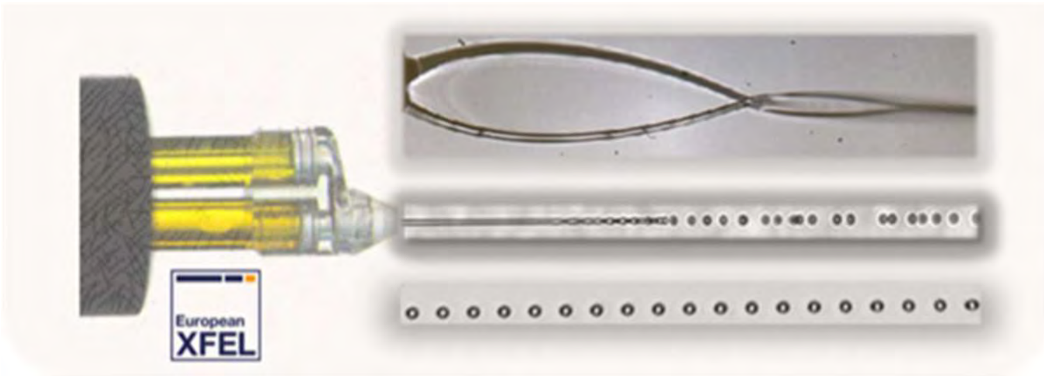
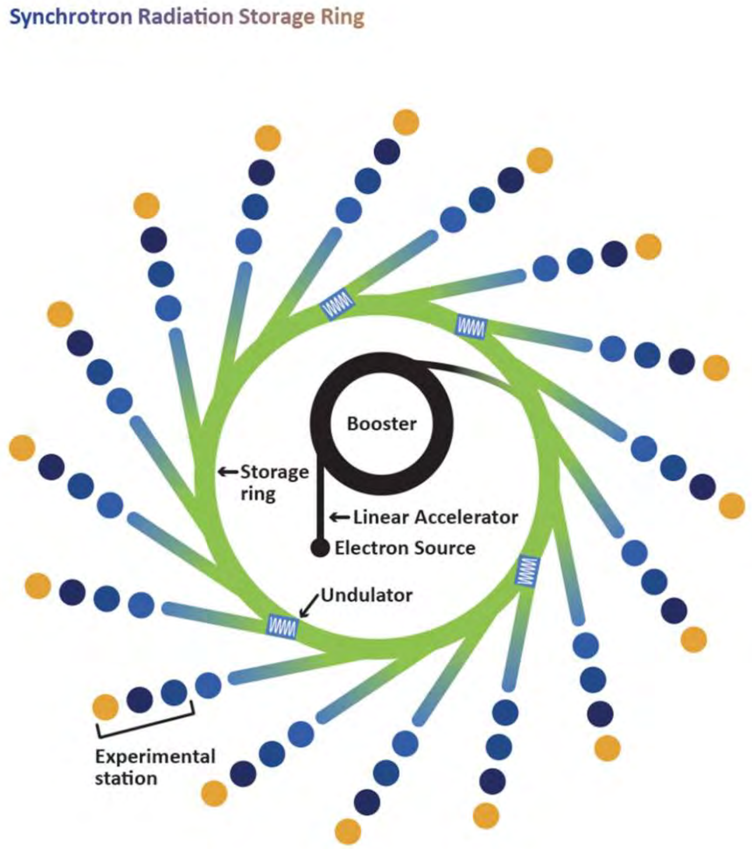
Sample Environments for X-ray Free-Electron Lasers

