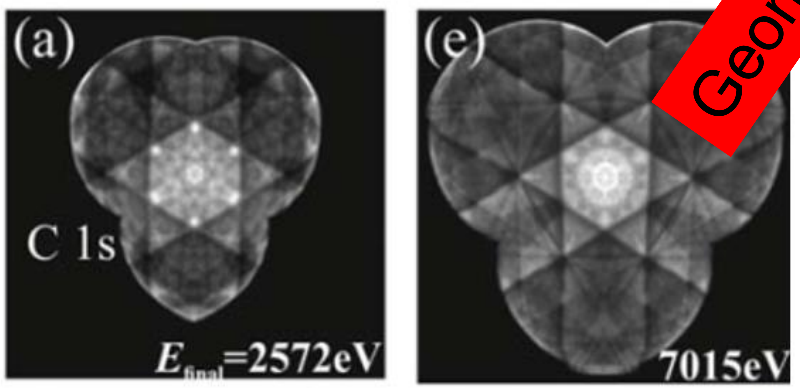
Geometry change



Kutnyakhov et al. Rev. Sci. Instr. 91, 013109 (2020)

- Small molecule chemistry
- FLASH: electronic dynamics via XPS or XAS
- XFEL: Coulomb Explosion imaging of the geometry

- Material dynamics with k-Microscope
- FLASH: ARPES and XPS
- XFEL: Photoelectron diffraction



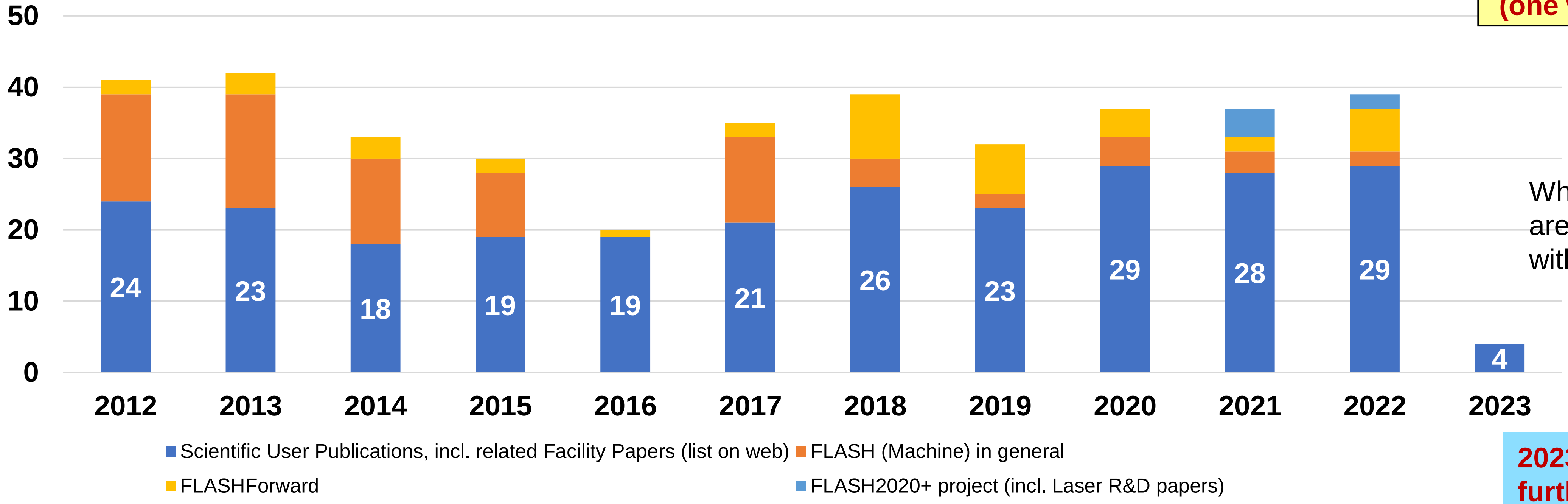
Fedchenko et al. New. J. Phys. 21, 113031 (2019)

Photon-Science at FLASH

Publications

FLASH results made it on two cover pages in 2022 (one with inhouse research)!

FLASH User Publications + peer-reviewed machine and laser publications

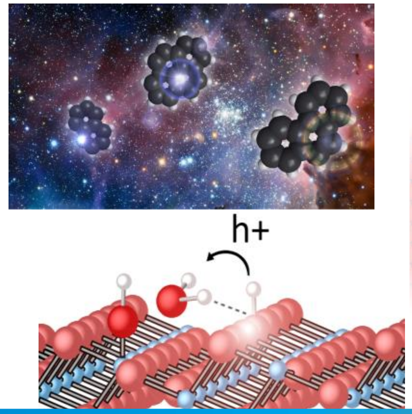


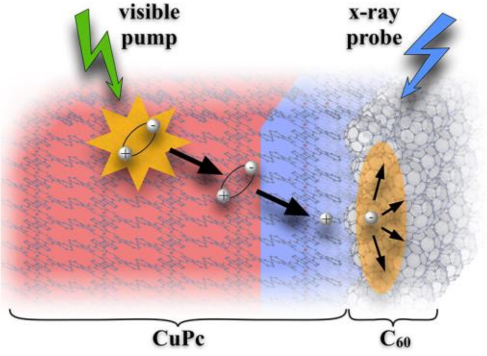
White numbers for blue bars are the usual numbers without the machine papers

2023:
further 7 papers submitted (partially in 2022)

Courtesy: Rolf Treusch

The evolution of FEL science





Stable source
Sensitive methods
Smaller datasets

Early 2000s

Pioneer experiments
Strong x-ray matter interaction

X-ray only experiments

Experiments highly adapted to the source

Often done by people at the facilities

Now

X-ray expert experiments
Linear x-ray interaction for probing out of exuilibrium states

Laser pump – FEL probe

Experiments on model systems

Expert community whith strong bounds to the facilities

The next decade

Topical expert experiments
Linear x-ray interaction for probing out of equilibrium states

Laser pump – FEL probe

Experiments on relevant samples

Goal: we want to reach a wider community that is not involved at the facilities yet and has a low entry barrier to performing the experiment and analyzing the data

Workshop on Future of science at FLASH

September 25-27, 2017 @ DESY + further topical workshops in the years before

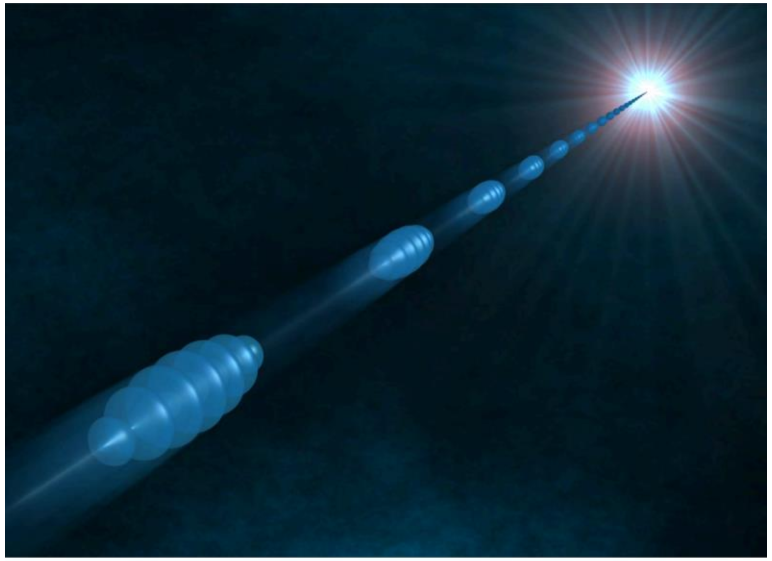
- ~120 participants
- 15 contributed talks
- 6 facility talks
- Poster session
- Breakout sessions on **AMO, chemistry and life sciences and condensed matter science**

Keynote speakers:

• Catalysis	Anders Nilsson
• Nonlinear science	Claudio Masciovecchio
• Functional materials	Hermann Dürr
• Imaging	Daniela Rupp
• Astrochemistry	Melanie Schnell
• Magnetism	Jan Lüning
• AMO	Markus Gühr

Coutesy: Rolf Treusch

FLASH2020+ a unique machine in the FEL landscape



FLASH1	FLASH2
Seeded	SASE
Full polarisation control	
Quick and easy tunability	✓
Small bandwidth	Extended wavelength range
Full coherence	(Sub-) femtosecond pulses
Flexible laser based pump confgurations	
Advanced two-color FEL schemes	

FLASH 2020+

Making FLASH brighter, faster and more flexible
Conceptual Design Report

Deutsches Elektronen-Synchrotron DESY
A Research Centre of the Helmholtz Association

