First results from experiments using PP laser, and general fs timing stability between XFEL and optical lasers

Jan-Patrick Schwinkendorf and Motoaki Nakatsutsumi
on behalf of HED and HiBEF team

20th Jan. 2021

European XFEL, Users' meeting – Satellite meeting

Current and future experiments at the HED instrument Reports from the HIBEF UC.

Outline

- PP laser at HED
  - Parameters, activities in 2020, future improvements

- PAM (pulse-arrival monitor – timing tool).
  - Commissioning status and future improvements

- First demonstration experiment using the PP laser at HED instrument
  - Proposal #2716 in Oct. 2020: “Probing of phase transition kinetics at the surface of femtosecond laser-heated warm-dense gold with grazing-incidence x-ray diffraction”
  - One example of possible sciences that can be explored with the PP laser.
    ►Scientific talk on 27th, Wed, at 17h (plenary session).

3(+1) 'high-power' or 'high-rep' optical lasers at the HED instrument

- Pump-Probe (PP) >10^{17} W/cm^2
  - → 0.2–2 mJ, 0.1–4.5 MHz, ~15 fs
  - → 1–40 mJ, 0.1–4.5 MHz, ~1 ps

European XFEL

- ReLaX ≥100 TW
  - → >10^{20} W/cm^2
  - → ≥ 3 J, 30 fs, 5 & 10 Hz

HiBEF USER CONSORTIUM

- DiPOLE-100X
  - → 100J, 2–15ns, 10Hz

HiBEF USER CONSORTIUM

DAC heating laser
(λ = 1030 nm, 100 W, > 10 ns)
Contact: Z. Konopkova and C. Prescher

DFG Deutsche Forschungsgemeinschaft

Pump-Probe (PP) laser ‚designed to work in 4 basic ‚set points'

G. Palmer et al., J. Sync. Rad. **26**, 328 (2019).

	λ	800nm	1030nm
Pulse duration	τ_{FWHM}	15…300fs (nearly transform limited)	<1ps or 400ps (chirped)

Intraburst repetition

Set point	f_{rep} [MHz]	E_{pulse} [mJ] @ 800nm	E_{pulse} [mJ] @ 1030nm
1	4.5	0.05	1
2	1	0.2	4
3	0.2	1	20
4	0.1	2	40

2 mJ / 15 fs/ 5 μmϕ

→ **> 10^17 W.cm^-2**

40 mJ / 1 ps

→ ~ **10^17 W.cm^-2** @ 5 μmϕ

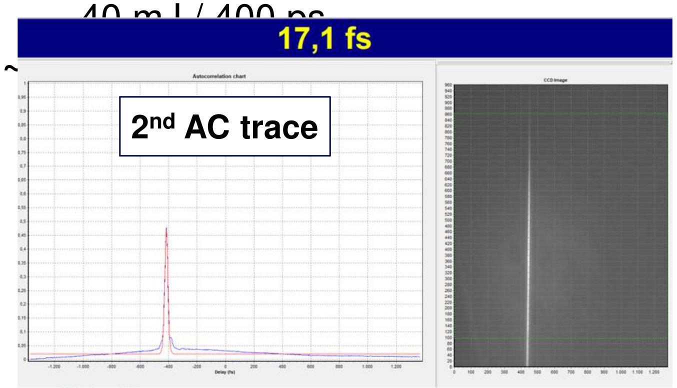
→ **≥ 10^14 W.cm^-2** @ 100 μmϕ

40 mJ / 400 ps

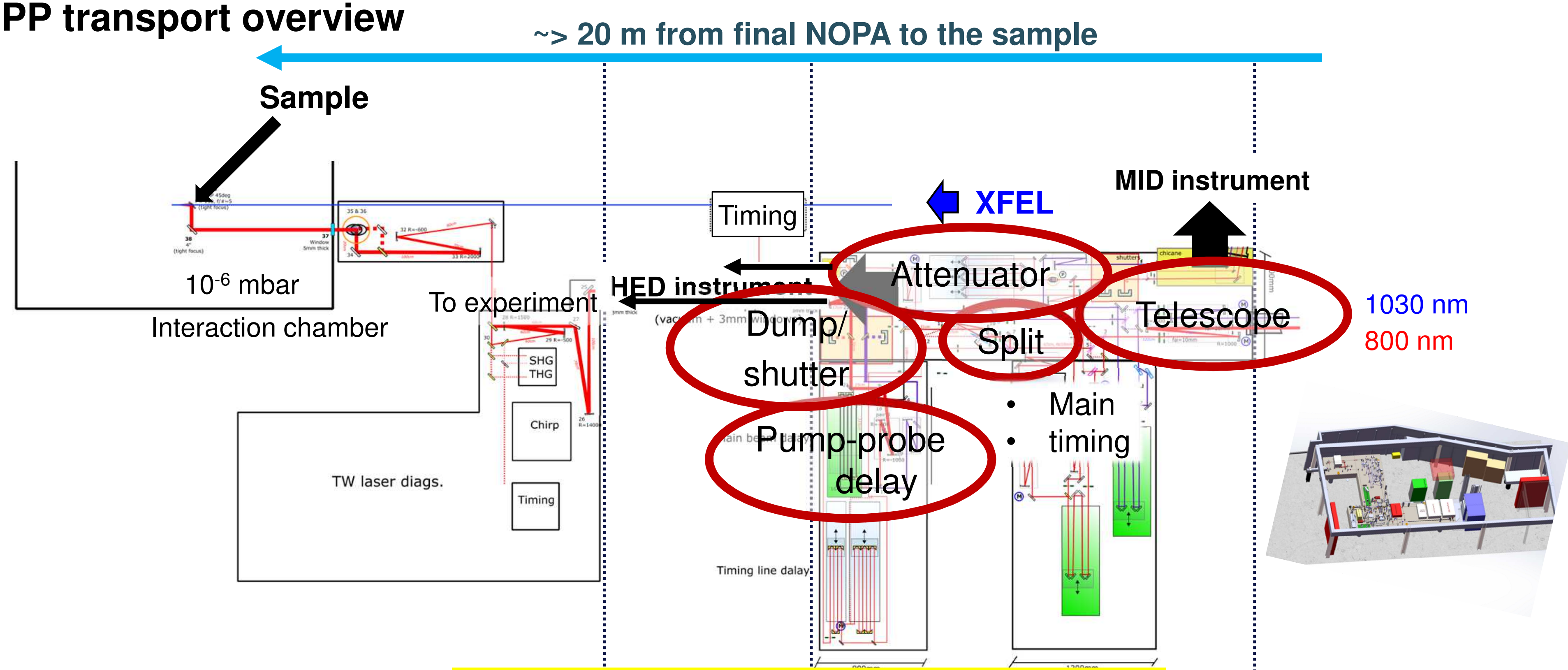
→ ~

Demonstrated in 2020 (λ = 800 nm):

- ~1.6 mJ after the final NOPA, ~1 mJ on sample
- Pulse was able to be compressed to 17 fs FWHM, but chirped to ~100 fs at the sample location – initial negative chirp was insufficient and/or too much dispersion material during the transport.



PP transport overview

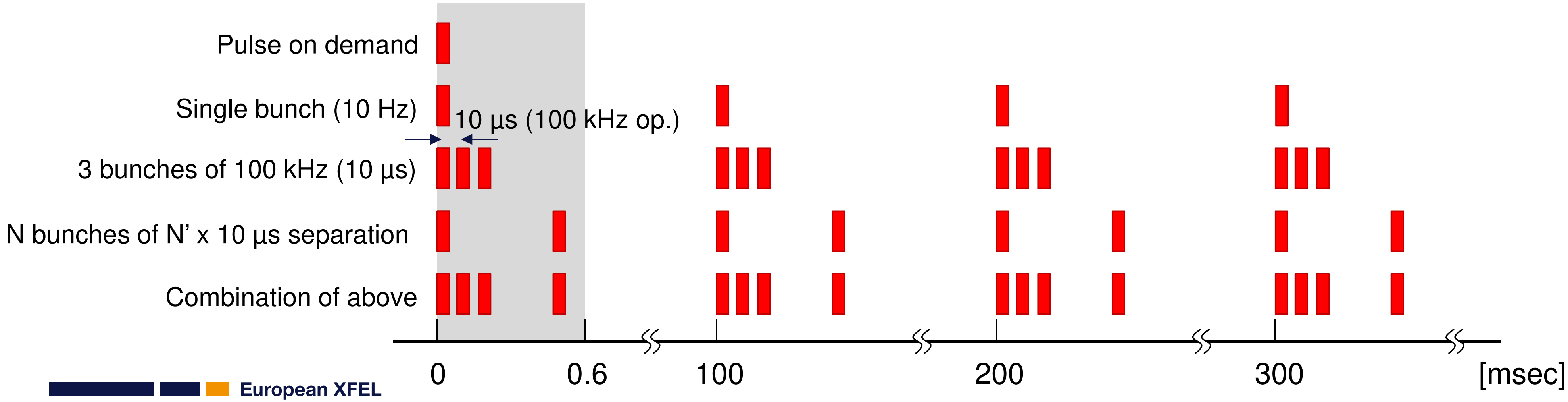


λ = 800 nm: all aligned, and tested, used

HED-EXP	HED-OPT	COM - LAS	SA2-LAS
Experiment	**Timing**	**Manipulation**	**Laser hutch**
		Instru. Sci.	OL group (M. Lederer)

