(Linear) – XUV and Soft X-ray Spectroscopy at FLASH, LCLS and FERMI

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Source comparison

FLASH LCLS FERMI Sincrotrone Trieste

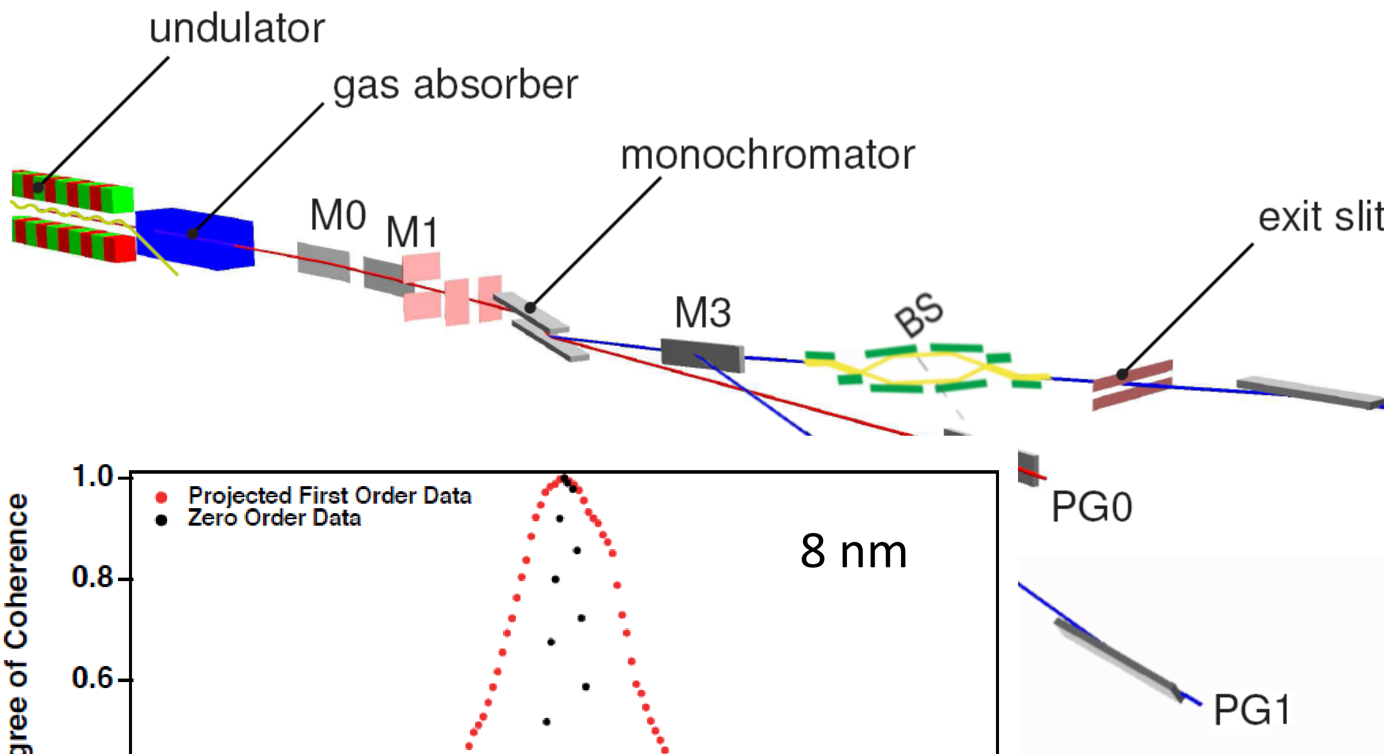
	Photon energy	Photons/ pulse	Pulse duration	Spectral bw. (FWHM)	Pulses/ s	Polarization
FLASH	30-300	$\leq 10^{14}$	3 -200fs	~1%	$\leq$ 8000	Linear
LCLS (SXR)	280-2000	10^{13} 10^{11}	10-200fs	~0.5%	120	Linear Circular
FERMI FEL-1 FEL-2	 12-60 (100) 60-310	 10^{14}-10^{13} 10^{13}-$3\cdot10^{11}$	 <100fs <100fs	 <0.1% 0.1-0.2%	 10-50 10-50	 Variable Variable

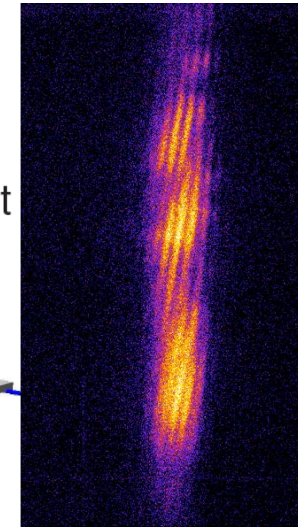
SASE requires monochromatization (SXR@LCLS, PG@FLASH)

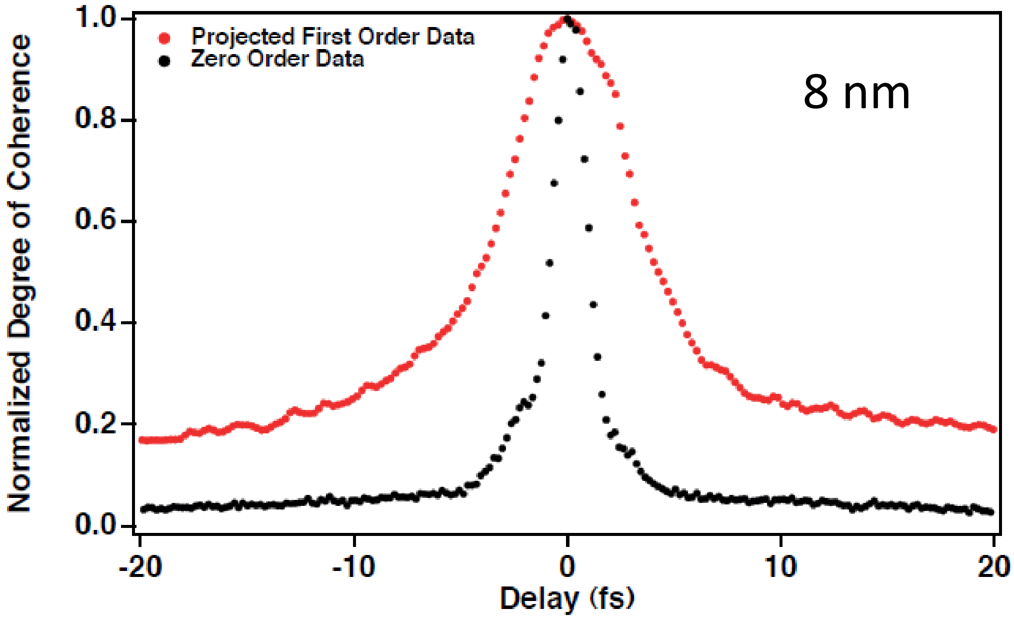
SXR: W.F. Schlotter et al., Rev. Sci. Instr. 83, 043107 (2012)

PG: M. Martins et al. Rev. Sci. Instrum. 77, 115108 (2006)