CDR in 2020
Based on science workshop
Sept. 2017

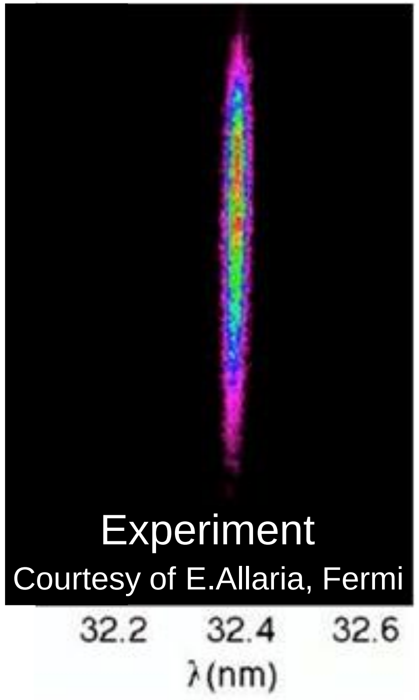
Coutesy: Rolf Treusch

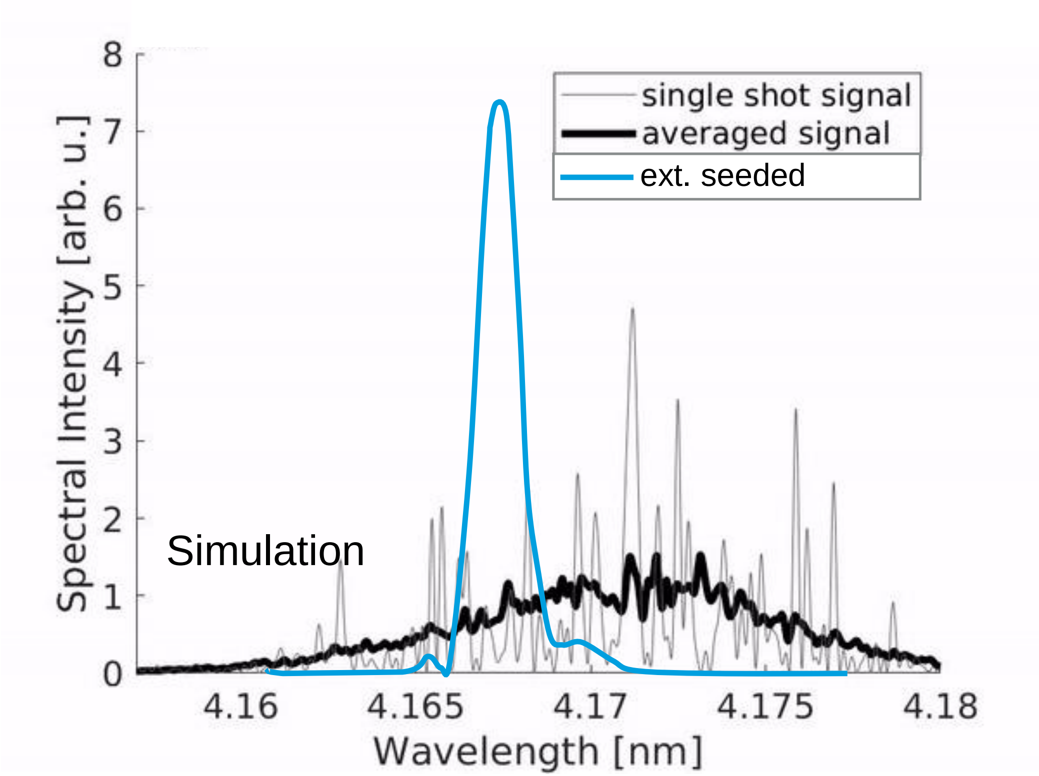
	FLASH1 (Seeded)	FLASH1 (SASE)	FLASH2	
Wavelength range	4 – 60	4 – 60	1.3 – > 60*	nm
Pulse energy	<100	<1000	<1000	µJ
Pulse duration (FWHM)	30	5 - 200	0.1 – 200	fs
Spectral width	Fourier limited	0.5 - 2	0.5-2	%
Pulses per second**	10 – 5000	10 – 5000	10 – 5000	

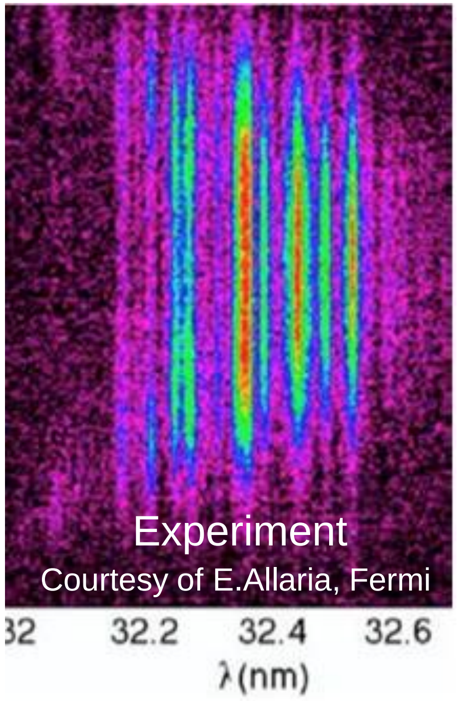
*including third harmonic **Shared between FLASH1 and FLASH2

External seeding

Narrower Bandwidth, Stability and Coherence at unique repetition rate







Seeded **FLASH1**

- Narrow bandwidth
- Stability
- Longitudinal coherence

Facility will provide both simultaneaously

Courtesy: Lucas Schaper

SASE **FLASH2**

- Low complexity
- Pulse energy
- Shortest pulses
- Repetition rate

FLASH2020+ upgrade

FLASH: towards a high repetition rate seeded soft X-ray FEL

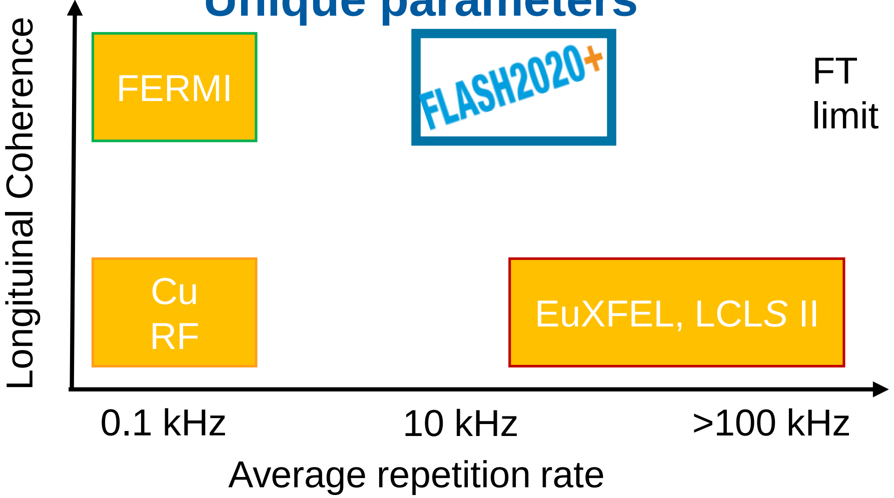
2021 — 2022 — 2023 — 2024 — 2025

- **2022:** Energy upgrade nitrogen K-edge
- **2023:** Variable polarization at third harmonic
- **2025:** Externally seeded operation up to the carbon K-edge

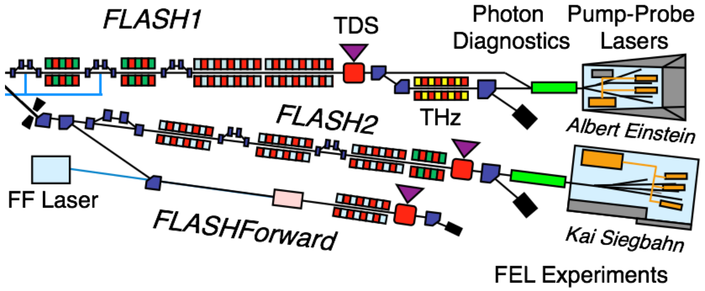
Temporal and spectral stability

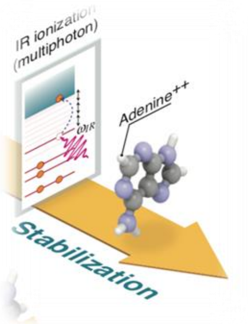
Credit: Forschungsverbund Berlin e.V. (FVB)

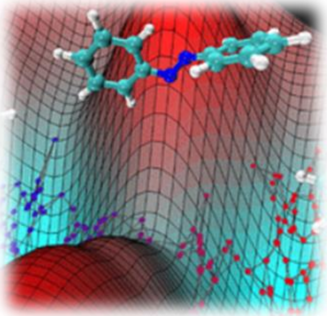
Unique parameters

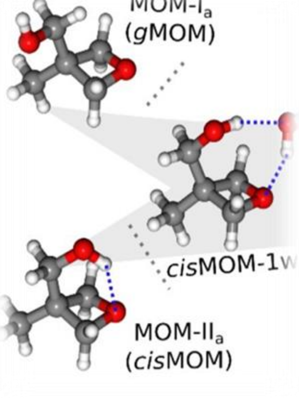


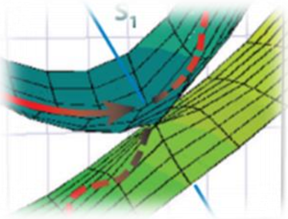
Parallel operation - more beamtime

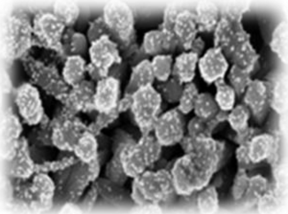








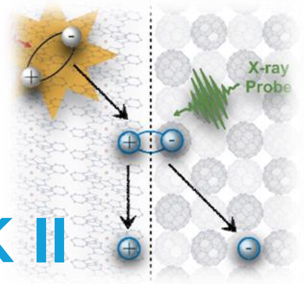


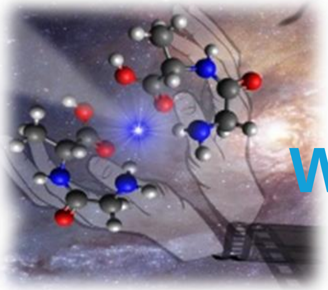


Two important points:

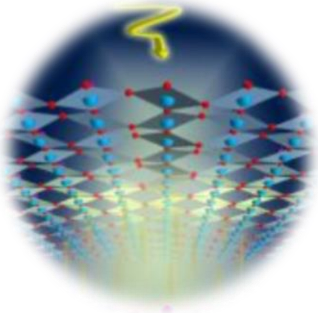
We needed to formulate science challenges for 2020+

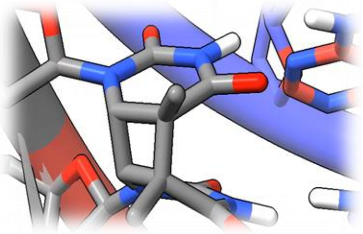
We need to strengthen the connection between LK I and LK II

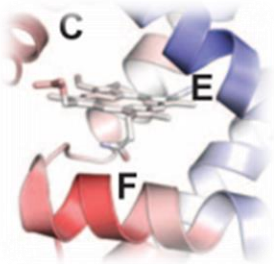


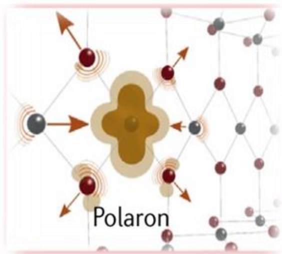


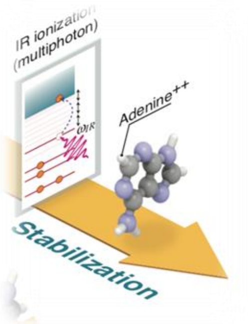
Result: 12 Science Challenges

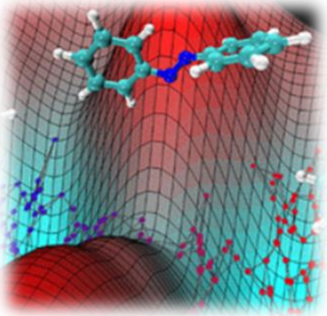


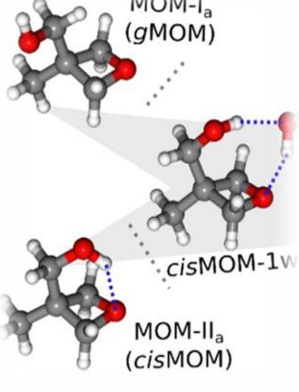


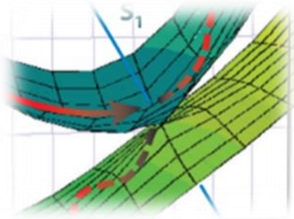


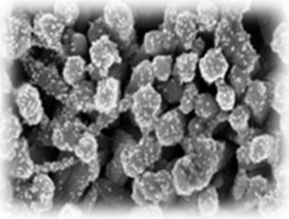




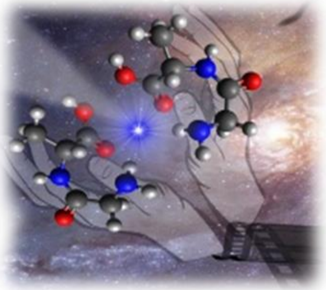


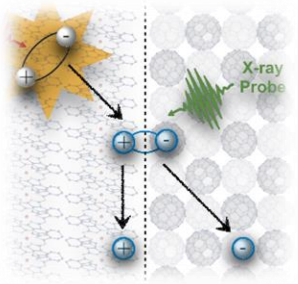




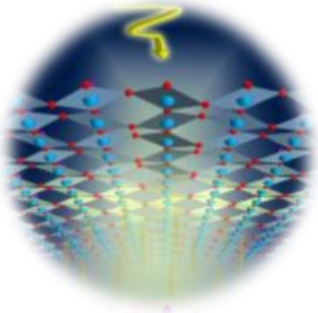


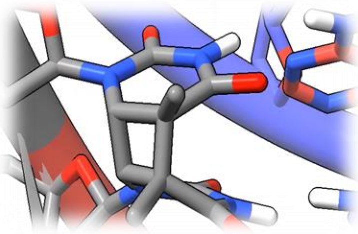
12 Challenges
uniquely benefit from FLASH2020+

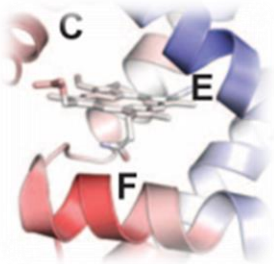


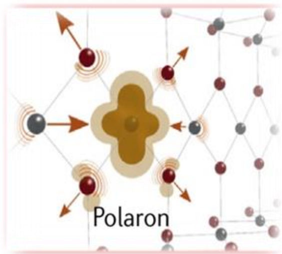


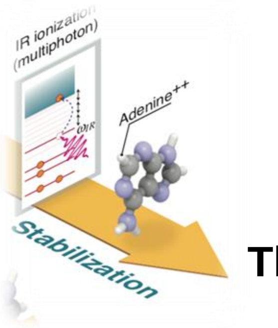
Sadia Bari, Martin Beye, Francesca Calegari, Henry Chapman, Benjamin Erk, Ulrike Frühling, Tais Gorkhover, Markus Gühr, Niels Huse, Markus Ilchen, Jochen Küpper, Tim Laarmann, Heshmat Noei, Elke Plönjes-Palm, Nina Rohringer, Kai Rossnagel, Lucas Schaper, Melanie Schnell, Siegfried Schreiber, Lucas Schwob, Holger Sondermann, Andreas Stierle, Simone Techert, Kai Tiedtke, Andrea Trabattoni, Rolf Treusch, Sebastian Trippel, Charlotte Utrecht

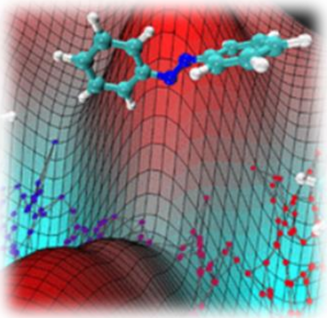


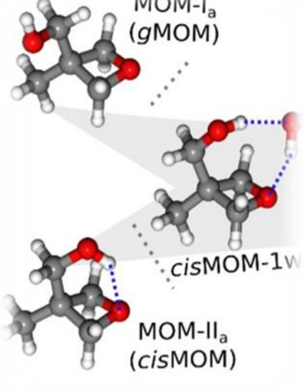


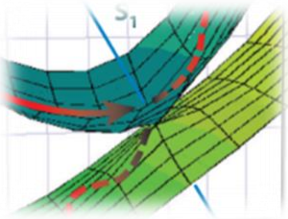


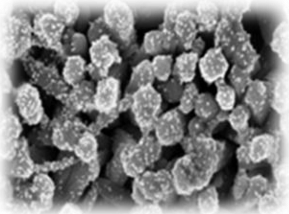












The role of electron correlation and coherences in charge-directed reactivity

Molecular ground state reactions

Steering molecular dynamics by controlled solvation

Understanding molecular photoenergy conversion

Plasmonic noble metal nanoparticles in photo-catalysis

Optimize efficiency of solar cells

Resolving polaron dynamics

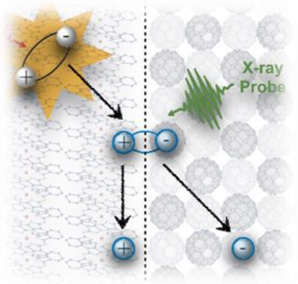
Control and stabilization of functional phase transitions

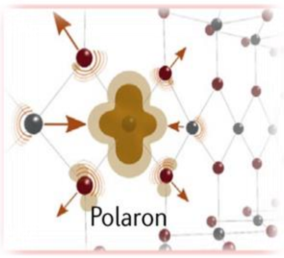
Understanding biomolecular functionality through its dynamics

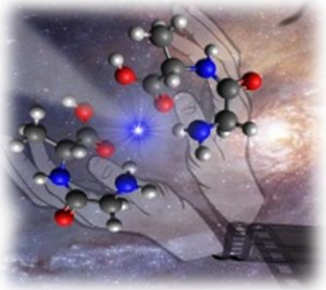
Understanding life’s photoprotection mechanisms

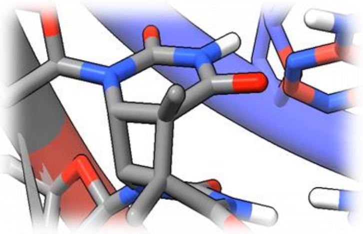
Emergence of molecular complexity in space

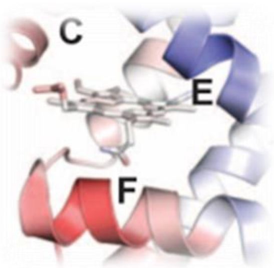
Nature’s molecular asymmetry in highly transient matter and processes