Summary – PP laser parameters and current status

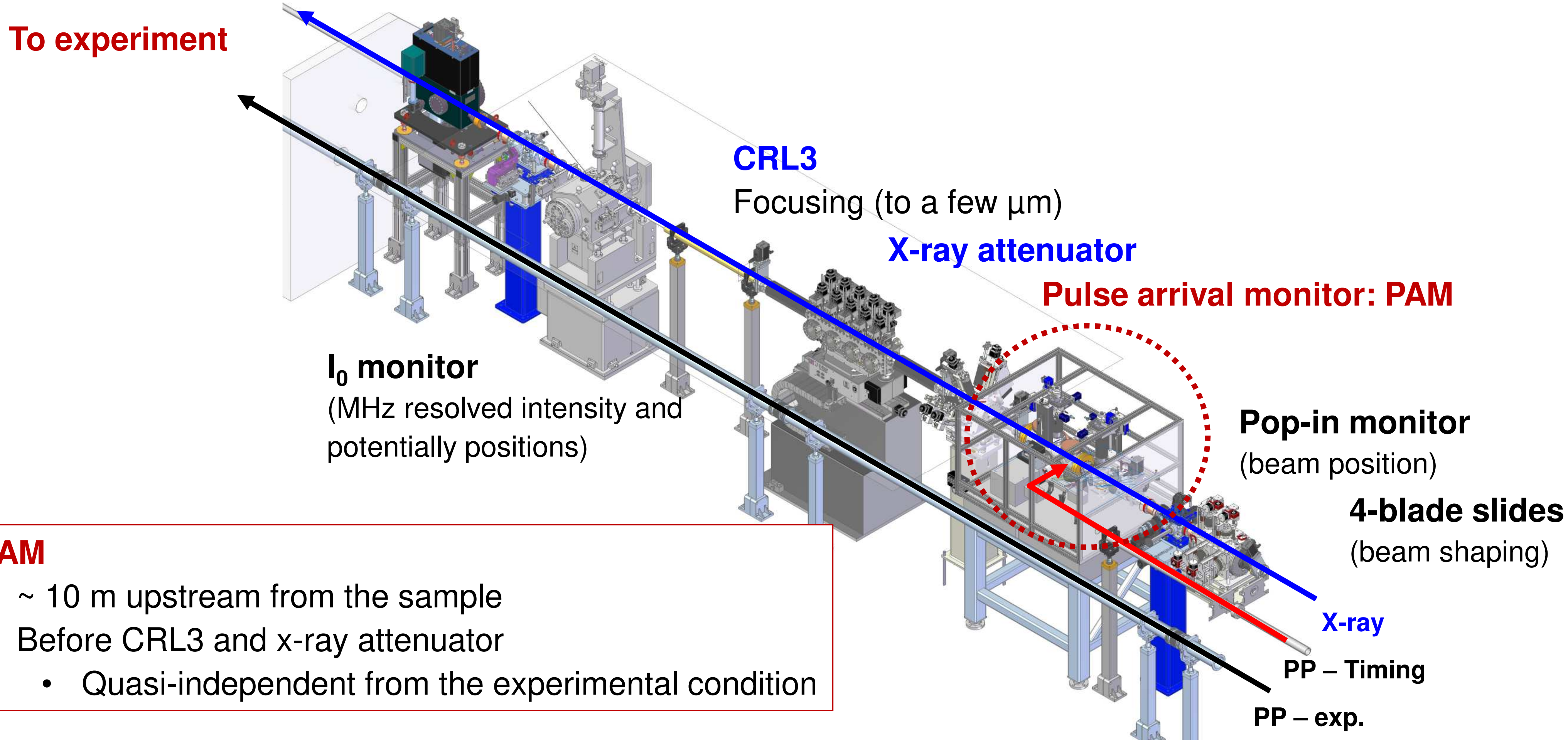
- PP laser 800 nm is ready for x-ray experiments with limited capability
  - ~ 1 mJ, 100 fs, no tight focus (~ 20 µm). SHG (λ = 400 nm) at ~0.1 mJ, shot-on-demand and 100 kHz
    - ► Pulse duration, focusing, SHG (and THG) performance to be improved
    - ► More characterization (e.g. spectral phase) of the laser.
- 1030 nm commissioning is planned in 2021
- Day-to-day beam stability is good (pointing, energy, …). The pulse pattern is easy to change.
- The laser is shared with MID. Coordination and communication appeared extremely important.



Pulse arrival monitor (PAM) – timing tool

Recent commissioning results and future improvements

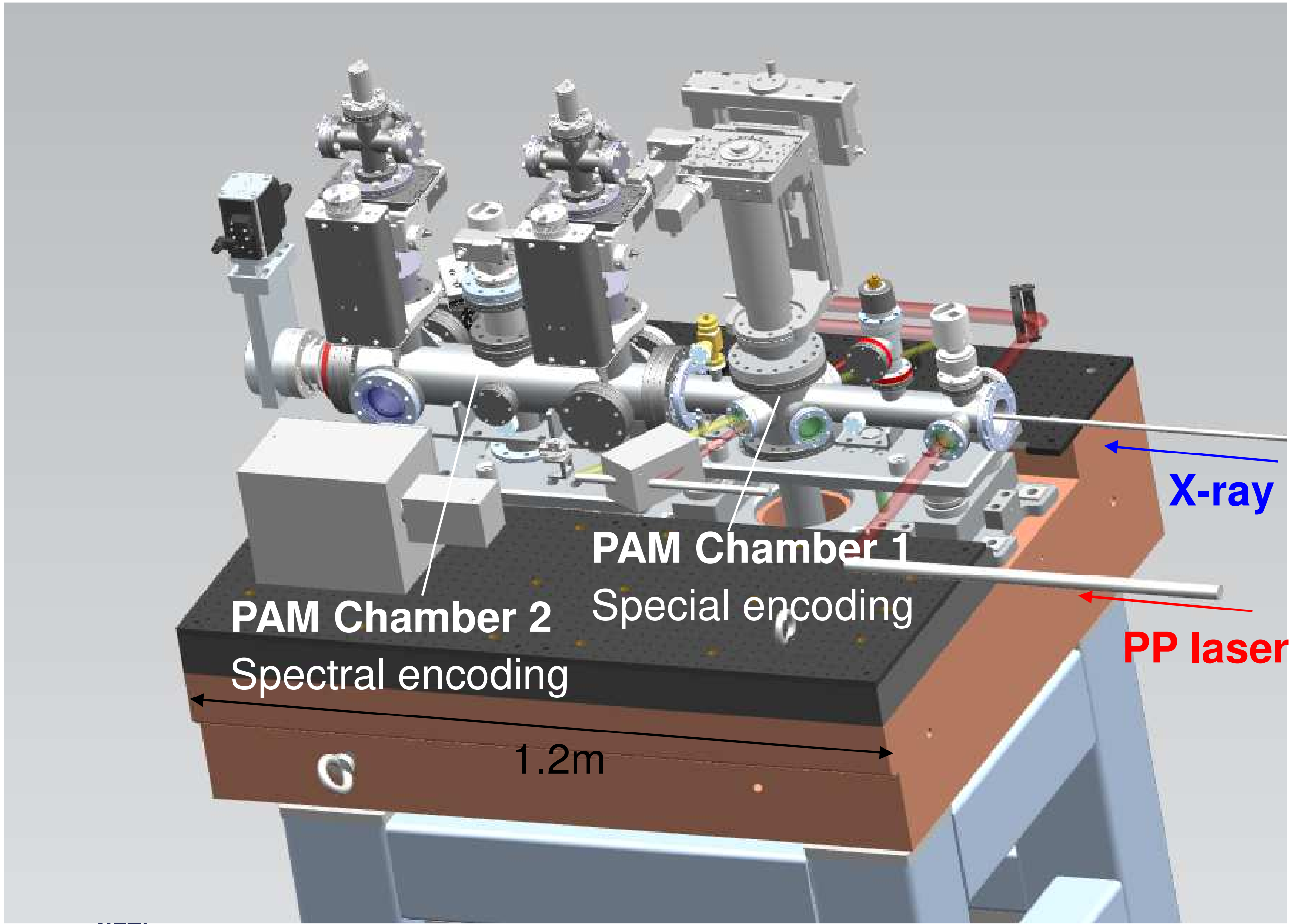
X-ray optics hutch (just before the experimental hutch)