SASE + mono: temporal coherence



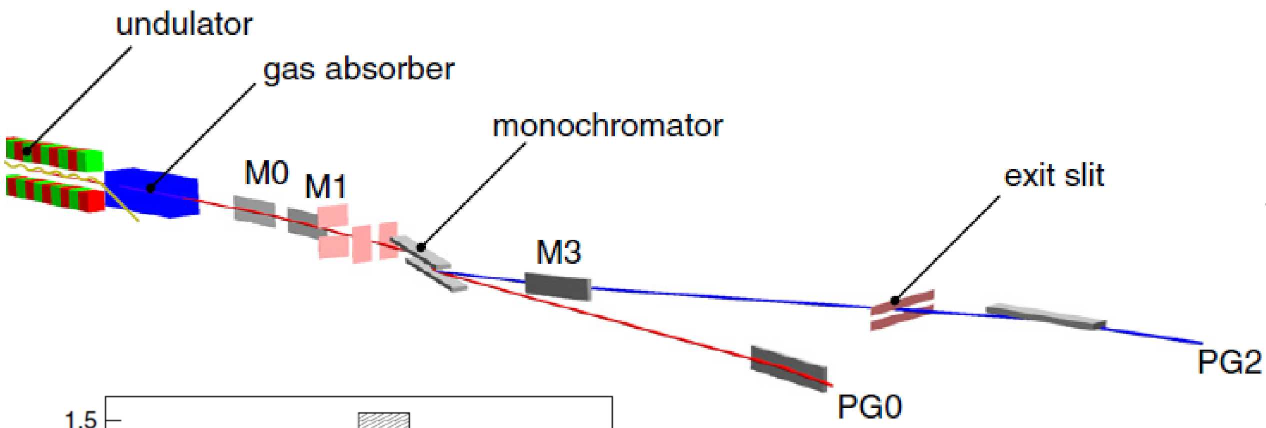




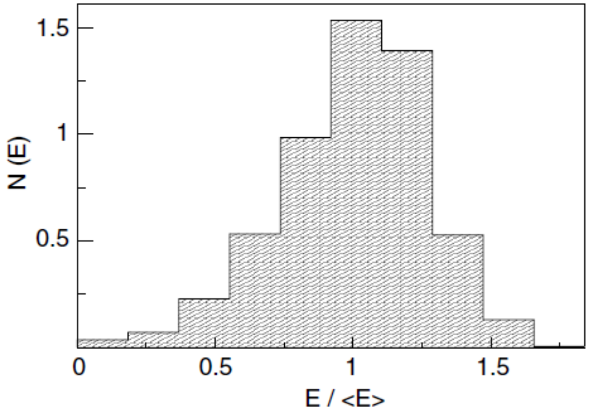
Longitudinal coherence increased

F. Sorgenfrei PH.D thesis Universität Hamburg (2012)

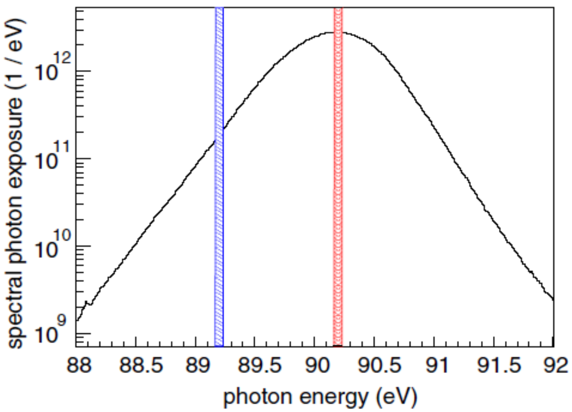
SASE + monochromator: statistical fluctuations



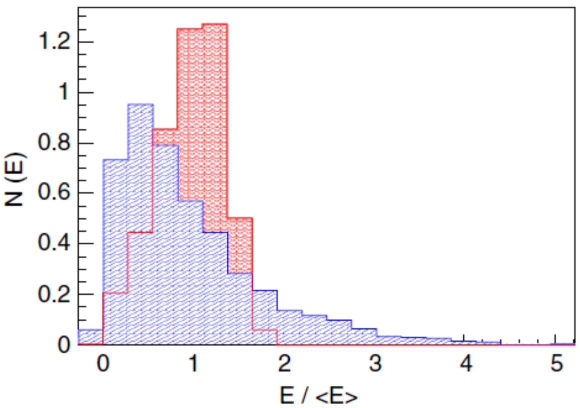
M. Wellhöfer et al., Performance of the monochromator beamline at FLASH
J. Opt. A: Pure Appl. Opt. **9** (2007) 749–756



SASE distribution

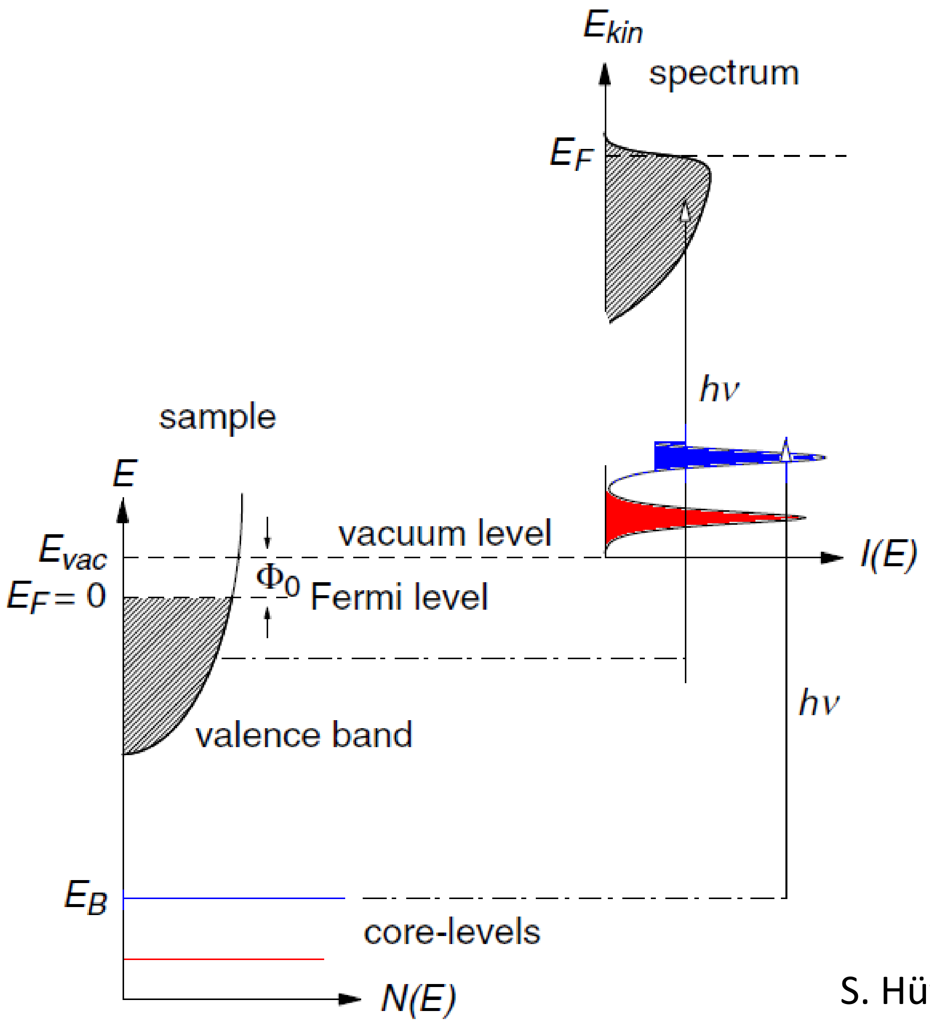


Average spectrum



Distribution behind mono

Soft X-Ray Toolbox: Photoelectron Spectroscopy



Full electronic structure information

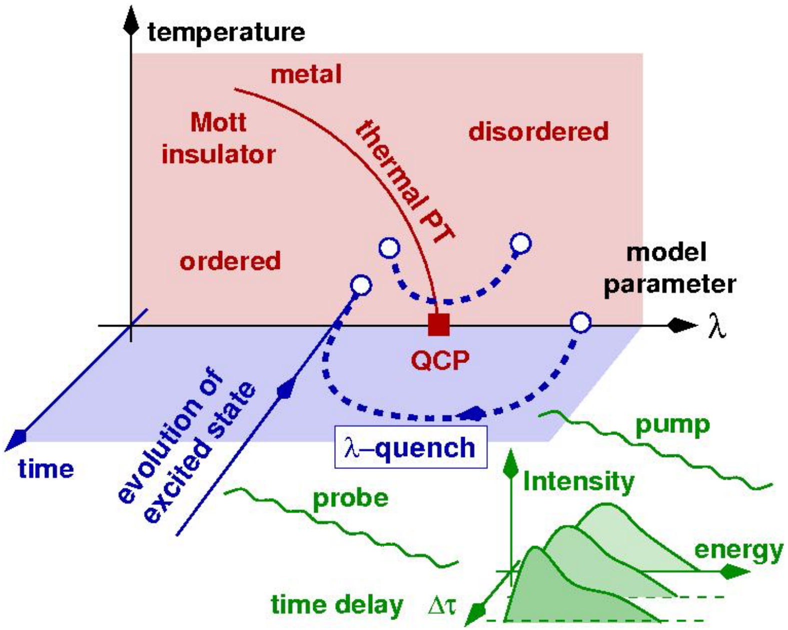
Drawback:

- Space charge

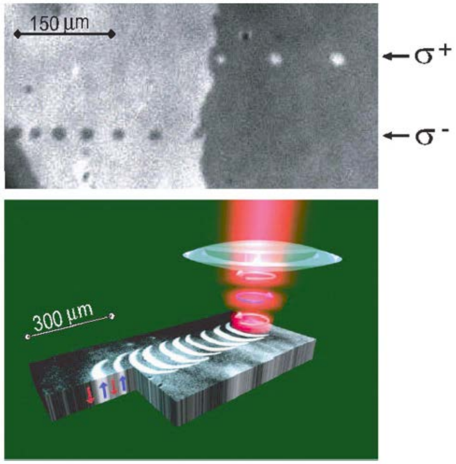
S. Hüfner, Photoelectron Spectroscopy, Springer 2008