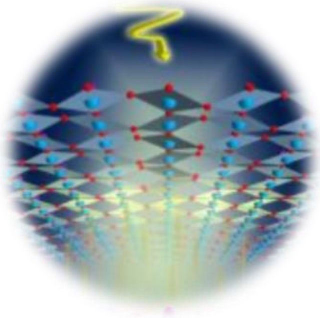










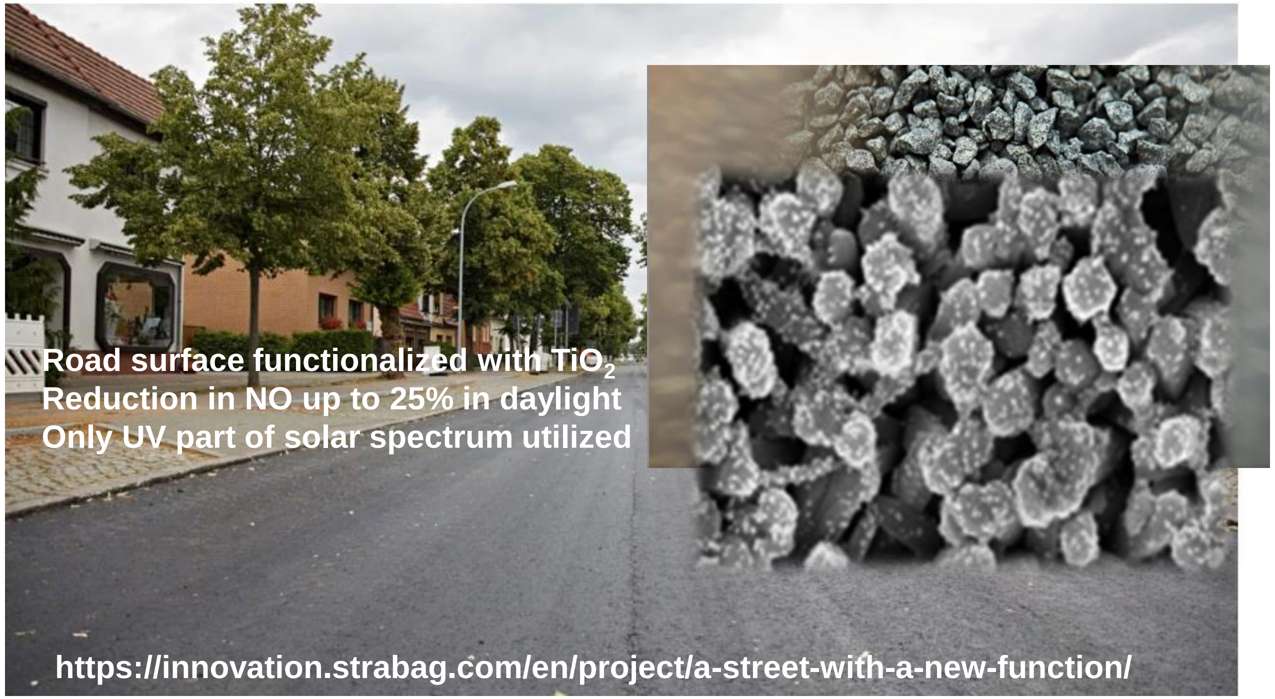


FLASH challenge

Stable, narrowband source for XPS

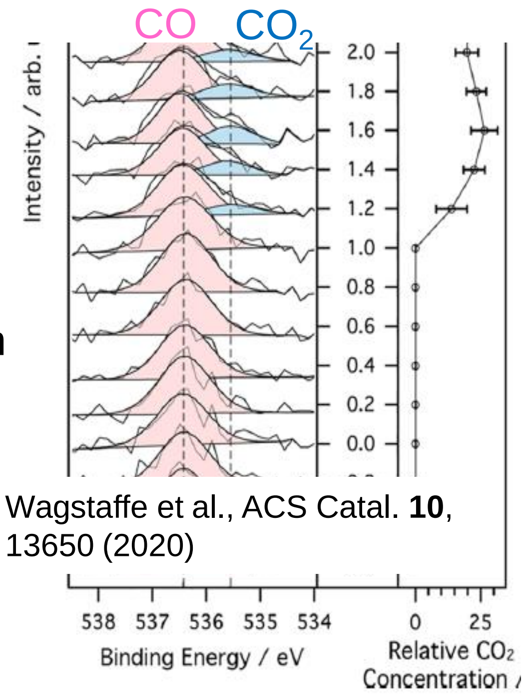
Photocatalysis innovation

- Plasmonic nanoparticles for sensitizing cheap UV photocatalysts
- Needs ultrafast x-rays to identify losses in charge transfer and bond activation

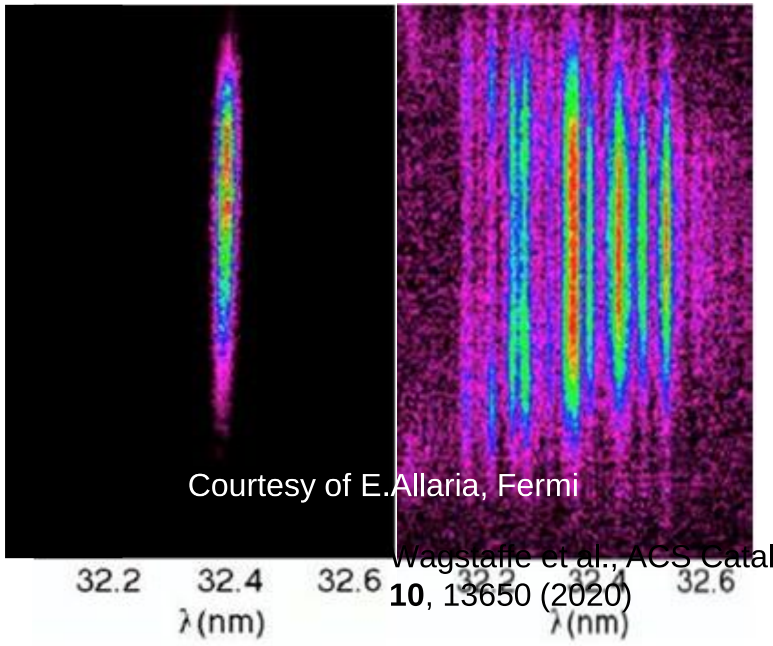


X-ray photoelectron spectroscopy with seeding

- XPS needs high-rep. rate source to avoid space charge broadening
- Statistical pulses from SASE means strong fluctuations on space charge per pulse
- Seeding makes any XPS method much faster



Wagstaffe et al., ACS Catal. **10**, 13650 (2020)

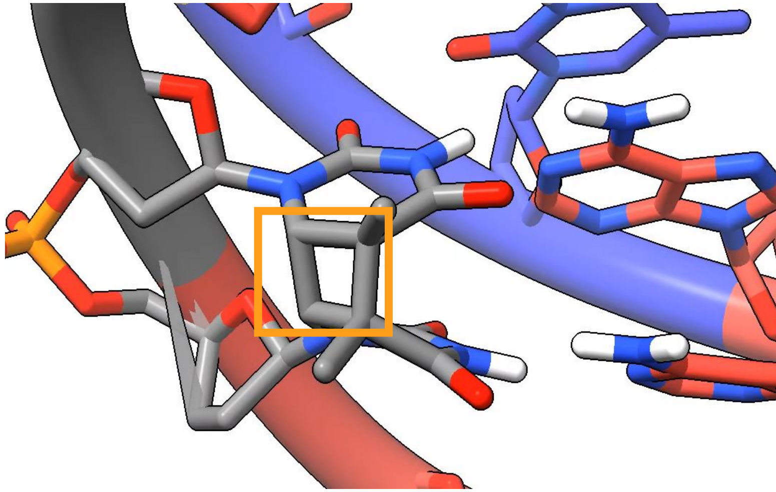


Courtesy of E.Allaria, Fermi

Wagstaffe et al., ACS Catal. **10**, 13650 (2020)

FLASH challenge

Controlled solvation/photochemistry/biological function

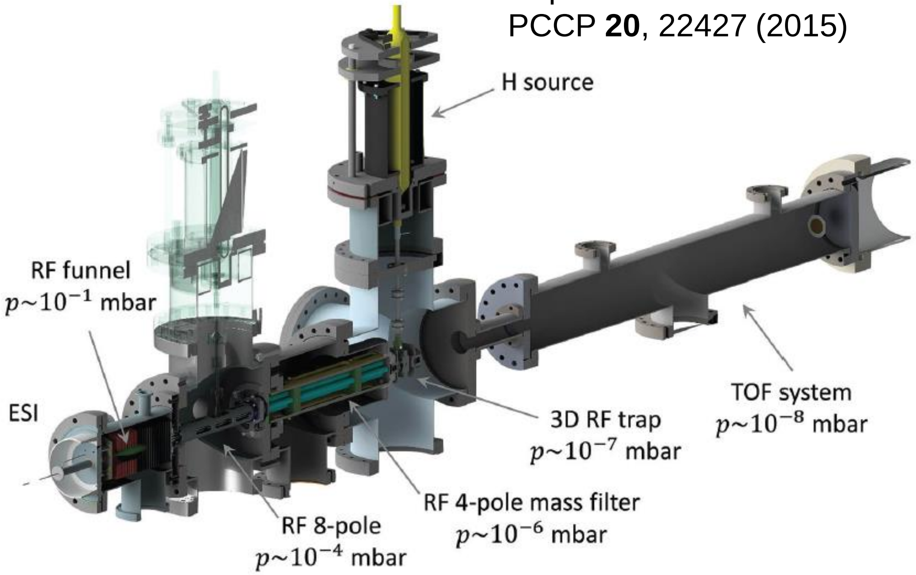


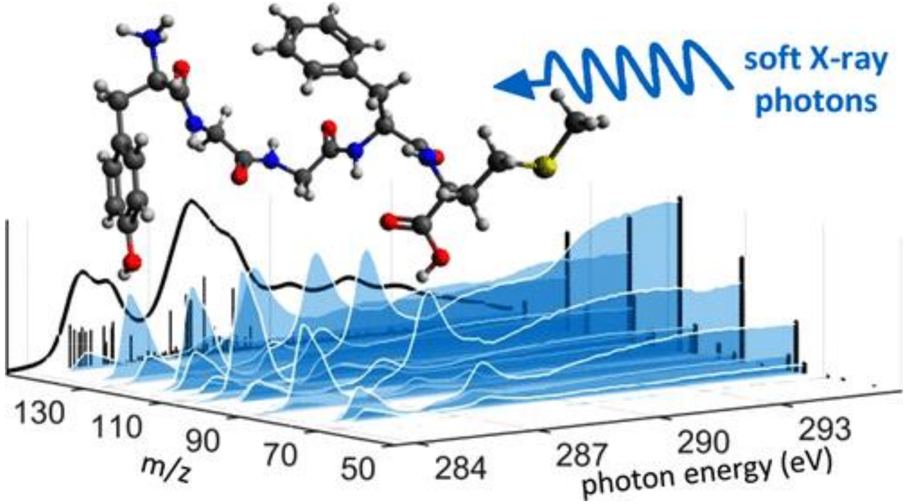
- All these efforts require ion-trapping and mass-spectrometry and ion mobility as novel tools
- Example here: UV induced lesions in biomolecules
- Ion-traps require long trapping and readout times, ideal for FLASH burst mode
- Averaging over whole burst ist required – seeded FLASH necessary
- **Funding + community building** together with FELIX and synchrotrons with a LEAPS workpackage