PAM

- ~ 10 m upstream from the sample
- Before CRL3 and x-ray attenuator
  - Quasi-independent from the experimental condition

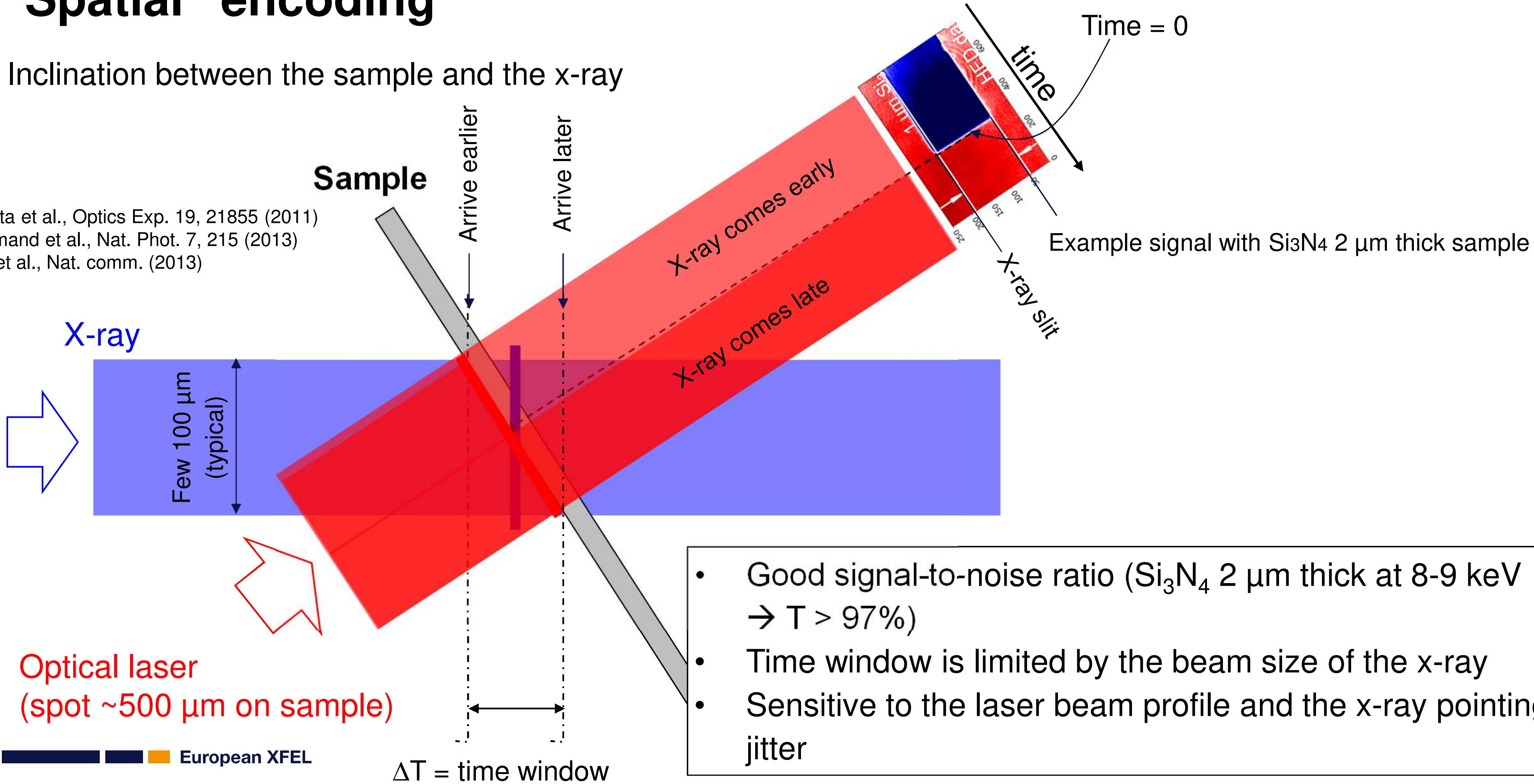
2 sample chambers for simultaneous measurement with 2 methods: spatial and spectral encoding.



"Spatial" encoding

Inclination between the sample and the x-ray

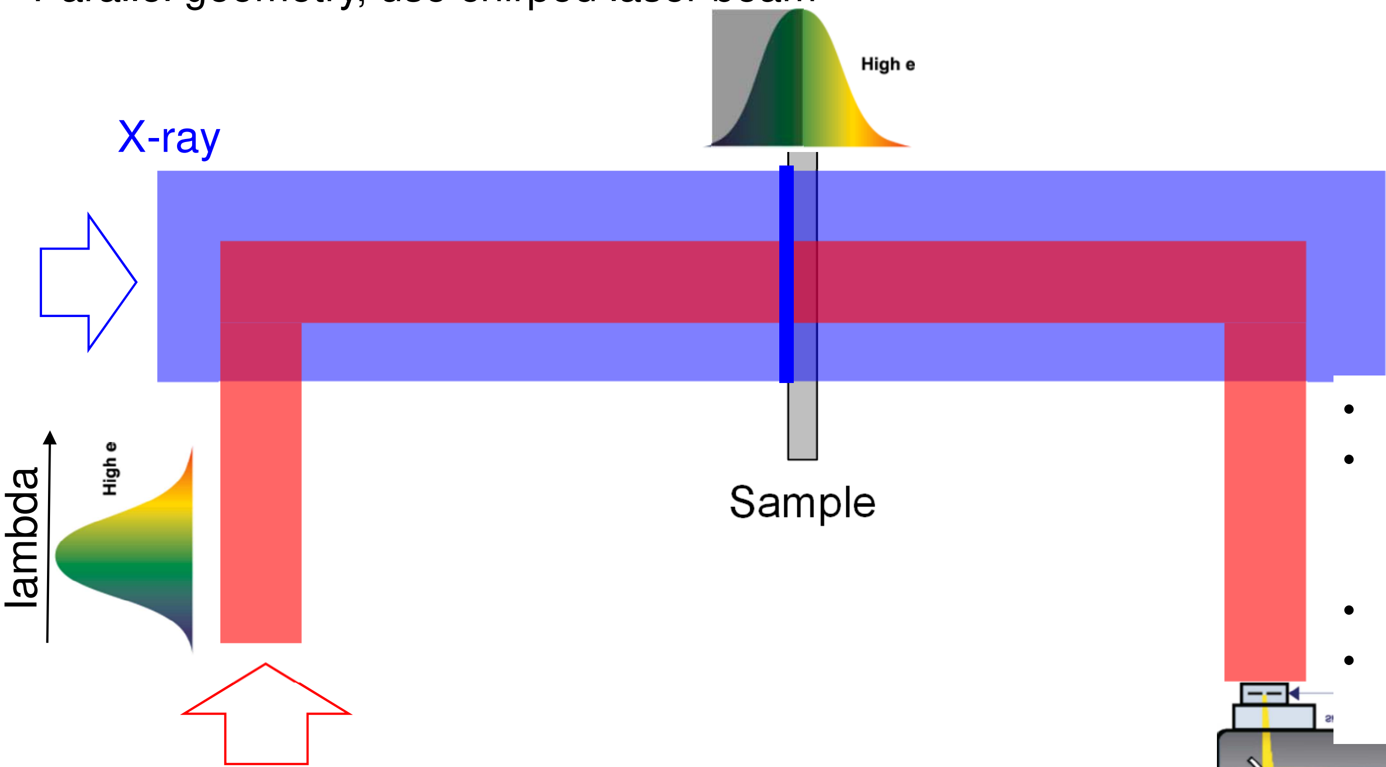
M. Bionta et al., Optics Exp. 19, 21855 (2011)
M. Harmand et al., Nat. Phot. 7, 215 (2013)
Riedel et al., Nat. comm. (2013)
…