Science drivers for TR-PES

Understand and control quantum phase transitions

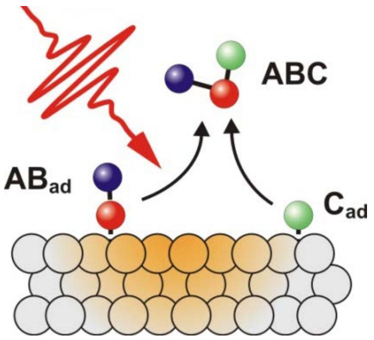


Light control of conductivity and/or magnetism



C. D. Stanciu, et al.,
PRL **99,** 047601 (2007)

Surface catalysis Observe transition states



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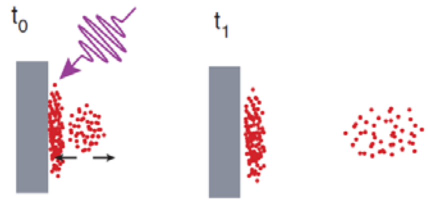
Methods	TR-SXARPES	T(σ)R-HAXPES	TR-XPS (ESCA)

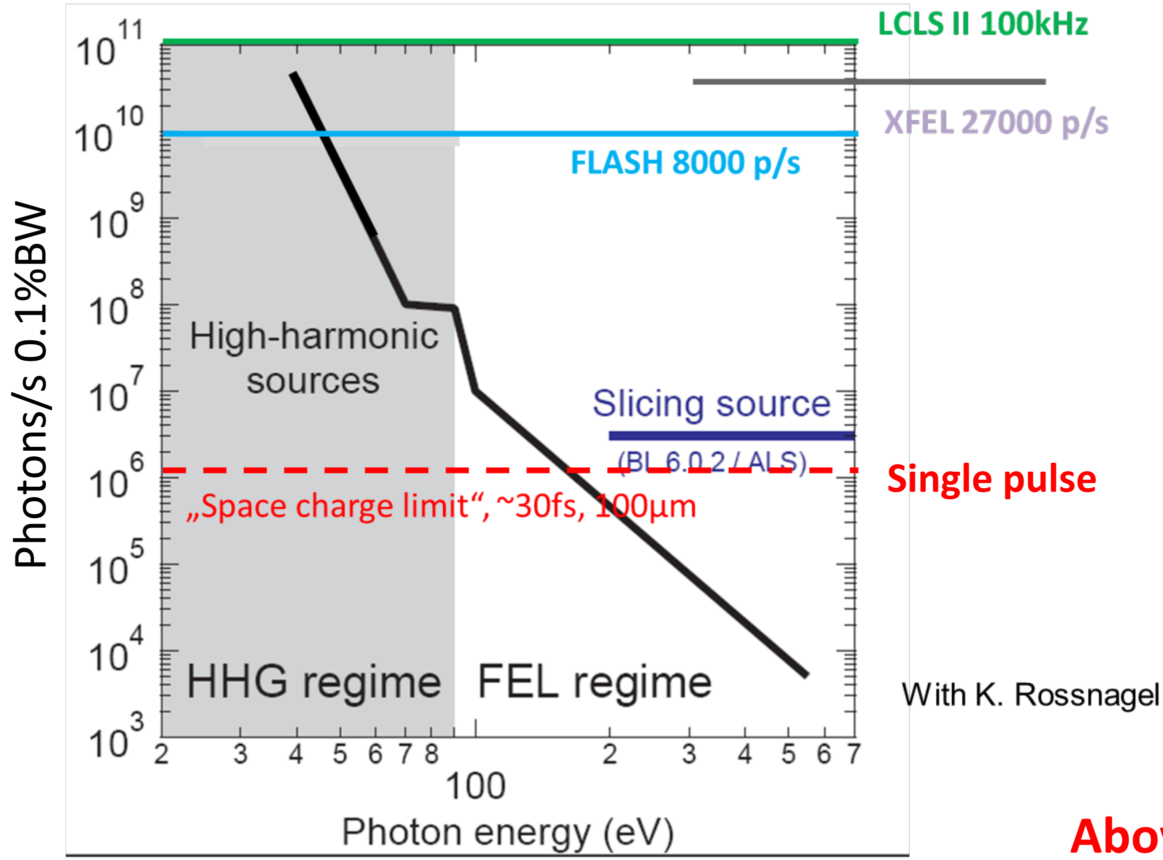
fs-photoelectron spectroscopy

Challenge space-charge



e.g. A. Pietzsch et al.,
NJP 10 (2008) 033004

However:



HHG adapted from T. Pfeifer, C. Spielmann and G. Gerber, Rep. Prog. Phys. **69** (2006) 443–505

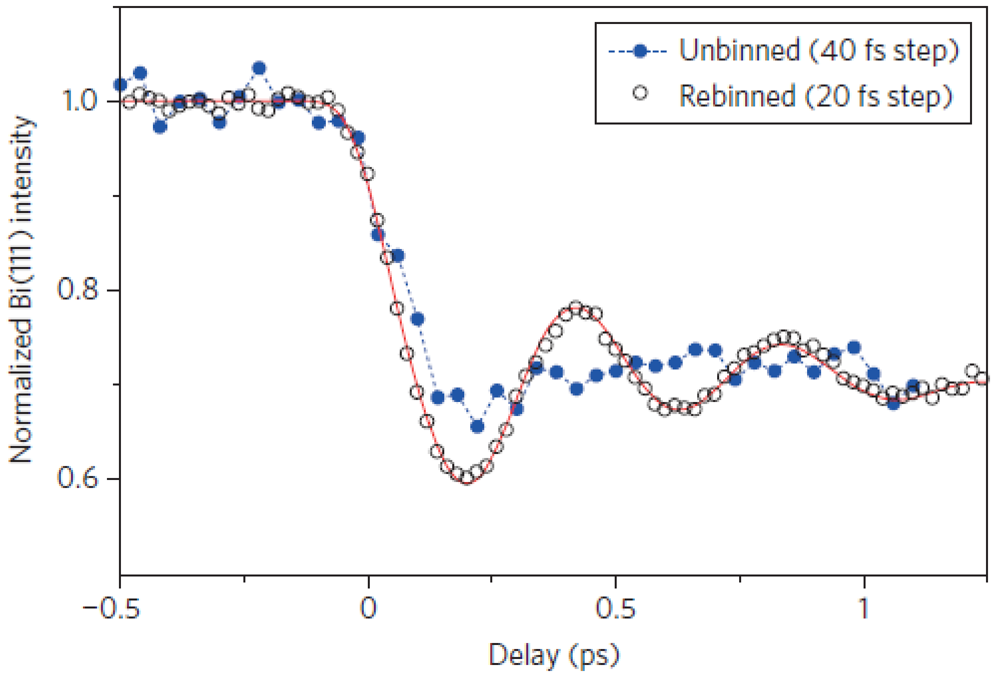
Above 200 eV <50fs
high-rep rate FEL's are absolutely unique!

Challenges for TR-exp.: Synchronisation and timing

Problem: Timing jitter between external lasers and FEL's

Solution:

a) Improved synchronisation (e.g. Seeding – FERMI 5fs rms, FLASH <30fs rms)
or b) shot-to-shot timing diagnostics