European Commission

LEAPS

Rapacioli...Schlathölter
PCCP **20**, 22427 (2015)





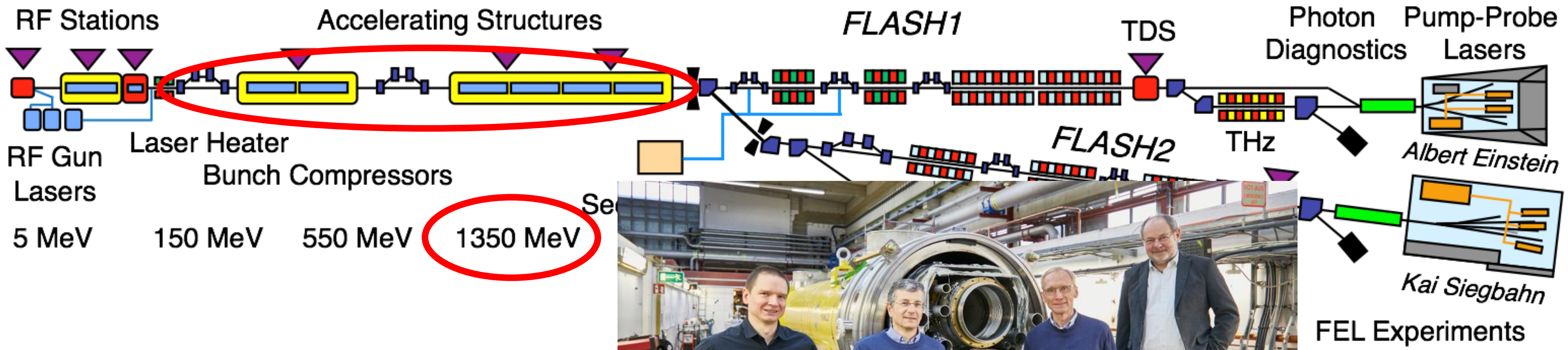
Dörner…Bari, J. Am. Soc. Mass Spectrom
32, 670 (2021)

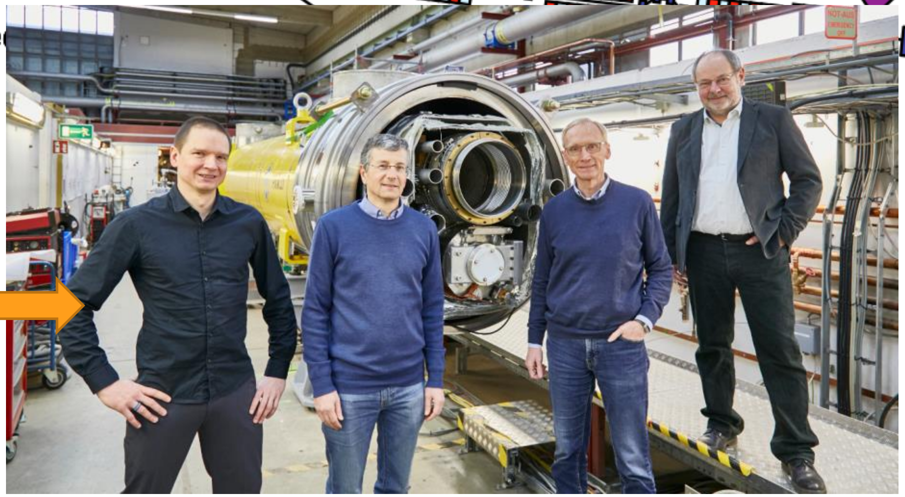
FLASH 2020+

Project overview

Energy upgrade — **Externally seeded operation**

2021 2022 2023 2024 2025

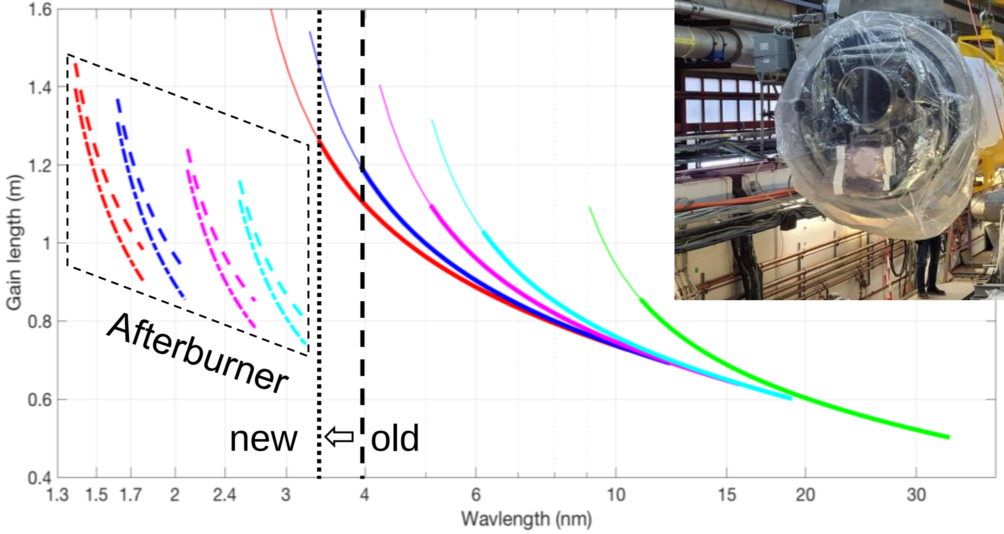


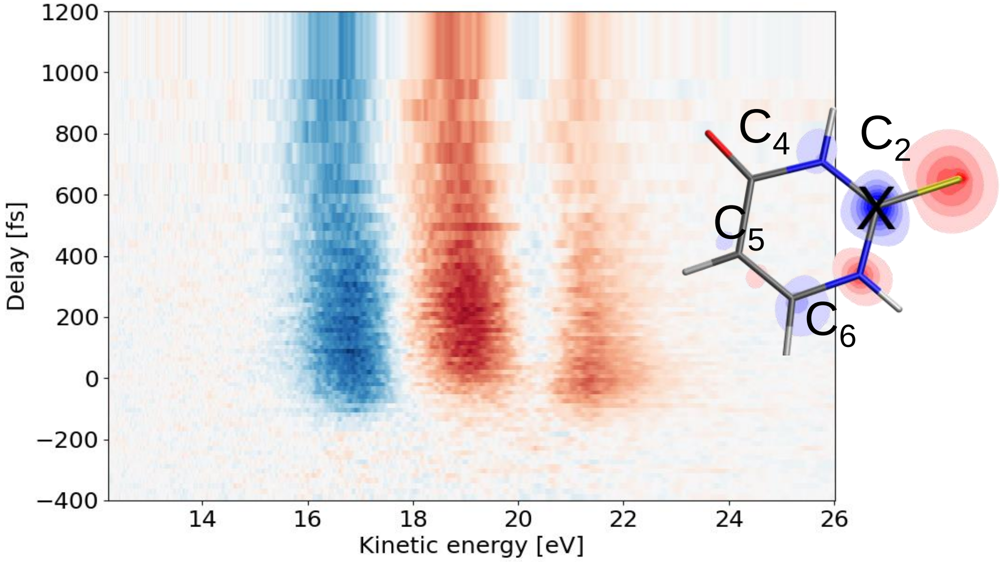


Courtesy Lucas Schaper
Project leader 2020+

First shutdown was a success!