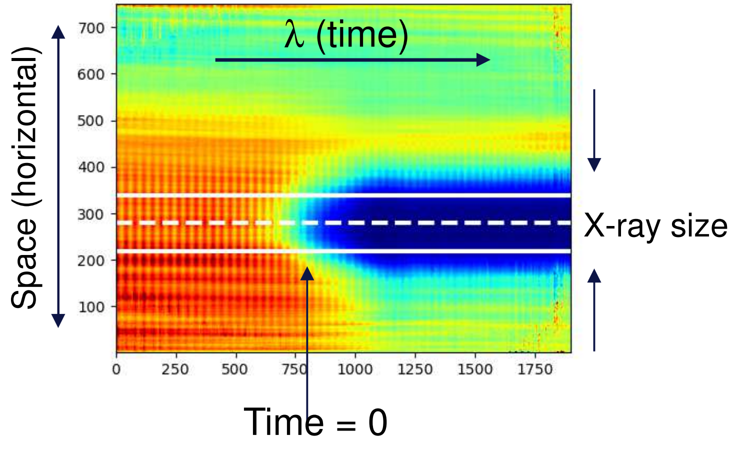
- Good signal-to-noise ratio (Si_3N_4 2 µm thick at 8-9 keV → T > 97%)
- Time window is limited by the beam size of the x-ray
- Sensitive to the laser beam profile and the x-ray pointing jitter

"Spectral" encoding

Parallel geometry, use chirped laser beam

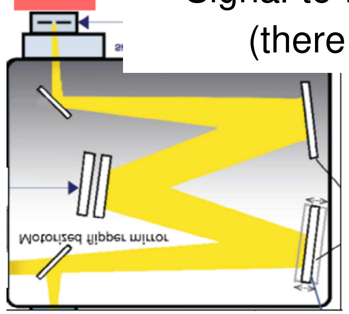


Chirped (> 1 ps) optical laser



- Variable time window (<1 – 30 ps range)
- More robust against
  - laser beam spatial profile
  - x-ray pointing jitter
- It works as a x-ray pointing monitor
- Signal-to-noise ratio is inferior to spatial enc.
  (there is a room for improvement though)

Spectrometer



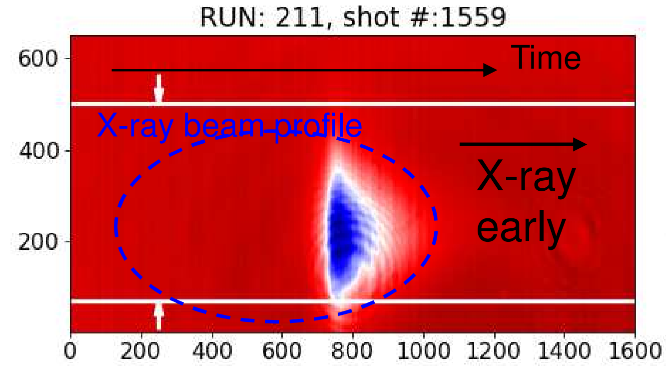
M. Bionta et al., Optics Exp. 19, 21855 (2011)
M. Harmand et al., Nat. Phot. 7, 215 (2013)
Riedel et al., Nat. comm. (2013)
…

Example of a single-shot data with *spatial* encoding
(2µm thick Si_3N_4 sample, X-ray: 9 keV, transmission ~98%)

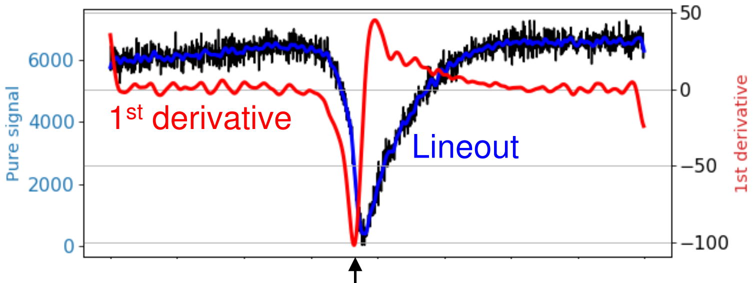
Single shot data

Earlier

Raw data

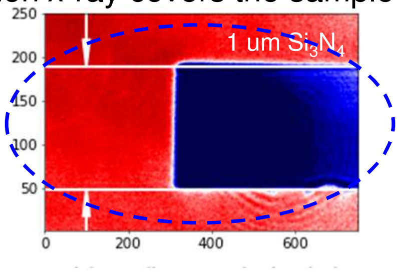


Lineout and Derivative

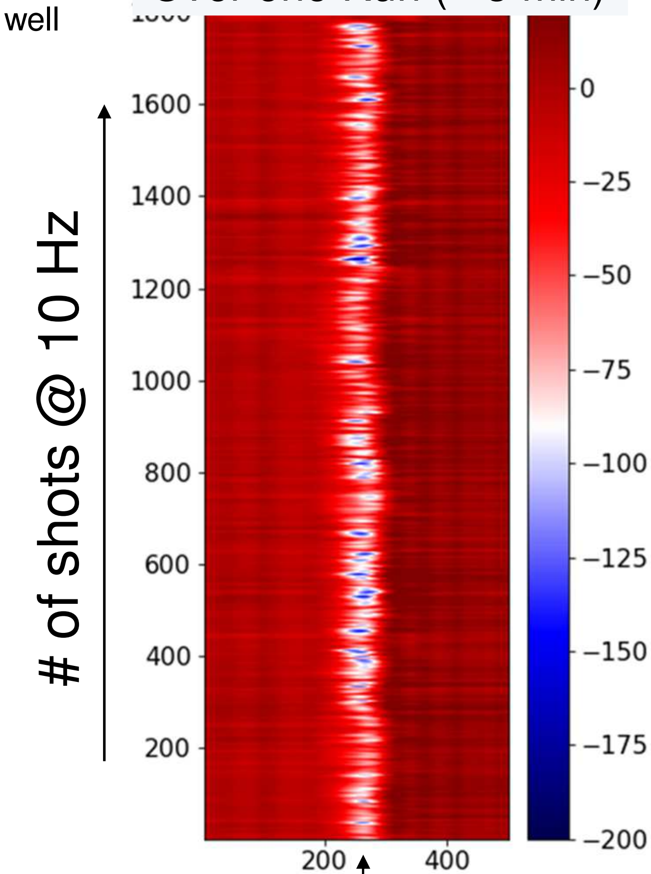


X-ray Arrival time

When x-ray covers the sample well



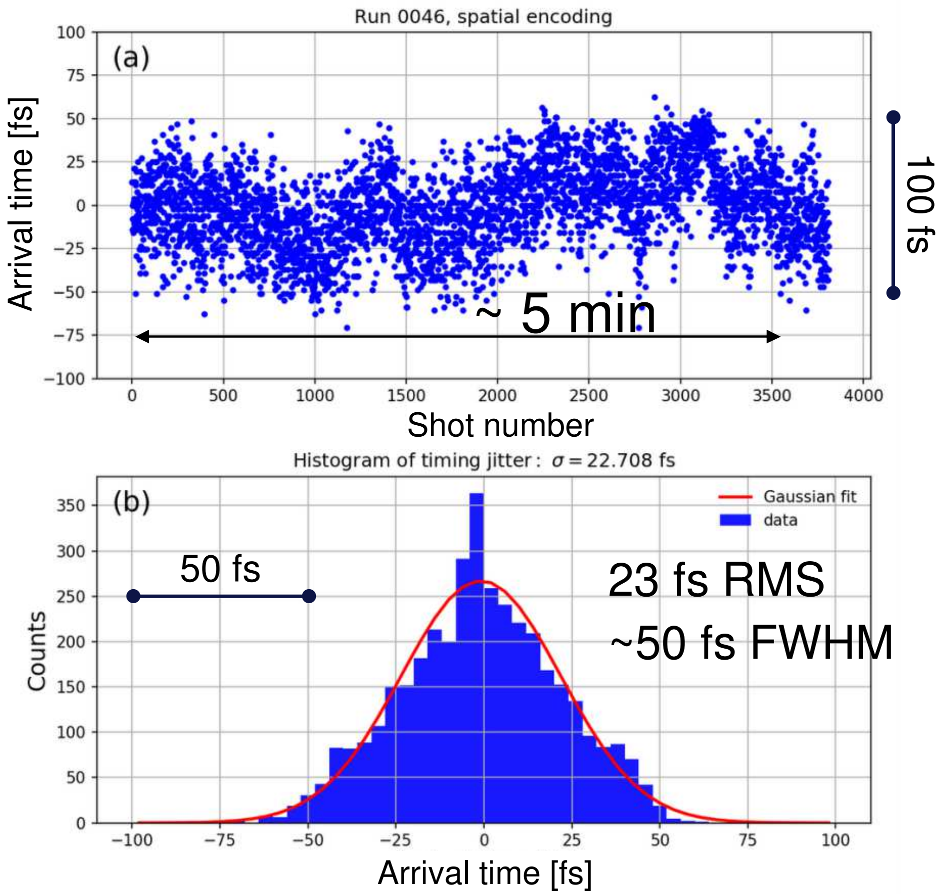
Over one Run (~ 3 min)