M. Harmand et al.,
Nat. Phot. 7, 215 (2013)

→ b) has to be combined with single-shot detection capabilities

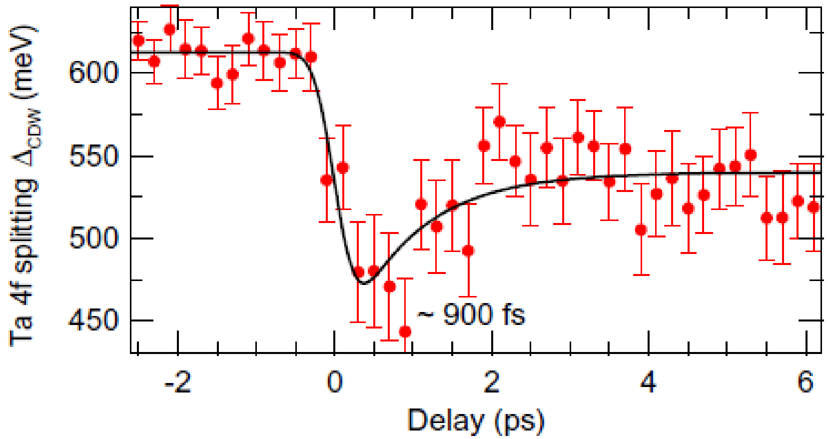
CAU

Metal-Insulator Transition

- Use XPS to monitor local charge state

S. Hellmann et al., PRL 105, 187401 (2010), ; NJP 14, 013062 (2012)

Ultrafast Melting of a Charge-Density Wave in the Mott Insulator 1*T*-TaS_2



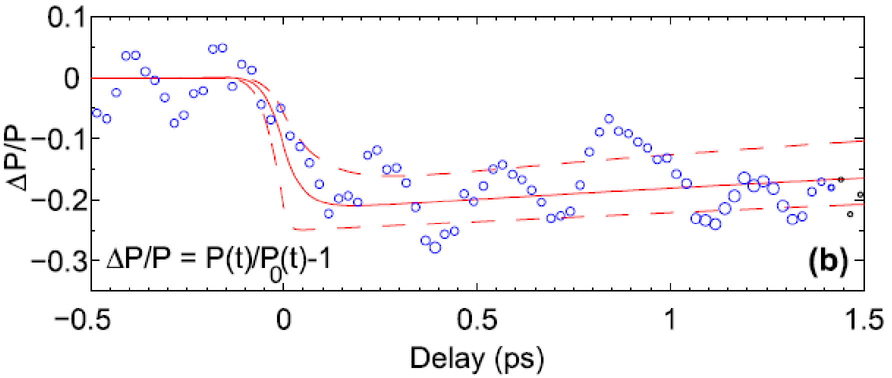
ETH zürich

Demagnetization dynamics

- Use spin-detection to monitor spin polarization

A. Fognini et al., APL 104, 032402 (2014)

Ultrafast reduction of the total spin polarization in iron

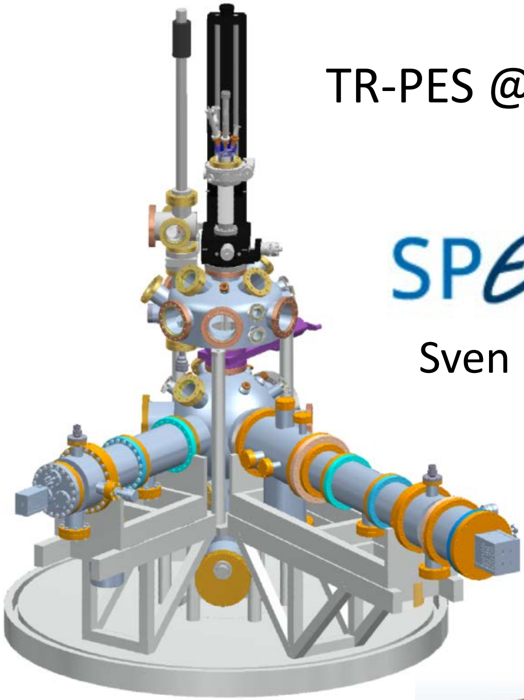


Conclusion

TR-PES needs efficient detection schemes

and high rep rate FEL sources (FLASH/XFEL/LCLS-II)

JG|U JOHANNES GUTENBERG UNIVERSITÄT MAINZ



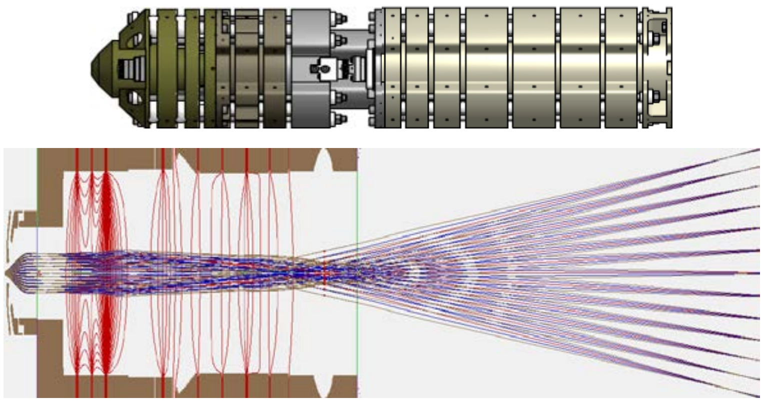
TR-PES @ FLASH

SPECS™

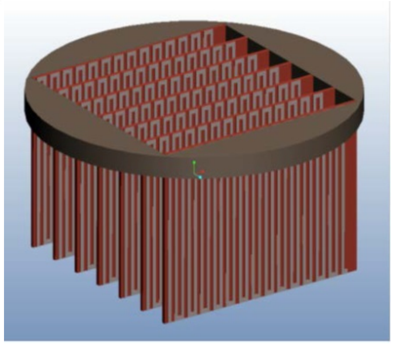
Sven Mähl

SURFACE CONCEPT

Andreas Oelsner

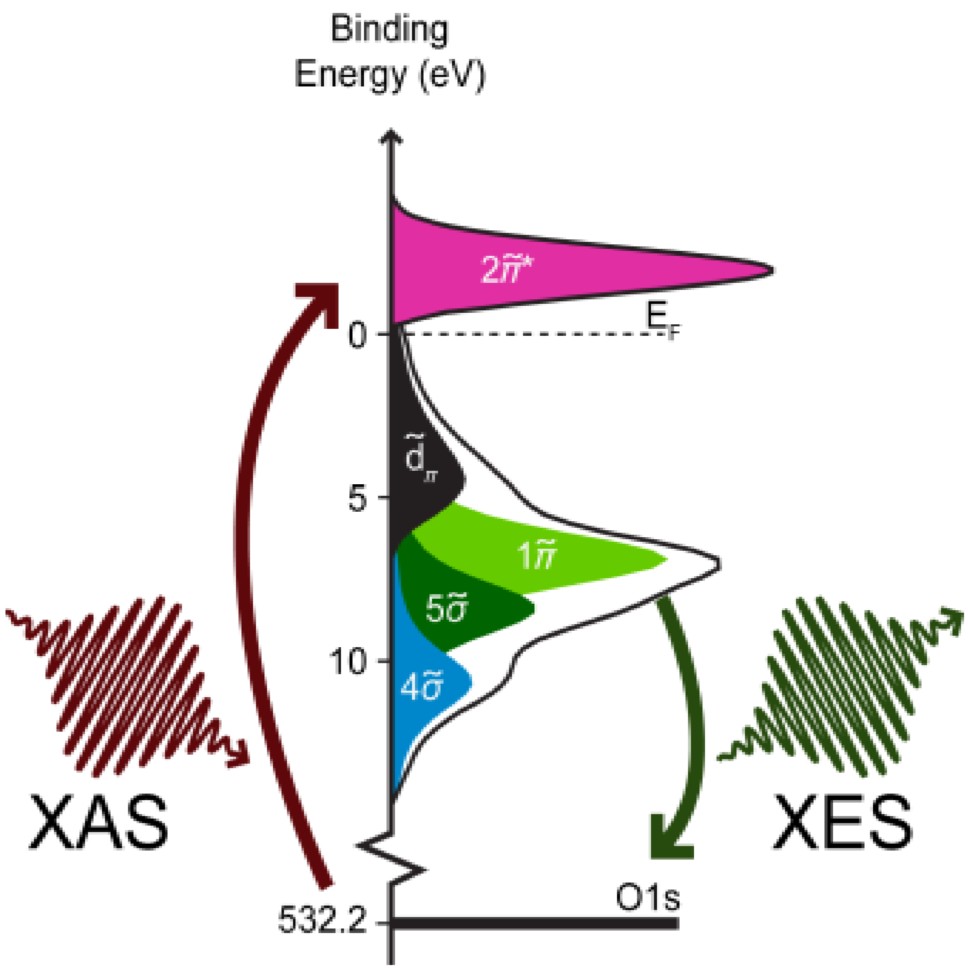


Momentum Microscope
See e.g. G. Schönhense et al., Ultramicroscopy159, 488 (2015)



Fast delay-line detector with 256 (512) independent channels

Soft X-Ray Toolbox
X-Ray Absorption and (Resonant) X-Ray Emission



Local electronic structure projected onto specific sites

Drawbacks:

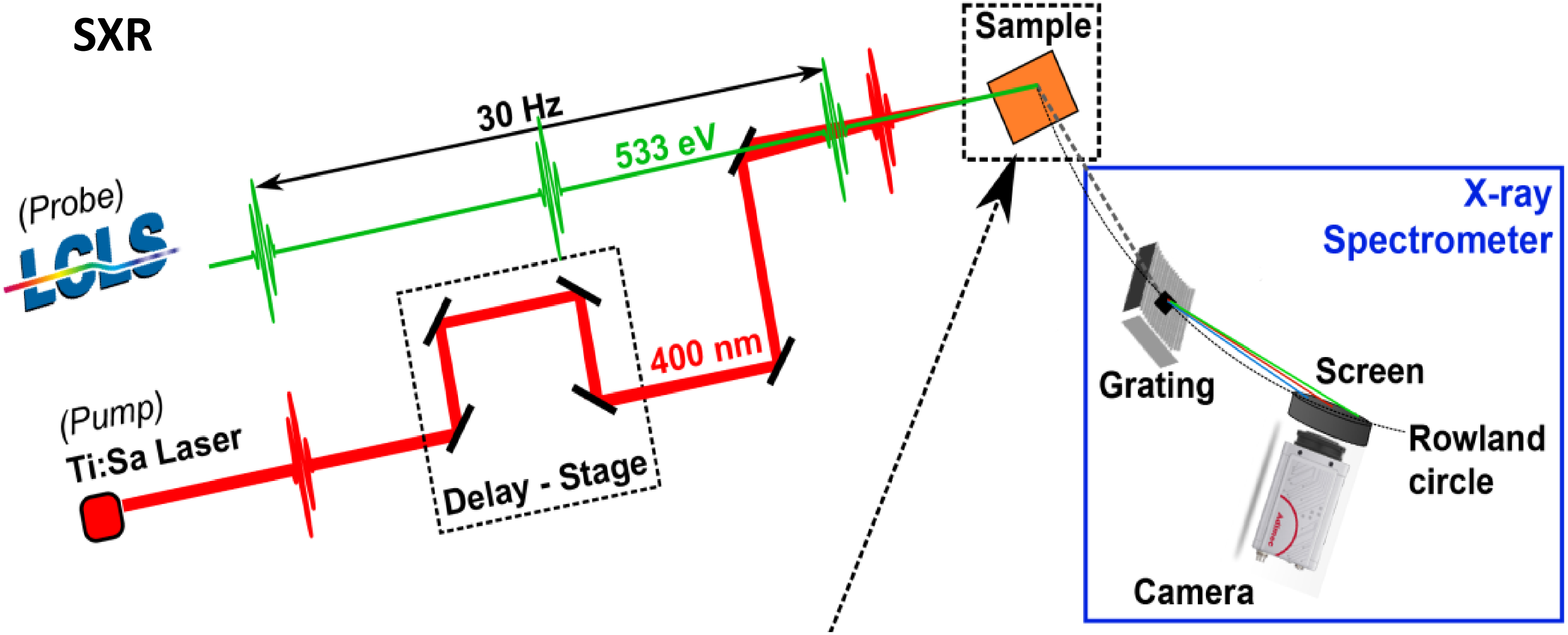
XAS

- needs widely tunable source with sufficient resolution ☑

XES

- low cross section (core excited states decay predominately non-radiatively by Auger electron emission)
- Low detection efficiency

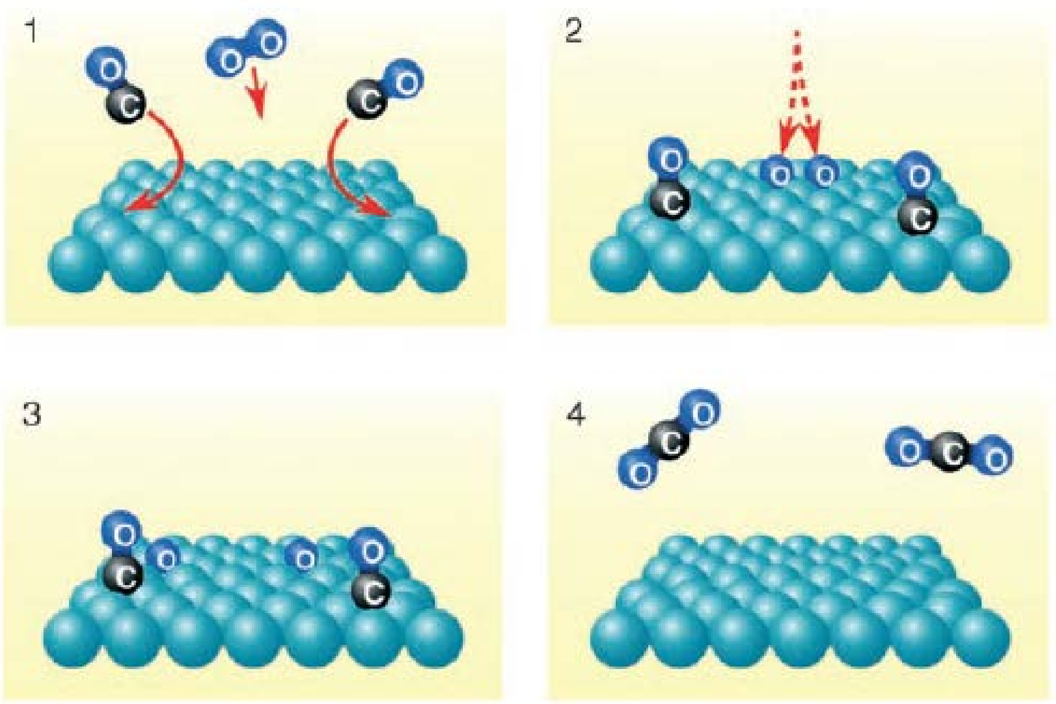
Time-resolved RIXS



Use low resolution resonant inelastic x-ray scattering (RIXS) as electronic structure probe (30 Hz, ΔE ~eV, ~mJ/cm^2)

Example: Real Time Observation of Surface Dynamics

Surface Science Collaboration at LCLS Spokesperson Anders Nilsson



Laser induced CO_2 formation on Ru(001)

CO desorption

Martina Dell'Angela et al., Science 339, 1302 (2013)

M. Beye et al., PRL 110, 186101 (2013)

T. Katayama et al, J. Electr. Spectrosc. 187, 9 (2013)

Oxygen activation

M. Beye et al. submitted

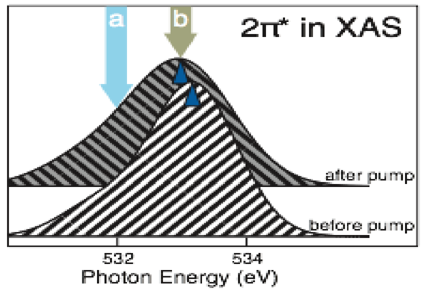
CO_2 formation

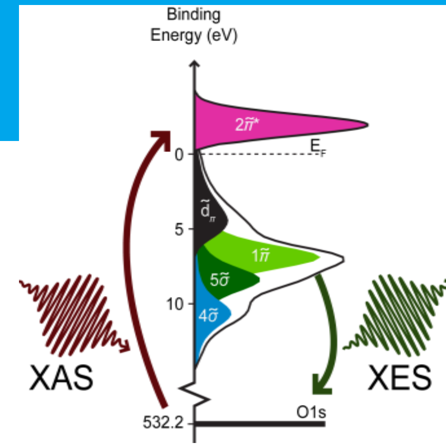
H. Öström et al., Science 347, 978 (2015)