New electron energy of 1.35 GeV allows pushing short wavelength limit





In FLASH shutdown 21/22 a major part of the linear accelerator was renewed

- 2 out of 7 acerating modules replaced → 100 MeV higher electron energy
- Allows for generation of shorter wavelength FEL radiation (red curve)

Courtesy: Lucas Schaper

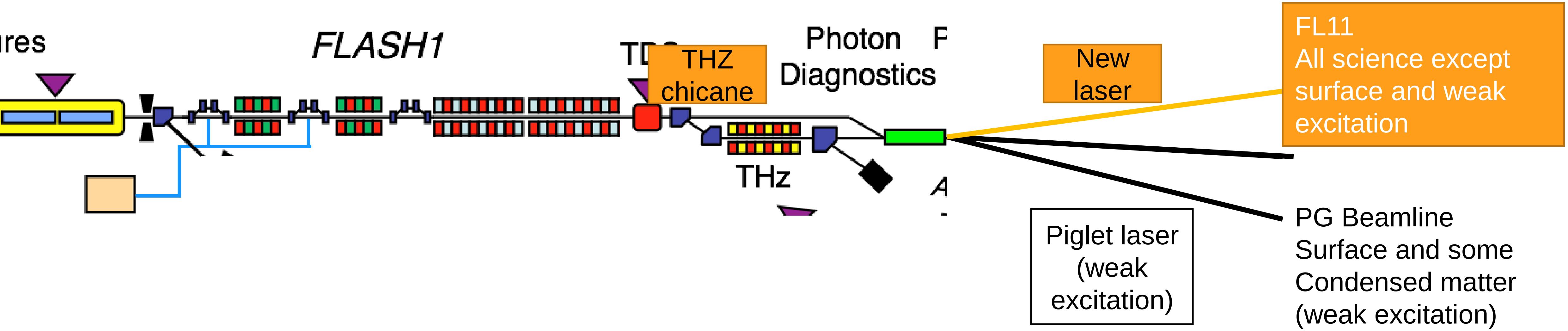
First beamtime after shutdown:
Time-resolved experiment with UV excitation and x-ray probe at 330 eV – enabled by the higher electron energy

Sample is a sulfur-substituted nucleobase, with applications in medicine

Extended x-ray energy allows to observe the electronic dynamics of the molecule at different sites - here carbon

FLASH 2020+

Project overview

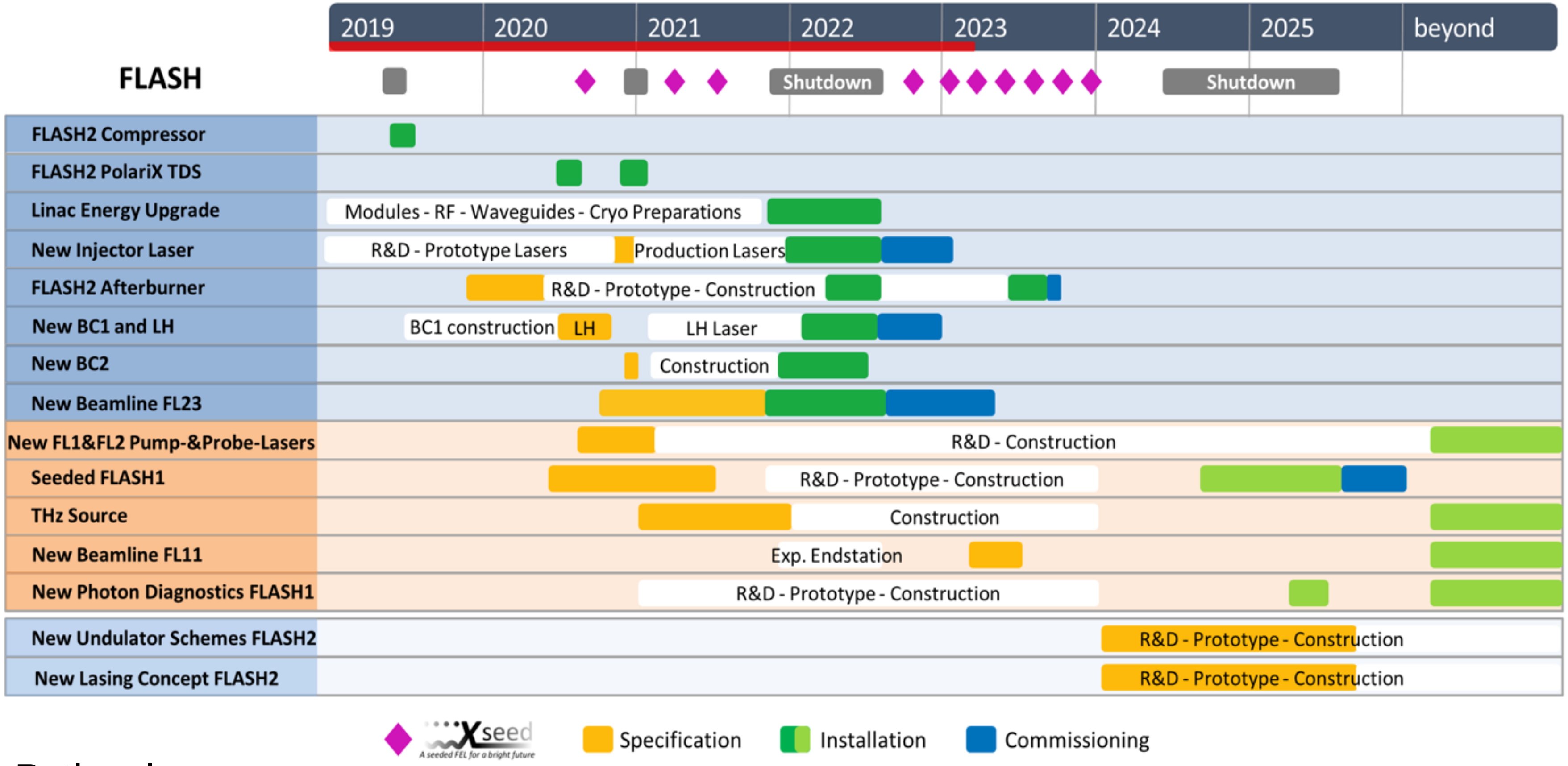


Major items:

- Seeding infrastructure
- THz chicane
- Photon diagnostics
- New beamline FL11 with new pump-probe laser

FLASH2020+ upgrade cuts

Courtesy: Lucas Schaper

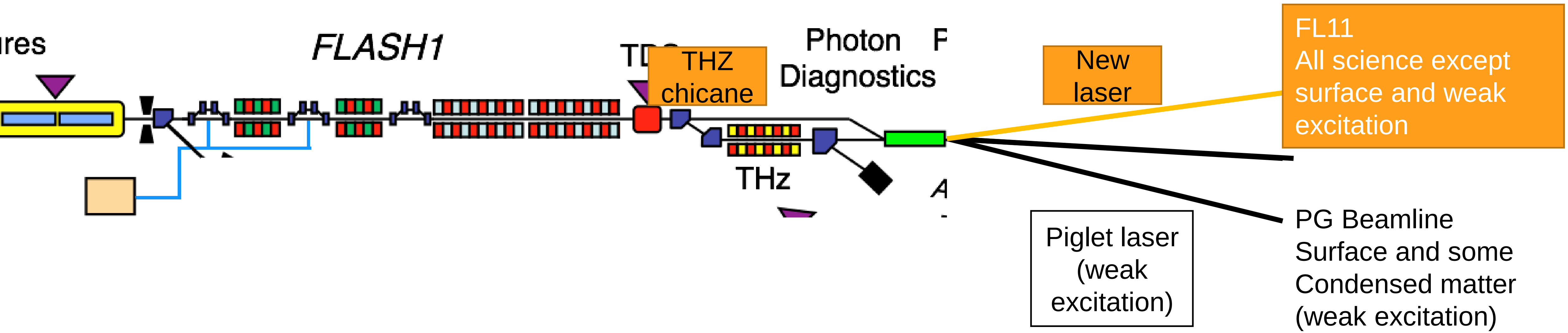


Rationale:

- Time: Finish everything inside tunnel before XFEL shutdown in second half of 2025
- Money: Current financial situation does not allow laser, THz, FL11, new photon diagnostics at end of 2025, but allows seeding
- Financing Model: major funds come out of the running budget of FLASH in M and FS divisions
- Crucial: scrutiny and control on operating budgets on M and FS side to finance everything up to seeding
- we must start with undulator purchasing now, there is no way back afterwards

FLASH 2020+

Project overview



We are actively seeking alternative funding

We might need your help with that!

Some operating funds in 26-27 can also be used, assuming we don‘t use them before for forward funding of seeding

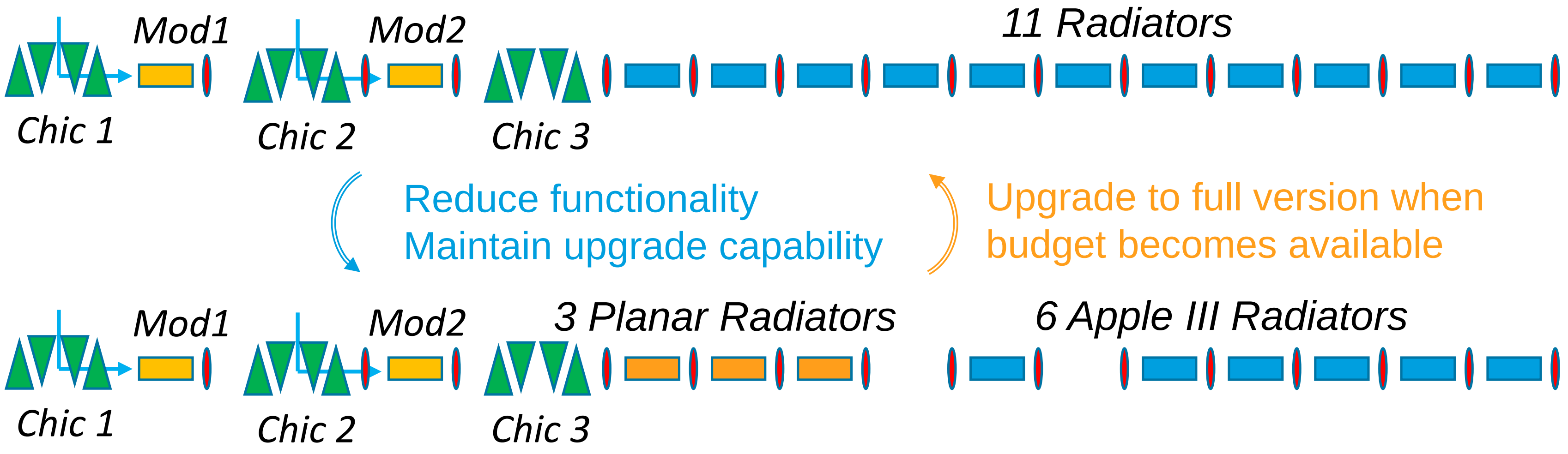
Seeded FLASH1 (reduced)