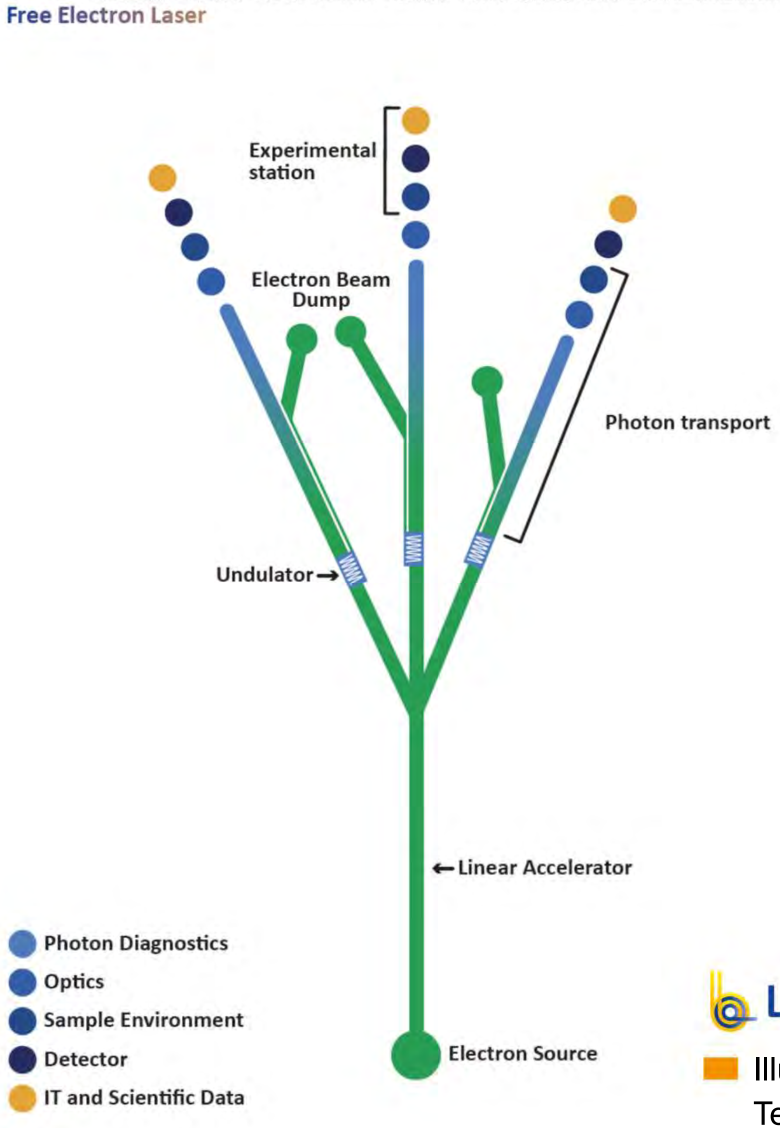
Opportunities and Challenges
(with a focus on the European XFEL)



Joachim Schulz
Sample Environment and Characterization

Synchrotron Radiation Source vs. X-ray Free-Electron Laser (XFEL)





Illustrations from LEAPS Technology Roadmap 2025

Peak Brilliance of Free-Electron Lasers

$$B = \frac{\dot{N}_{ph}}{4\pi\sigma_x\sigma_y\sigma_{x'}\sigma_{y'}\frac{d\omega}{\omega}}$$