„4-Dim“-RXES Maps – the Probe Step

XAS

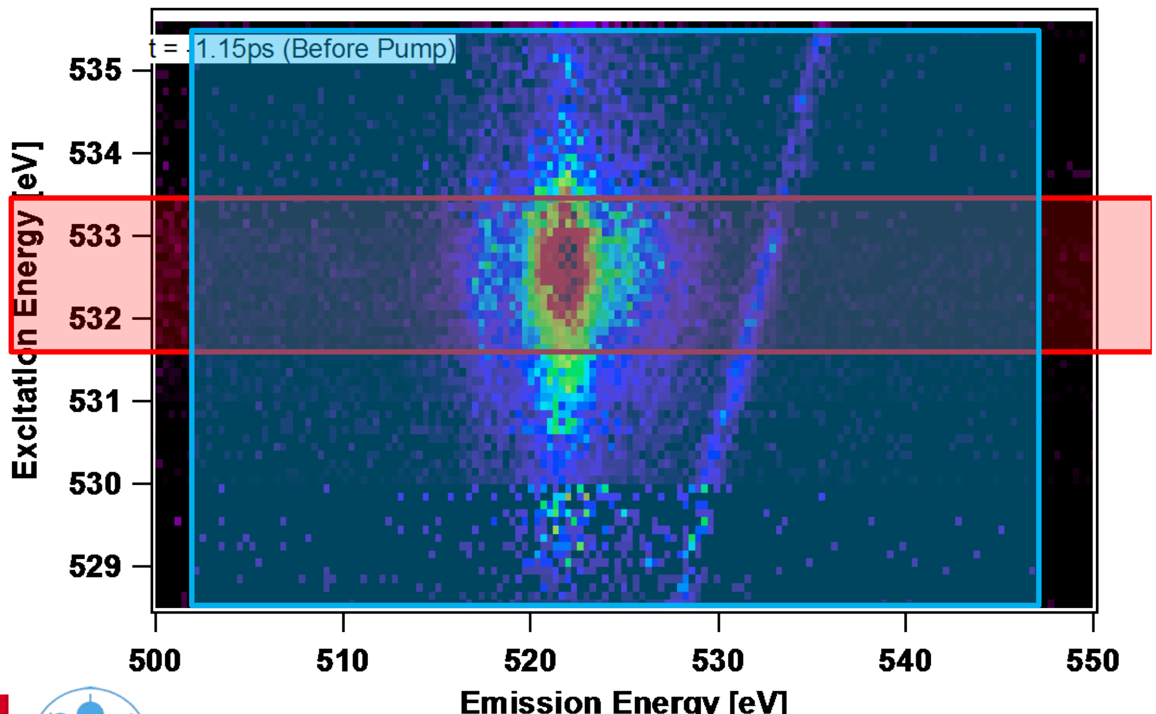
Unoccupied states

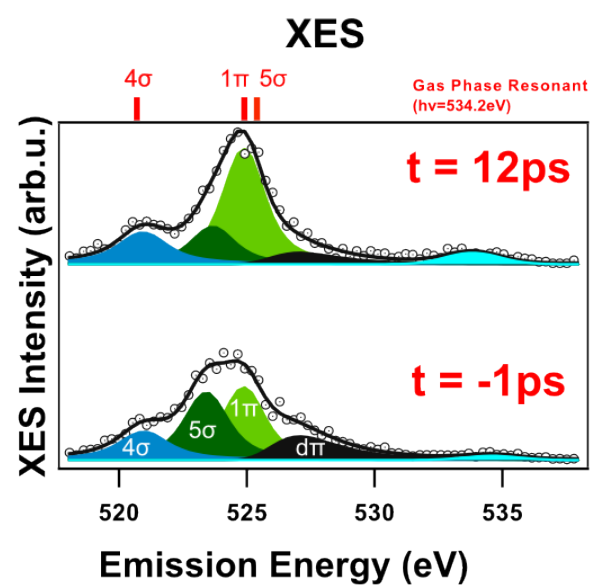




XES

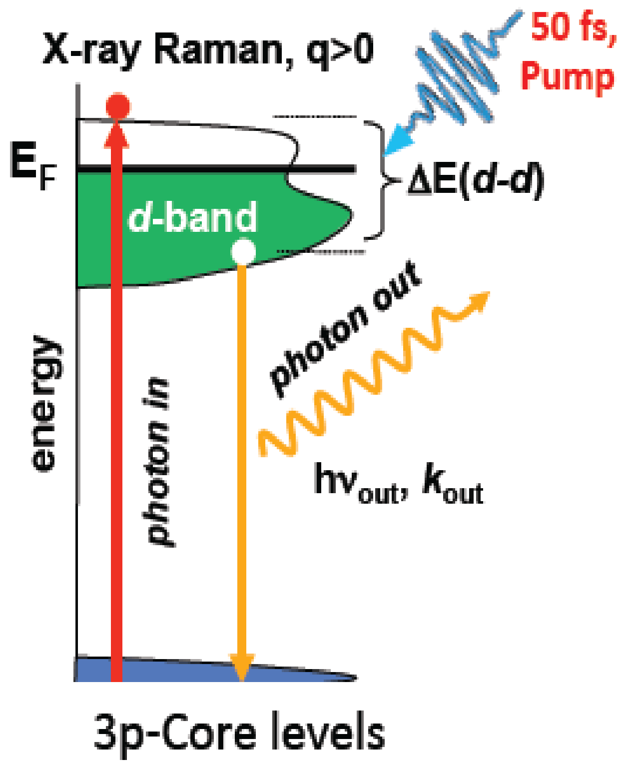
Occupied states





⇒ RIXS at (high rep-rate) seeded FEL's

„4D"-RIXS maps should greatly benefit from higher average brillance and **higher energy resolution** with seeded FEL's