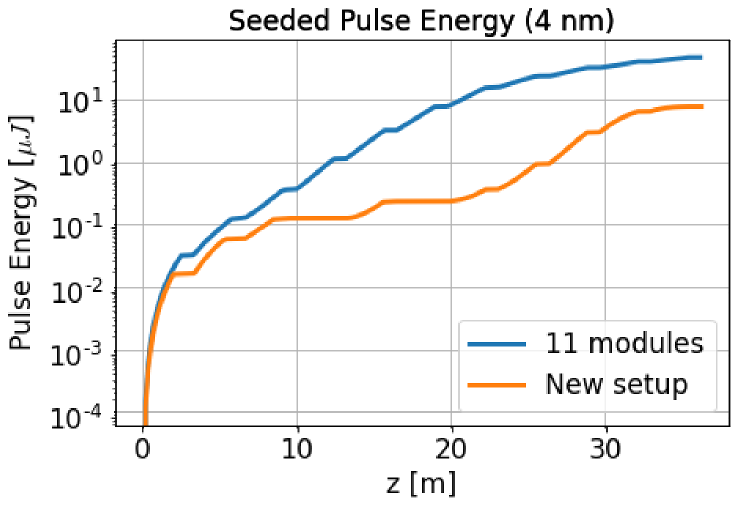
Courtesy Lucas Schaper

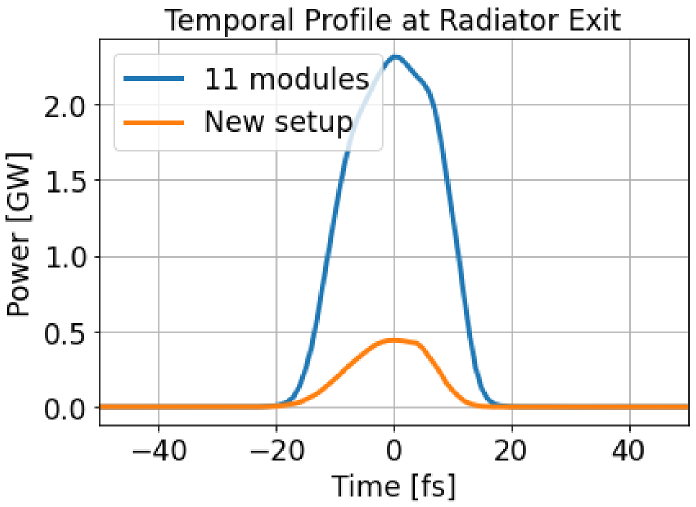
Adaptions required to stick within new funding frame

What had been envisaged in CDR:

- **Fully coherent** pulses with
- **Variable wavelength (60 – 4 nm)**
- **Tens of fs** duration
- **1MHz repetition** rate and
- **Variable polarization**

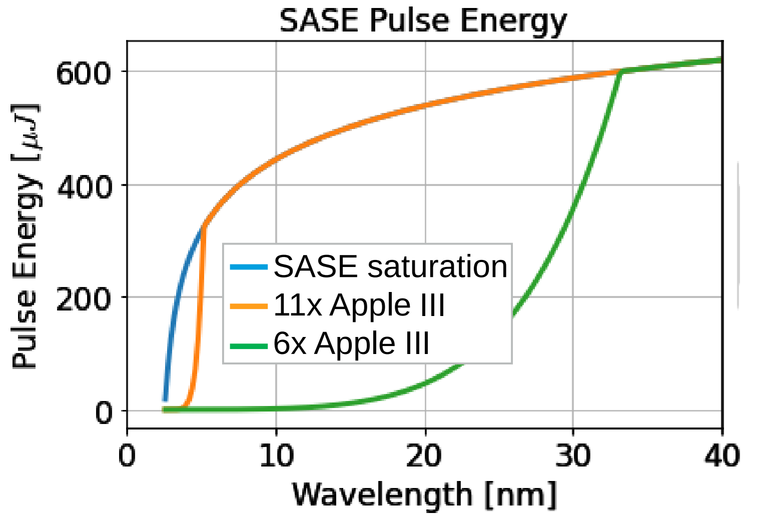






Output power reduced by a factor of 5

Polarisation contrast reduced to about 50



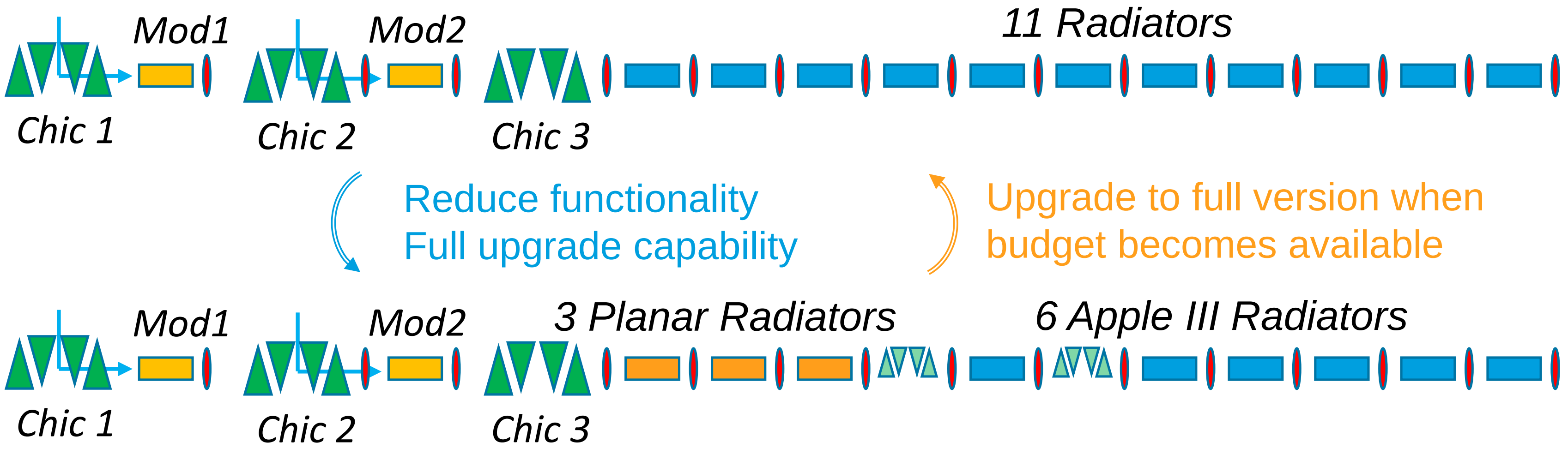
Opportunities

Courtesy Lucas Schaper

Fill the void

What will be realized:

- **Fully coherent** pulses with
- **Variable wavelength (60 – 4 nm)**
- **Tens of fs** duration
- **1MHz repetition** rate and
- **Variable polarization**



Gaps are possibilities

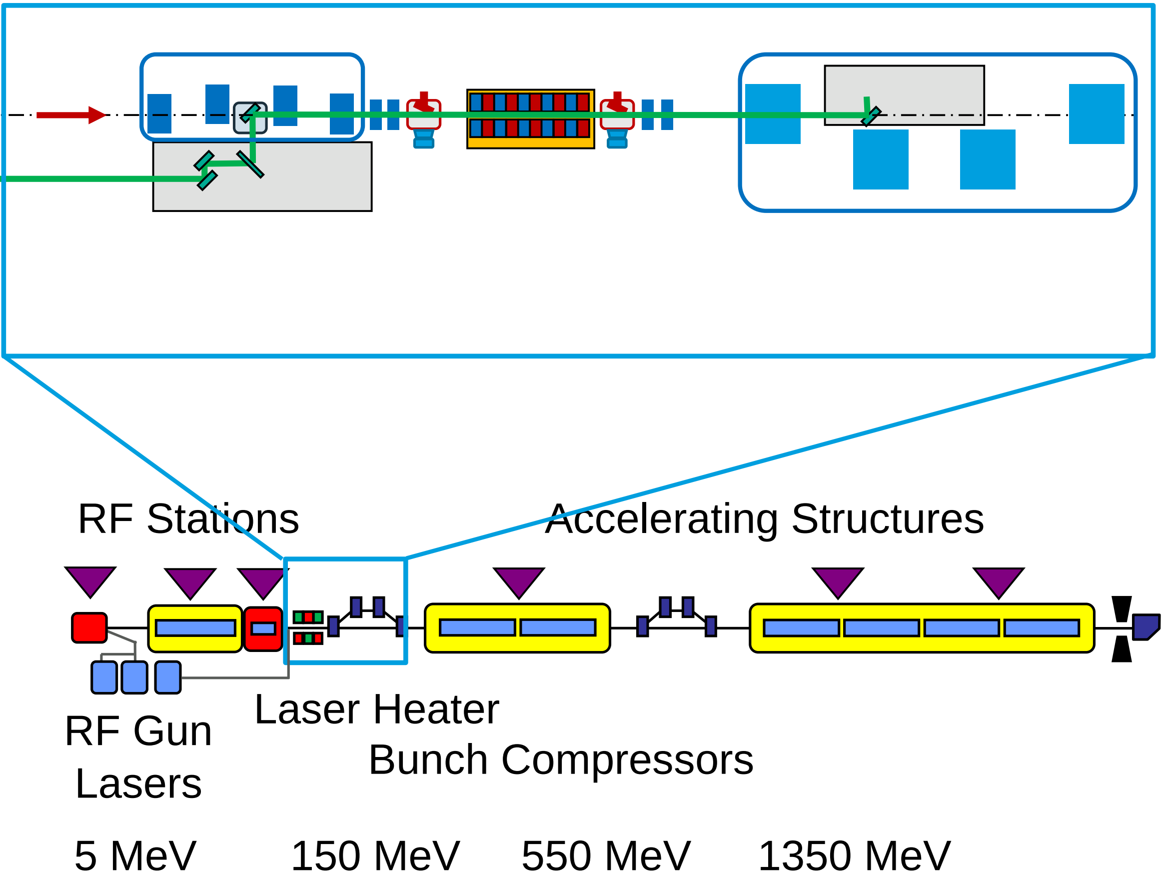
- Allows for new FEL science via installation of comparably cheap chicanes
  - Additional bunching options, fresh slice techniques, advanced seeding schemes, …