X-ray Arrival time

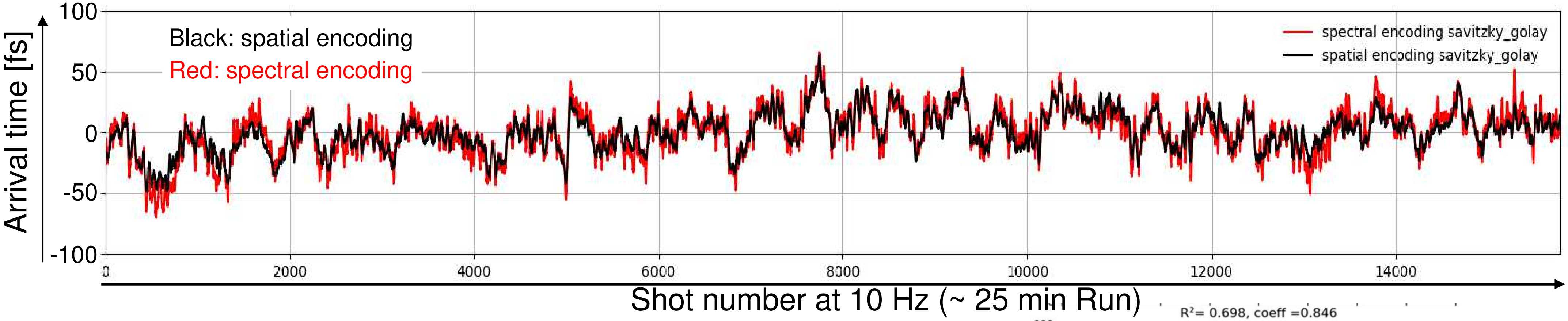
Analysis code was developed by Jia Liu (XPD group, EuXFEL)

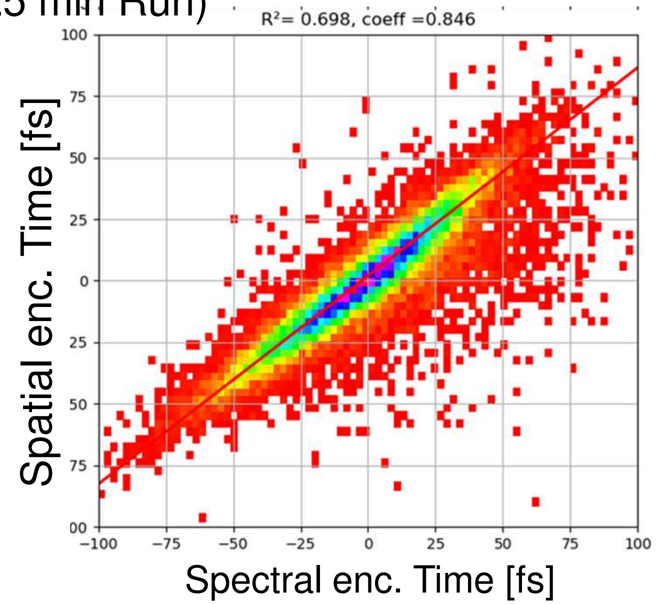
Pulse-to-pulse arrival jitter between x-ray and PP laser is < 30 fs RMS



Very similar RMS timing jitter is observed with ReLaX 100 TW laser too.
→ **Next session.**

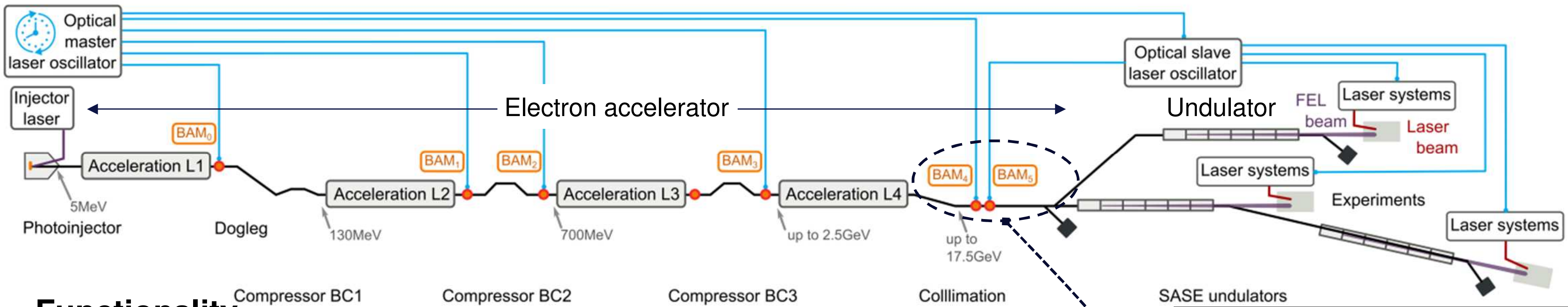
Good correlation between the spatial and the spectral encoding





Analysis code is developed by Jia Liu (XPD group, EuXFEL)

Additional (very promising) online timing tool: BAM (electron bunch arrival time monitors)

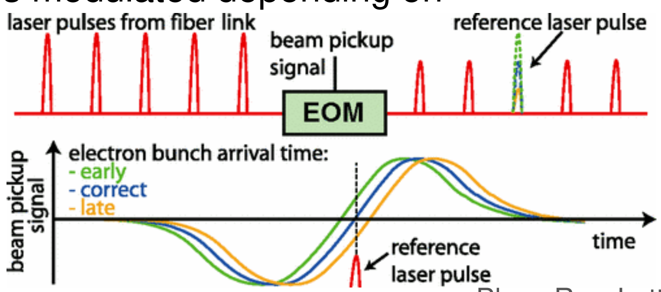


Courtesy: Marie K. Czwalinna(DESY MSK) and Jia Liu (XFEL, XPD)

Functionality

- Measures the time when an electron bunch passes an RF pick-up
  - ❑ RF pulse is applied to an electro-optical modulator (EOM)
  - ❑ amplitude of passing laser pulses from the laser-based synchronization system (lbsync) is modulated depending on the voltage.
- **4.5 MHz compatible**

Recent test at SPB instrument shows a good correlation between PAM and BAM (not conclusive, need further investigation and analysis)



BAMs 1932M.TL & 1932S.TL

- **Last monitor in front of undulators**
- Measuring all SASE1,2,3 bunches
- **Resolution: ~3 fs**
- DOOCS-Karabo bridge, **data available to user**

Phys. Rev. Lett. **104**, 144801 (2010)
MOPIK072, Proceedings of IPAC2017, Copenhagen, Denmark, 2017
TUPHA125, Proceedings of ICALEPCS2017, Barcelona, Spain, 2017