Needs photon energy stability
+
easy tunability
+
~Fourier-limited bandwidth

Seeded HGHG @ M-edges with 50Hz rep. rate

$$\frac{\Delta E}{E} \sim 10^{-3}$$

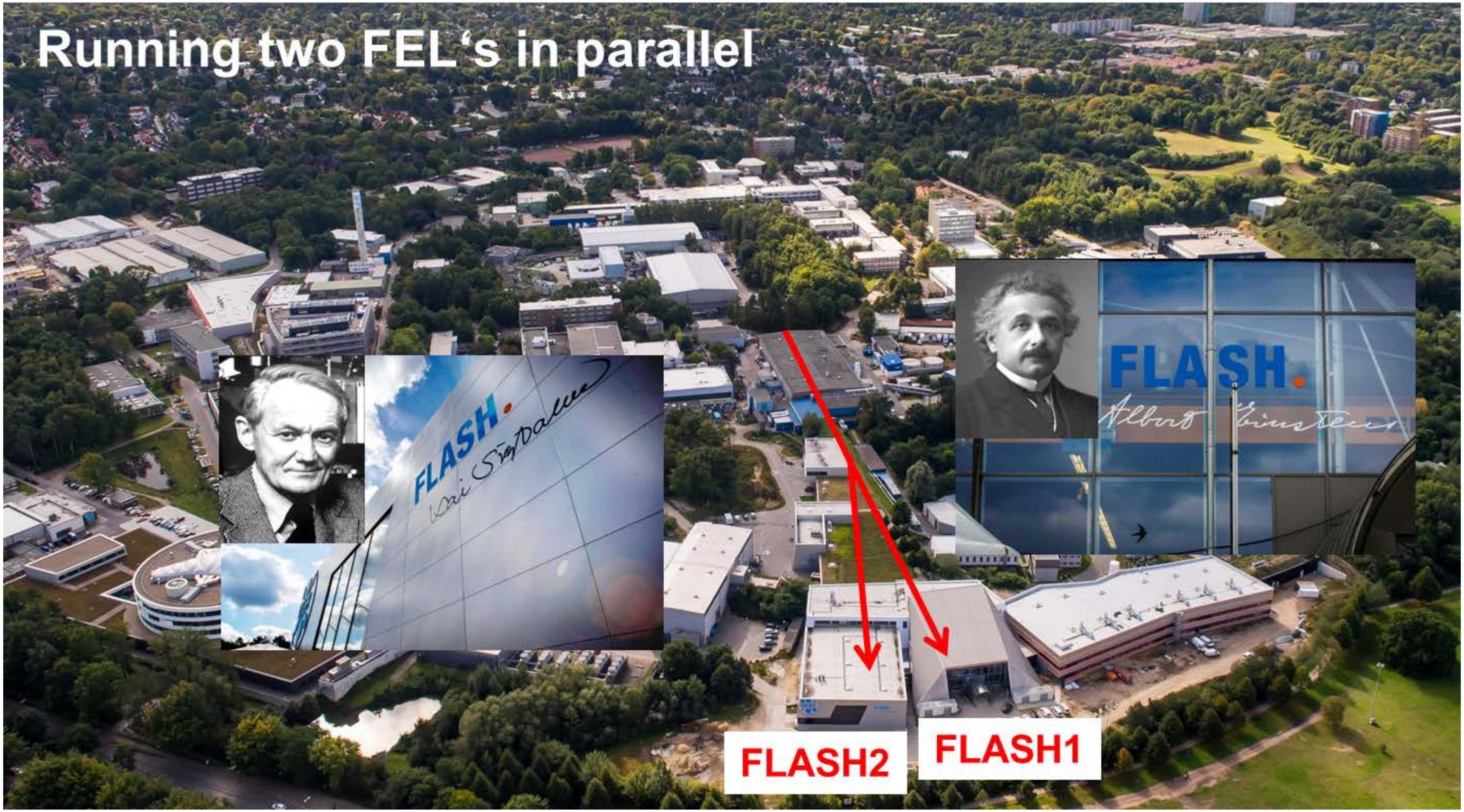
FERMI

Elettra Sincrotrone Trieste

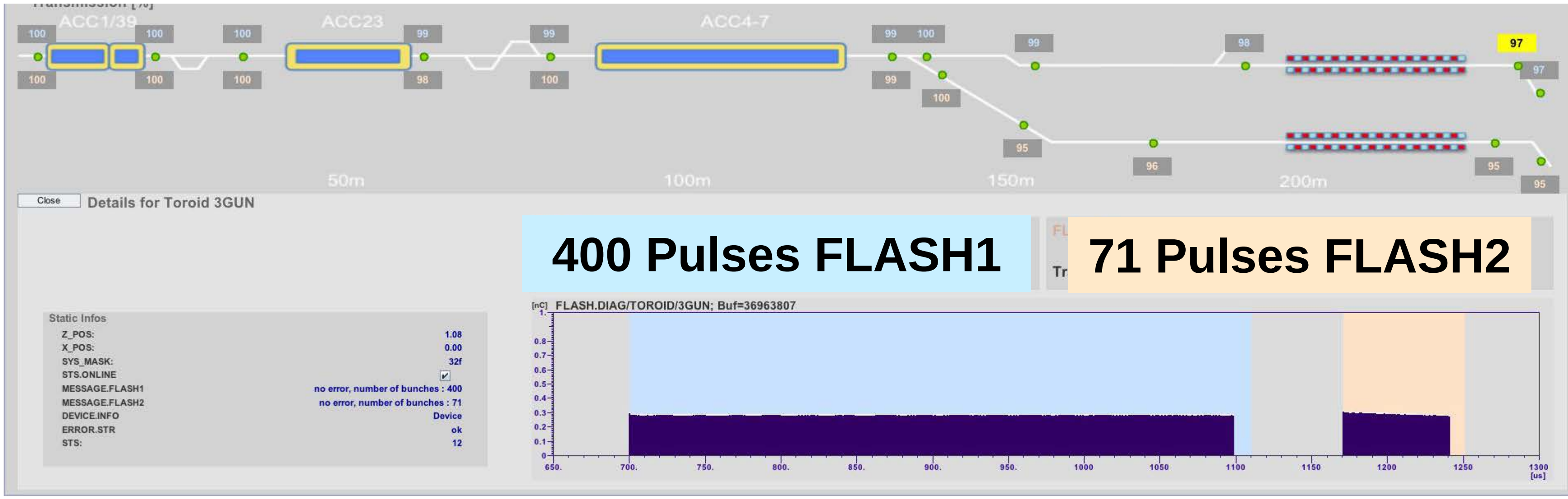
„General“ Conclusions

- Soft x-ray FEL‘s enable movies of electronic structure
- Even „delicate“ systems (such as adsorbates) can be studied with high brillance FEL`s
- Most soft X-ray spectroscopy techniques benefit from high-repetition rate sources
- Fourier-limited bandwidth is important

Developments – example: FLASH



Running two FEL's in parallel



Long Pulse trains at FLASH1 (400 pulses with 100-120 μJ average) and FLASH2 (71 pulses with 70 – 80 μJ average).

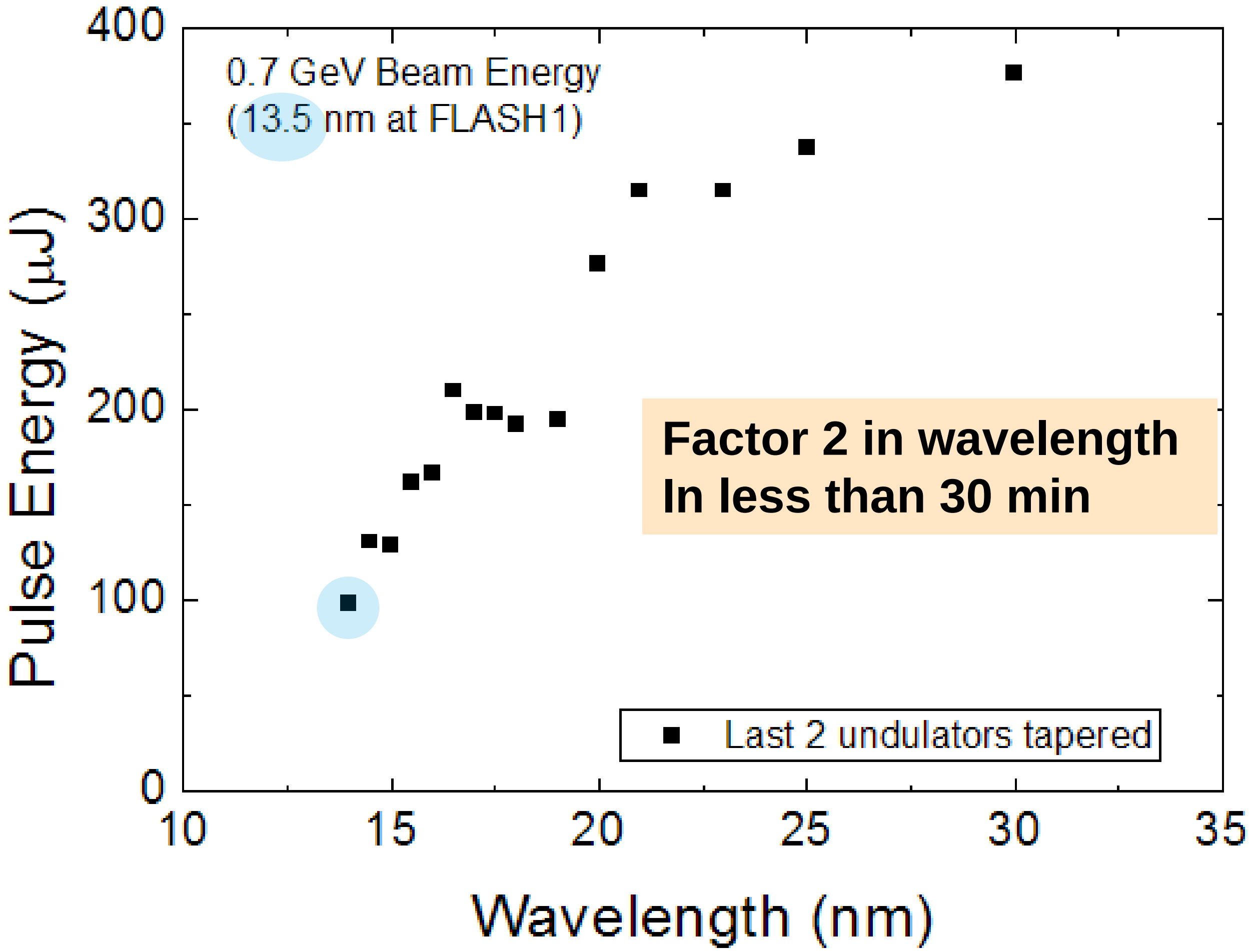
Wavelengths achieved at FLASH2

Bart Faatz

- Below 1.15 GeV (~5 nm at FLASH1): FLASH2 : λ_{FLASH1} to 3x λ_{FLASH1}
- Absolute maximum 90 nm
- Wavelength transported into the hall from 6.5 to 82 nm
- Pulse energies exceeding 50 uJ for wavelengths between 8 and 90 nm