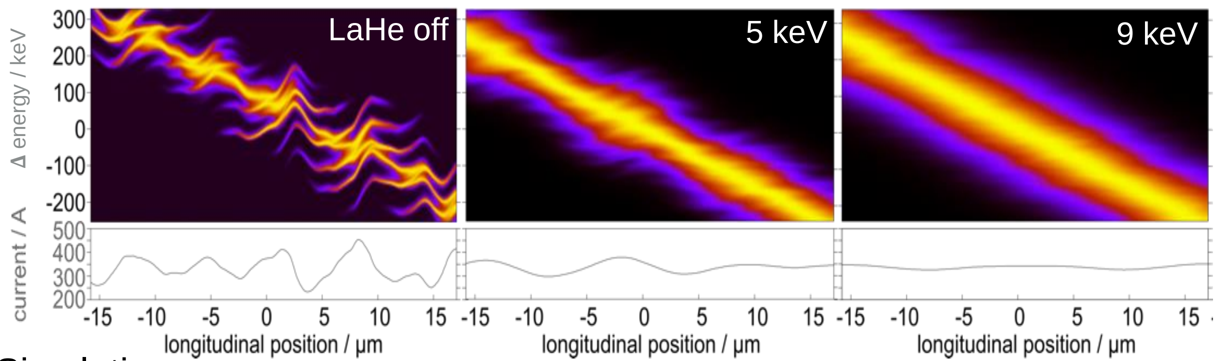
Stability is key: FLASH 1 will appeal to larger user community & new users

Smoothing electron bunches

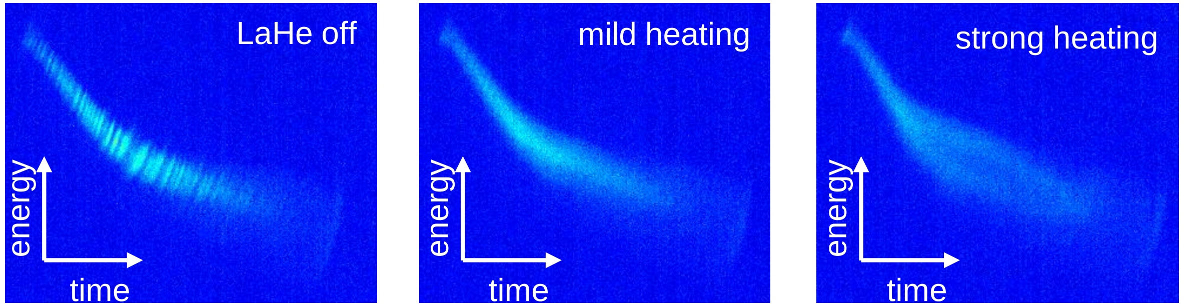
Reducing microbunching gain via laser heater

Courtesy Lucas Schaper

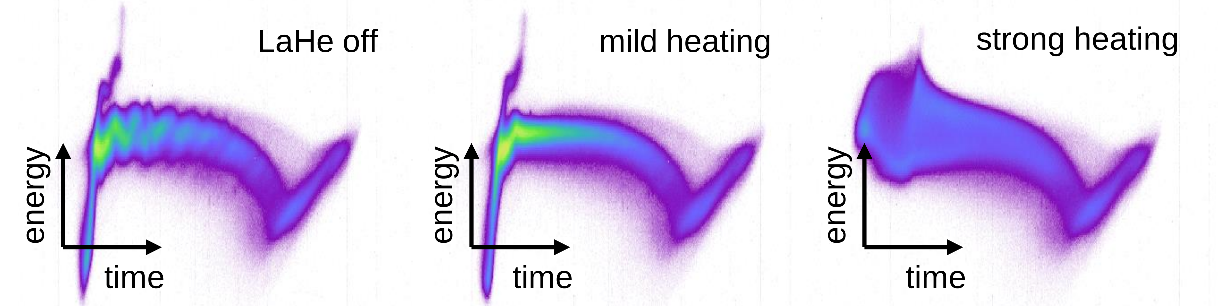




Simulation



FLASH1-LOLA

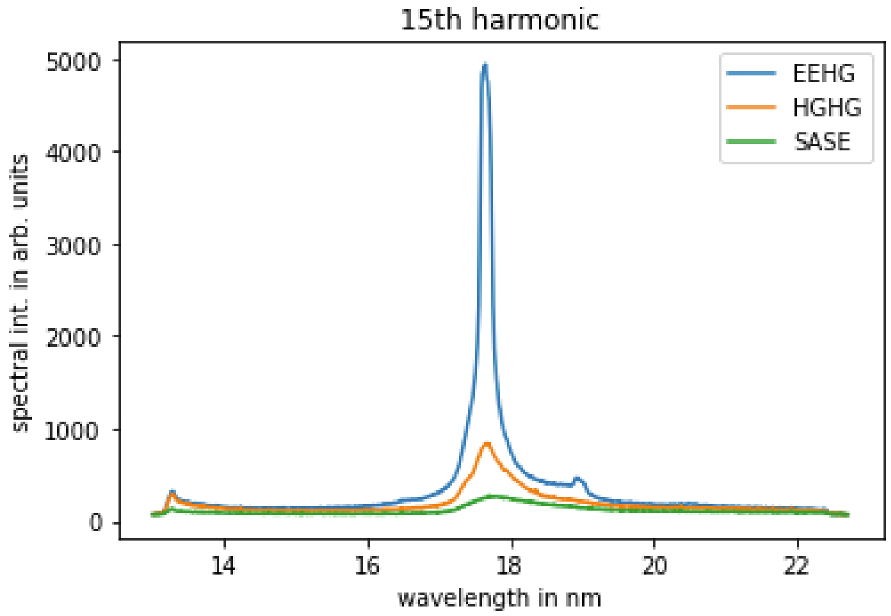


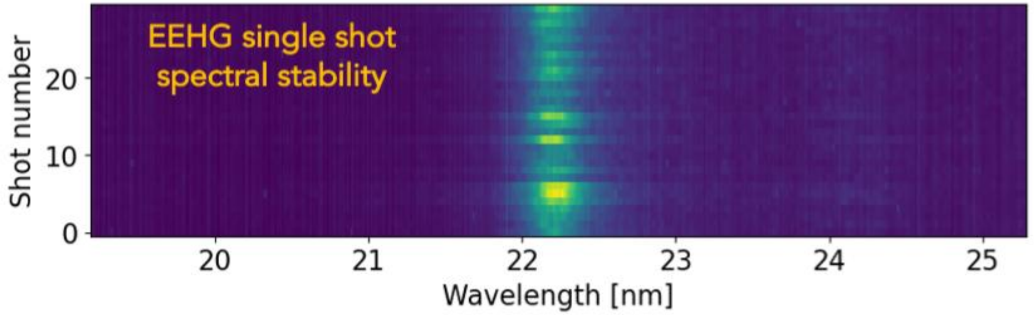
FLASH2-PolariX

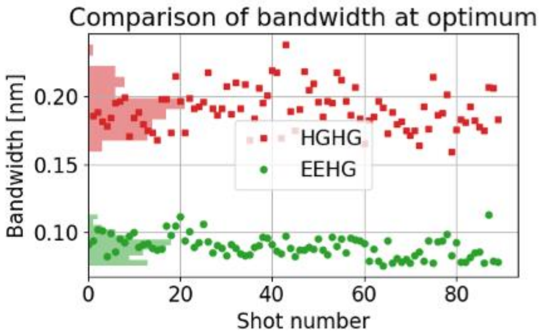
A major success: EEHG at FLASH

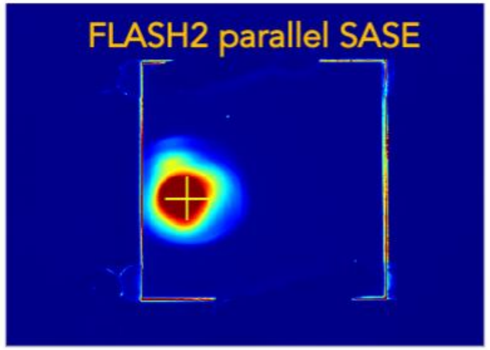
Worldwide unique: parallel operation of EEHG and SASE

- Initial EEHG setup looking at 12th harmonic took 3 full days!
  - Characterizing 12th harmonic
- Complete re-setup in only 4 hours
  - Characterizing harmonics 9, 12, 15 and 17
- Successful seeding of 2nd bunch
  - mimicking bunchtrain operation → full train to come
- Parallel SASE operation at FLASH2 with 30 bunches above 100uJ at 30 nm
- **A major team effort!**
  - Expertise vital for future seeded FLASH1
- Continued experiments offer unique chance
  - Shorten commissioning time
  - Develop tools and procedures
  - Allow early stage science experiments









Courtesy Lucas Schaper

Thanks for your attention

Thanks to the team at FLASH