PAM also delivers certain information about the x-ray pulse duration

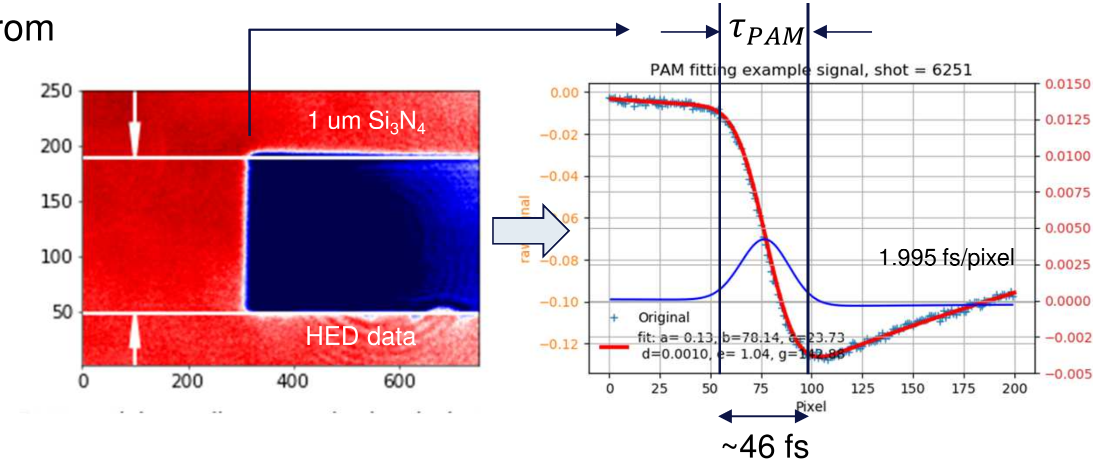
This slide is provided by Jia Liu (XPD group, EuXFEL)

- As we observe the transmission as a function of time with the resolution of a few fs, we can obtain further information from the falling time of the transmission, τ_{PAM}, which is not instantaneous but is the convolution of
  - τ_{FEL}: x-ray pulse duration
  - $\tau_{sample}(h\nu)$: sample response time (e- cascading)
  - τ_{PP}: laser pulse duration

$$\tau_{PAM} = \sqrt{\tau_{FEL}^2 + \tau_{sample}^2 + \tau_{pp}^2}$$

R. Riedel et al., Nat. Comm. **4**, 1731 (2013)

Measured falling time



- If we use $\tau_{PAM} = 46$ fs, $\tau_{sample}(h\nu\sim 9$ keV$) = 28$ fs*, $\tau_{PP} = 17$ fs, the x-ray pulse duration τ_{FEL} can be given by

$$\boldsymbol{\tau_{FEL}} = \sqrt{\tau_{PAM}^2 - \tau_{sample}^2 - \tau_{pp}^2} = \sqrt{46^2 - 28^2 - 17^2} \leq \mathbf{32\ fs}$$

* Estimating the sample response time is usually not an easy task and can contain errors. For Si3N4, there is a calculation available in N. Medvedev Appl. Phys. B **118**, 417 (2015).

Summary PAM

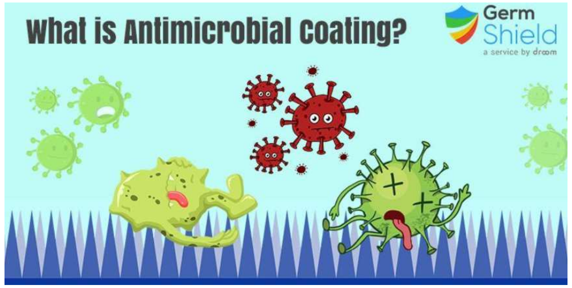
- Online pulse arrival monitor (PAM) is working. Signal quality is good enough as an online timing tool
  - $h\nu$ = 8 – 9 keV with Si_3N_4 2 µm thick sample ➔ T > 98%.
  - For higher photon energy operation, thicker samples are available (4 and 6 µm Si_3N_4 and 10 and 20 µm YAG) but untested.
- BAM (electron bunch arrival monitor) may become a useful complementary timing monitor
  - fully non-invasive,
  - max intraburst (4.5 MHz) compatible
- Limitations and near-future tasks
  - Currently only for 10 Hz. Also, no plan to improve it to MHz.
  - Huge issues on optical cameras' performance. New camera is bought. To be replaced in 2021.
  - Analysis and display tools should be improved to provide information to users immediately.
    ► Currently relying on offline analysis

One example experiment using the PP laser combined with surface-sensitive x-ray techniques

Nanoscale surface morphology plays decisive roles in versatile fields

Damaging bacteria

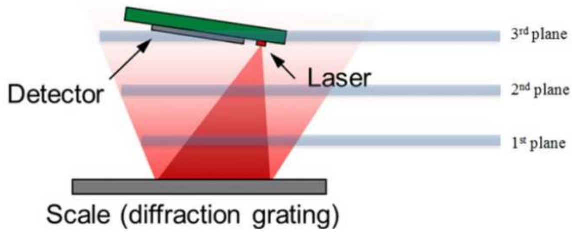
by producing reactive oxygen species



From: https://blog.droom.in/antimicrobial-coating

Optical encoder:

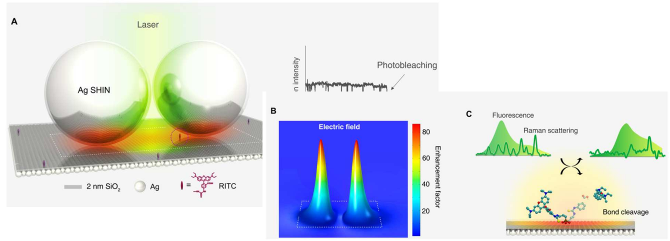
Optimizing reflection from the tape by nanostructuring



From: https://www.celeramotion.com/microe/optical-encoders/

Local field enhancement

Plasmon enhanced spectroscopy



Sci. Adv. 6, eaba6012 (2020)

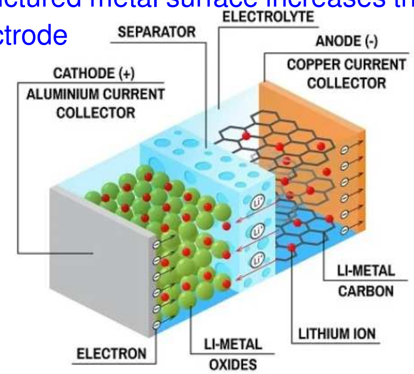
Dental implant, screws:

Structured surface bond much better with natural bones

From: https://www.bickfordandshirley.com/blog/expect-process-dental-implants-in-hiram/

Battery

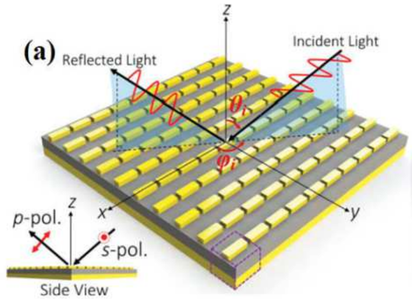
Structured metal surface increases the capacity of the electrode



From: sivVector/Shutterstock.com

Advanced optics

Subwavelength surface structure acts as optical anisotropic media



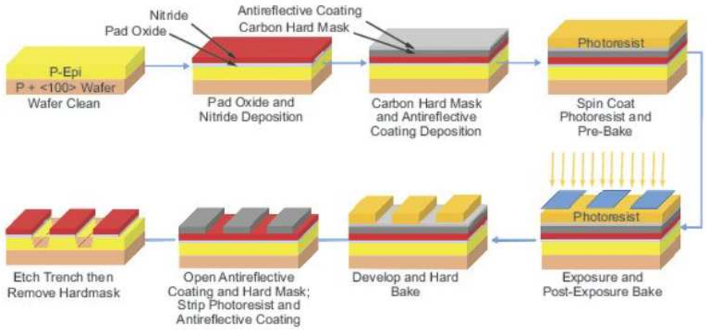
Shugi Chen et al., DOI: 10.5772/66036

Most of these application examples are from: Laser4surf
EU Horizon 2020
Research and Innovation

Surface nano-structuring by intense, femtosecond laser. It involves different physics on different time scales in intricate ways → needs appropriate tools to study dynamics

How to create a surface nanostructure in a controlled way?