FLASH2 – Fast Tunability

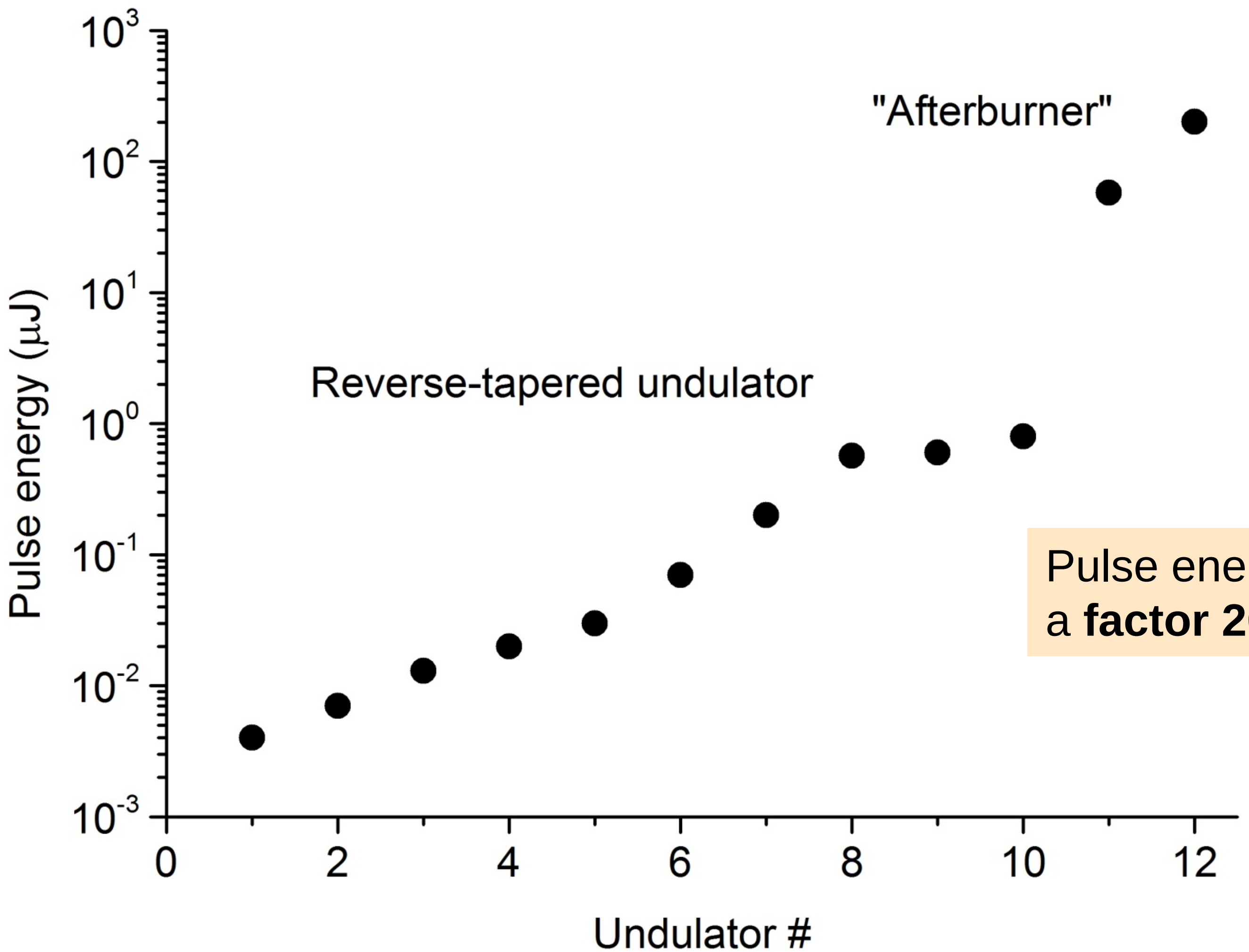
Variable gap undulators



Bart Faatz

Flexible undulators

17 nm, charge 300 pC



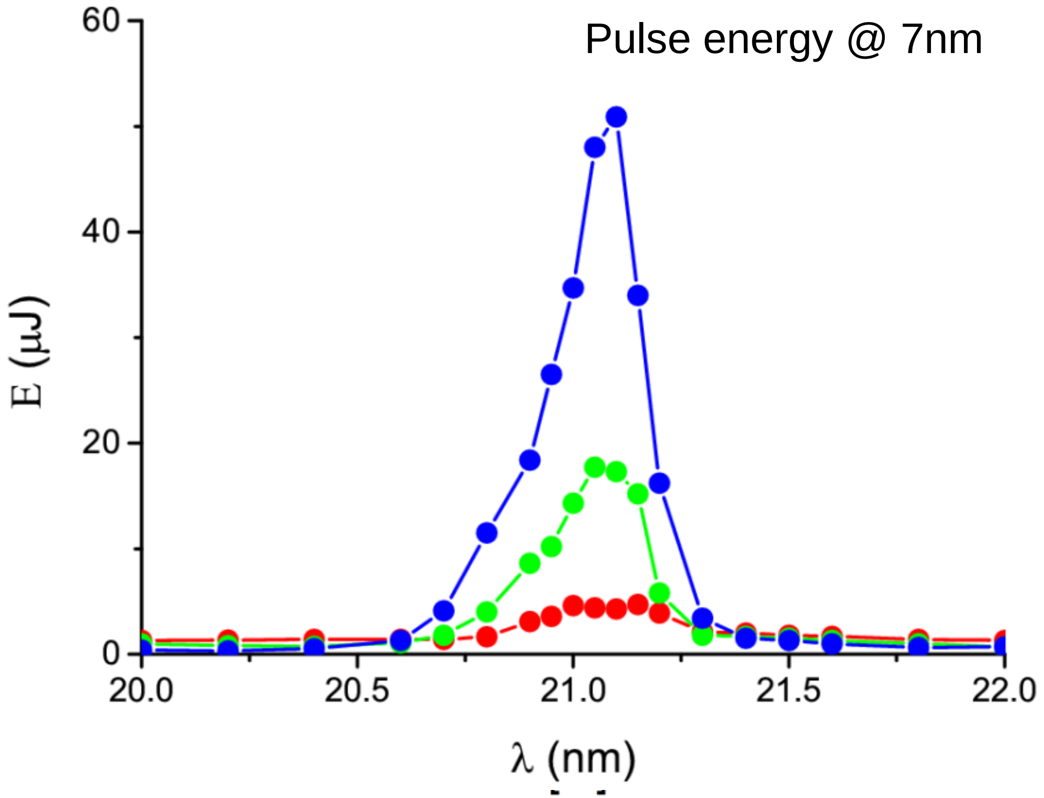
Pulse energy increase by a **factor 200** as predicted

E. A. Schneidmiller, M.V. Yurkov, PRSTAB 16, 110702 (2013)

Flexible undulators

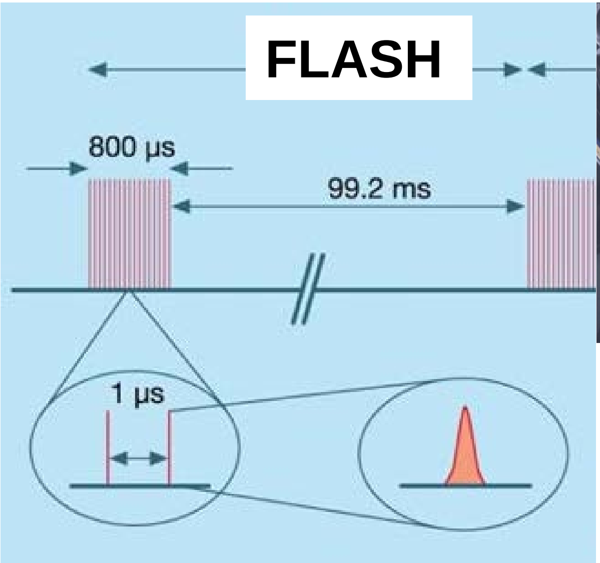
Latest results (May 1st):

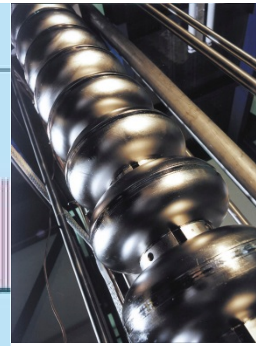
- **1mJ** @ 21nm (945 MeV)
- Transverse coherence measurement (~0.8)
- First demonstration of Harmonic Lasing Self Seeding
  - 3 undulators @ 21nm
  - 7 undulators @ 7nm
  - **50µJ** @ 7nm
  - As compared to 12µJ with 10 undulators
  - Increased temporal coherence predicted

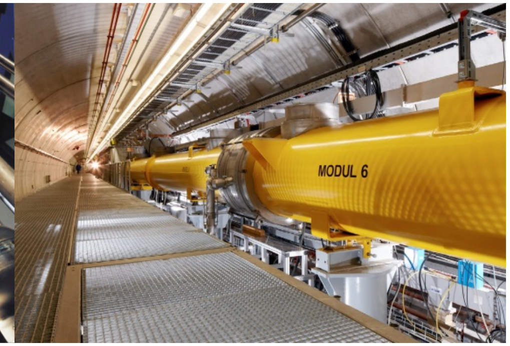


E. A. Schneidmiller and M.V. Yurkov

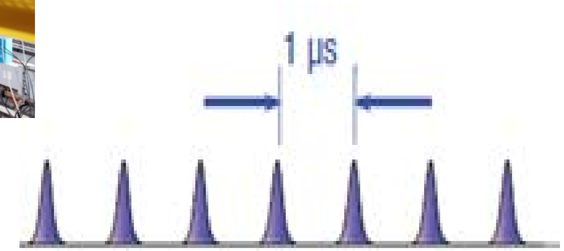
FLASH2020 – A High-Repetition Rate Soft X-Ray Camera







FLASH2020



Gain x 125

FLASH2020

- cw operation up to 1MHz
- fundamental 30-550eV
- multiple FEL lines with up to 100kHz
- external seeding up to 100kHz
- complementary to XFEL/LCLS-II

SCIENCE

- Time-resolved coincidences
- Electronic structure movies
- Single-shot nanoscale imaging
- Light-induced dynamics and control
- Nonlinear spectroscopy

DESY HELMHOLTZ GEMEINSCHAFT