Lithography → several step process



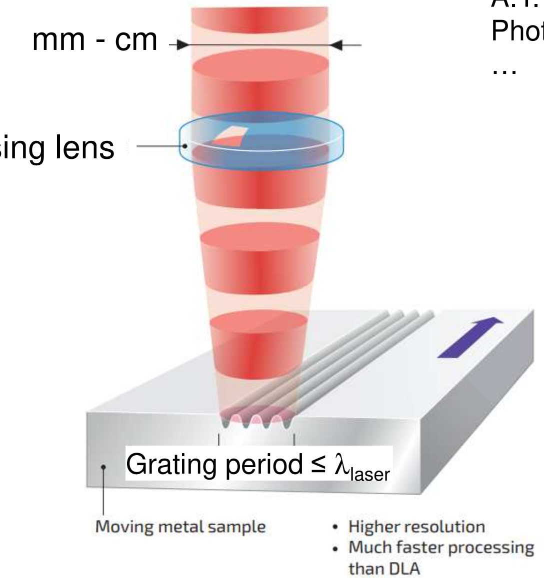
From: https://www.newport.com/n/photolithography-overview

- ~fs 1. Laser absorption into solids
- fs – ps 2. Phase transition, plasma creation
- > ps 3. Thermalization with lattice and ions
- ps – ns 4. Surface ablation, expansion
- ns - µs 5. Re-solidification

Surface 10 – 100 nm scale

Laser ablation, Laser-Induced Periodic Surface nano-Structure (LIPSS)

→ Single-step process



https://www.laser4surf.eu/wp-content/uploads/2019/01/Laser-comparing.pdf

J. Bonse and S. Gräf, Las. Phot. Rev. **14**, 2000215 (2020).
A.Y. Vorobyev and C. Guo, Laser Photon. Rev., **7**, 1, (2012)
…

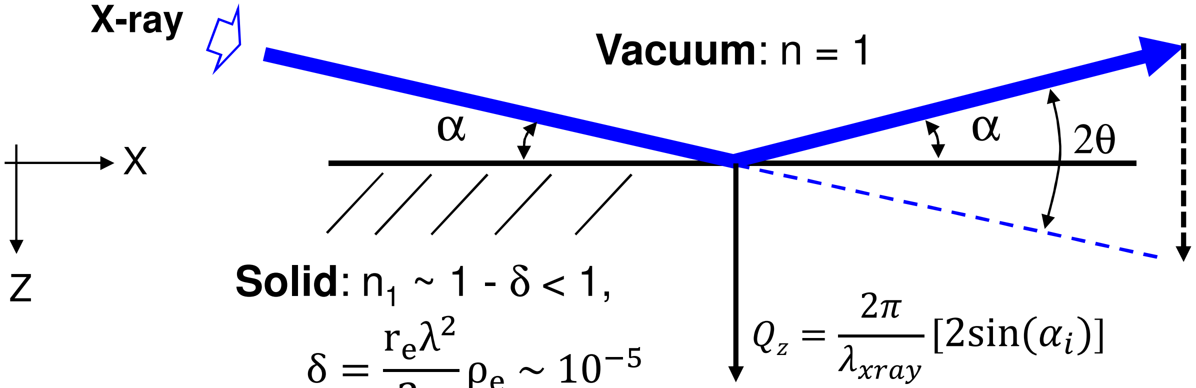
Laser4surf

EU Horizon 2020
Research and Innovation

- Optics
- Medicine
- Tribology
- Fluidics…

➔ Need for *in situ* visualization of surface and subsurface with relevant (Å to sub-µm) resolution with fs – ps precision

X-ray can be surface sensitive when going to the grazing-incidence (≤ 1 deg.)



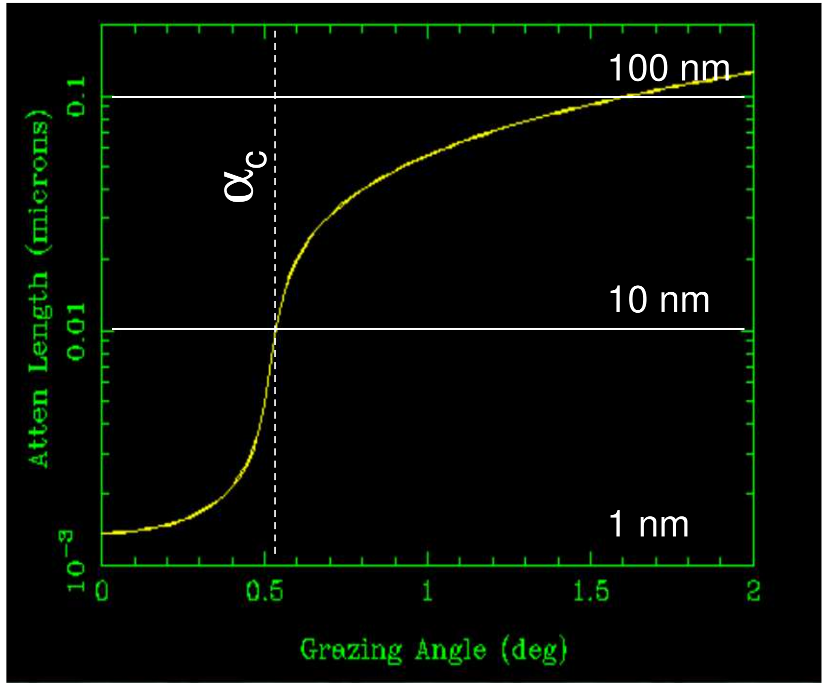
Solid: $n_1 \sim 1 - \delta < 1$,

$$\delta = \frac{r_e \lambda^2}{2\pi} \rho_e \sim 10^{-5}$$

for x-ray

$$Q_z = \frac{2\pi}{\lambda_{xray}} [2\sin(\alpha_i)]$$

Penetration depth for Ta at $h\nu = 8$ keV



http://www.cxro.lbl.gov/

Critical angle (α_c) for total external reflection
for Ta (Z = 73), hν = 8 keV,

$$\alpha_c = \sqrt{4\pi\rho r_0}/k = \mathbf{0.52°}$$

J. Als-Nielsen and D. McMorrow
Elements of modern x-ray physics
John Wiley & Sons, Ltd. (2011)

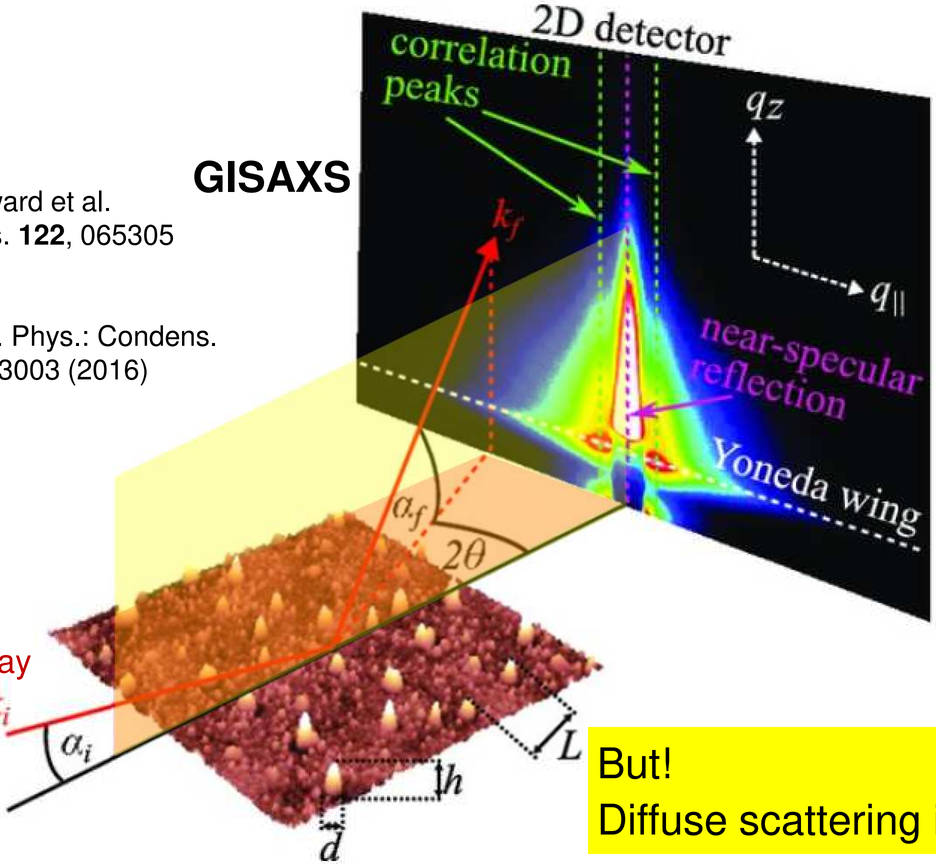
X-ray penetration depth is tunable from a few nanometer (nm) to few 100 nm by changing the grazing-incident angle

➔ **surface sensitive**

Grazing-incidence small-angle/wide-angle x-ray scattering (GISAXS/GIWAXS) to obtain longitudinal and horizontal density profile as well as atomic structure

J. M. Woodward et al. J. Appl. Phys. **122**, 065305 (2017)

S. V. Roth, J. Phys.: Condens. Matter **28** 403003 (2016)



But!
Diffuse scattering is weak.

GISAXS, in-plane scattering

$$Q_z = \frac{2\pi}{\lambda}(\sin\alpha_i + \sin\alpha_f) \sim 0.01 \text{ – few nm}^{-1}$$

→ : Information about longitudinal density profile, RMS roughness

GISAXS, out-of-plane scattering

$$Q_{||} = \frac{2\pi}{\lambda}(\cos\alpha_f \sin 2\theta) \sim 0.01 \text{ – few nm}^{-1}$$

→ Correlation lengths, mutual distance and size of nanostructures

GIWAXS (GID) – large 2θ

$Q \gg 1 \text{ nm}^{-1}$

→ Atomic structure from the x-ray penetration depth ~ 10 – 100 nm

First successful experiment of **single-shot** GIXS: L. Randolph et al., arXiv: 2012.15076 – Talk on 27th

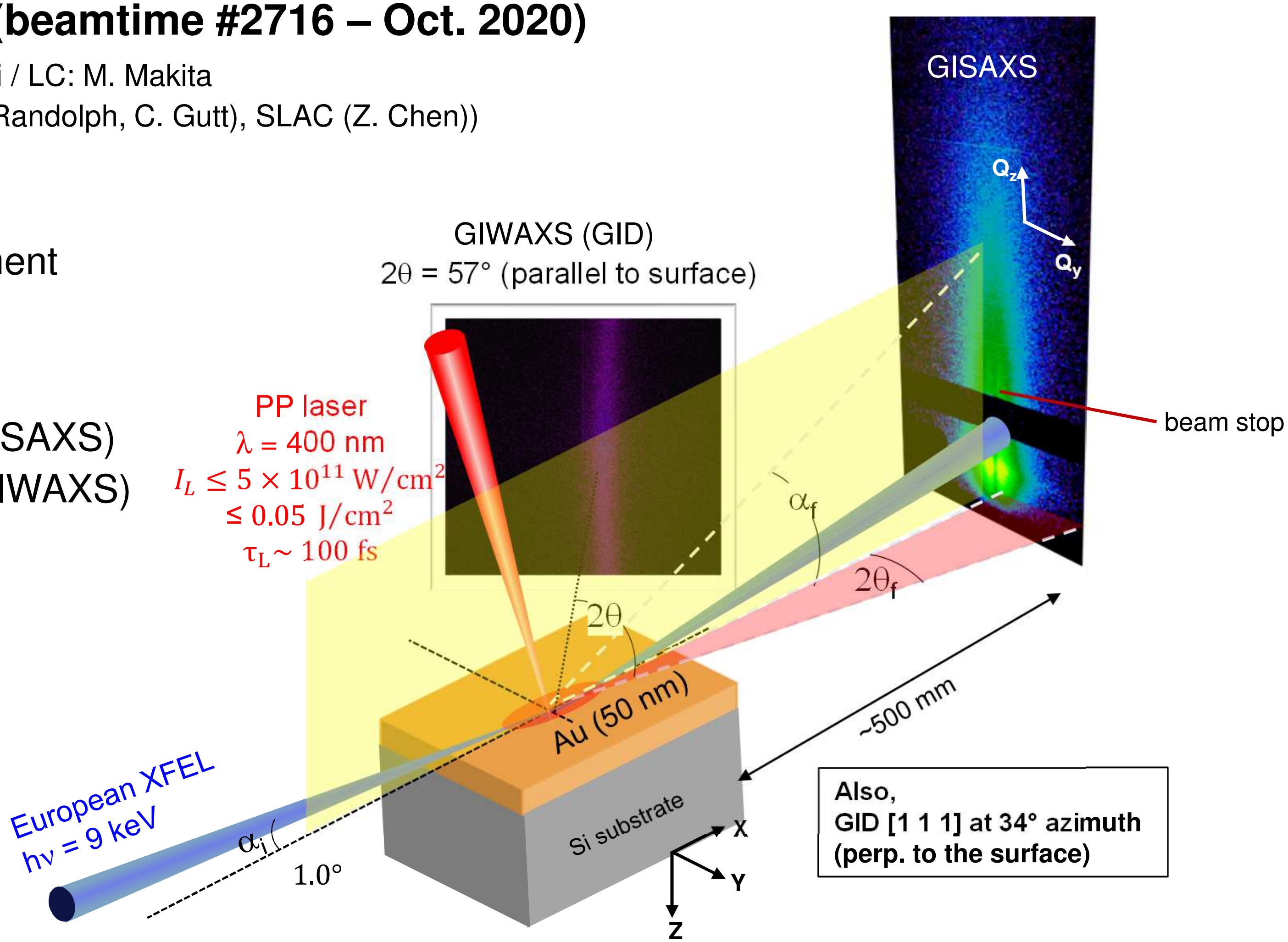
Simultaneous detection of atomic and nanoscale dynamics at the surface of femtosecond laser-heated warm-dense gold (beamtime #2716 – Oct. 2020)

(PI: J-P. Schwinkendorf / MP: Nakatsutsumi / LC: M. Makita
with main contributions from U. Siegen (L. Randolph, C. Gutt), SLAC (Z. Chen))

- First PP laser experiment
- First grazing-incidence x-ray experiment

- Surface nano-structure dynamics (GISAXS)
- Solid-liquid phase transition (GID / GIWAXS)

M. Z. Mo et al., Science **360**, 1451 (2018);
Z. Chen et al., PRL **121**, 075002 (2018);
V. Recoules et al., PRL **96**, 055503 (2006)



Summary: first experiment using the PP laser at HED

- Surface diffraction and scattering from only 50 nm thick gold with a single XFEL pulse.
  - Change in texture, rearrangement of surface morphology.

- One can pursue nice science with the PP laser at HED.
  - Phase transition dynamics in *isochorically* heated **warm-dense-matter** with optical laser.
    - ► Electron ballistic range = 50 – 100 nm ~ x-ray probing depth.
  - Surface **ablation** dynamics
    - ► *In situ* surface morphology (roughness or specific nanostructures) *during* and after re-solidification
  - Exploit high repetition rate of the PP laser (100 kHz - 2 mJ, 4.5 MHz - 50 µJ).
    - ► Change in the surface morphology after each laser irradiation (potentially with MHz detectors - HiBEF).
    - ► Change in laser absorption (and subsequent phase transition) due to the surface structure.
    - ► Gives a new insight into *e.g.*, LIPSS dynamics.

Conclusion & near-future tasks

- **PP laser 800 nm is ready for x-ray experiments with limited capability**
  - ~ 1 mJ, 100 fs, no tight focus (10 – 20 µm). 400 nm at ~0.1 mJ, shot-on-demand and 100 kHz
  - Pulse duration, focusing, SHG (and THG) performance to be improved
  - Beam transport and better characterization (e.g. spectral phase) should be done

- **1030 nm commissioning is planned in 2021**

- **Online pulse arrival monitor is working with good quality at 8 – 9 keV**
  - Si_3N_4 2 µm thick sample with T > 98%. Thicker samples for higher x-ray hν available (4 and 6 µm Si_3N_4 and 10 and 20 µm YAG) but yet untested.
  - Issue in optical cameras' performance. New camera is bought. To be implemented.
  - Analysis tools (now rely on *offline* analysis) should be improved

- **Additional**
  - **FDI** reflectometry, polarimetry is setup. Still needs tests.
  - **White light generation** of PP to produce 600-700 nm wavelength as a probe
  - **High-order harmonic spectrometer** for 5 – 80 nm wavelength will be bought and implemented

Acknowledgements

EuXFEL, HiBEF

HED/HiBEF: J-P. Schwinkendorf (PI of #2716), H. Höppner, M. Makita (LC of #2716), E. Brambrink, T. Toncian, M. Banjafar, L. Wollenweber, Th. Preston, A. Pelka, S. Göde, S. Di dio Cafiso, D. Müller and U. Zastrau

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And, ***all other* HED/HiBEF colleagues and EuXFEL support groups**

U. Siegen L. Randolph and C. Gutt (X-ray analysis, experiment)

HZDR Th. Kluge (plasma simulation, experiment)

U. Mainz G. Jakob, M. Kläui (Sample preparation)

SLAC Z. Chen, S. Glenzer (Warm dense gold, FDI, discussions, experimental planning)

CEA, France V. Recoules, L. Soulard (Warm dene gold, MD/DFT simulations)

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TU Darmstadt C. Rödel (inspiring discussions)

GIST, Korea Byong-ick Cho (discussions, experimental planning)