Photons per second

Beam size

Beam divergence

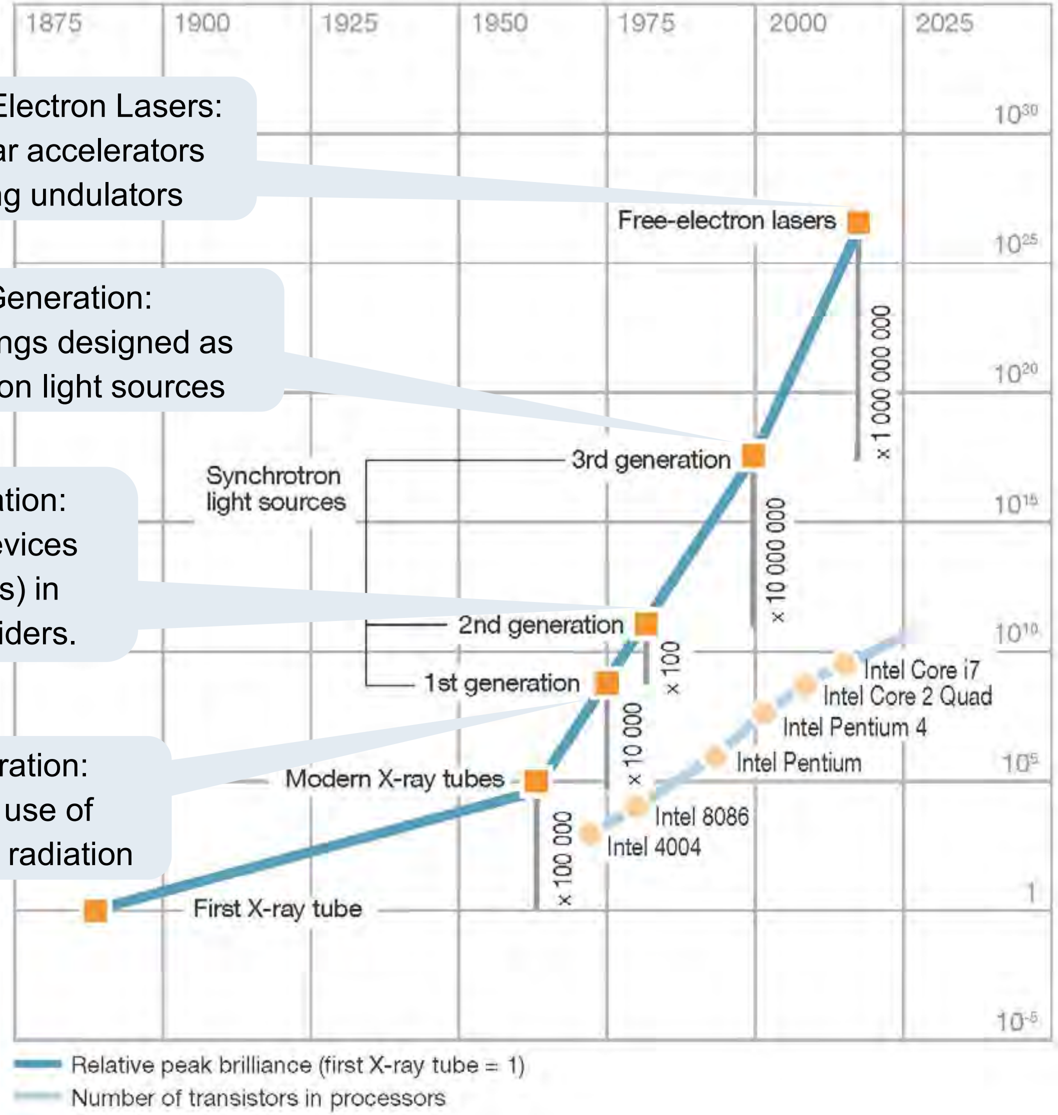
Bandwidth

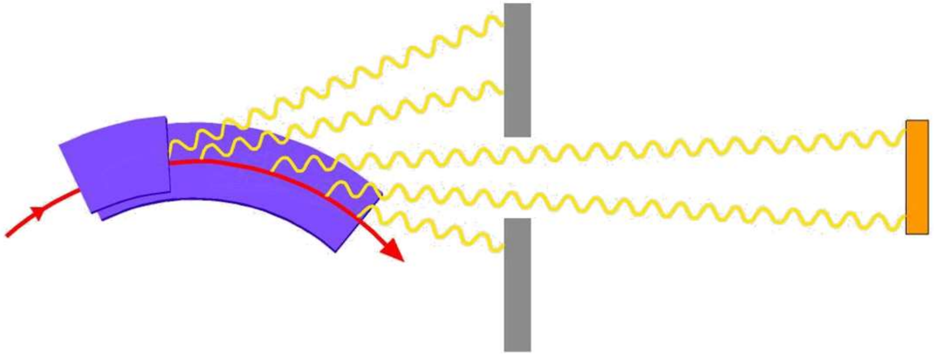
Free-Electron Lasers:
Linear accelerators
Long undulators

3rd Generation:
Storage rings designed as synchrotron light sources

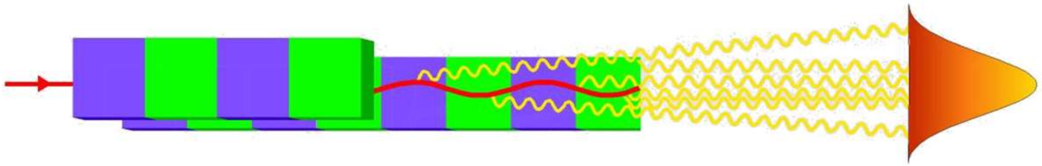
2nd Generation:
Insertion devices (undulators) in particle colliders.

1st Generation:
Parasitic use of synchrotron radiation



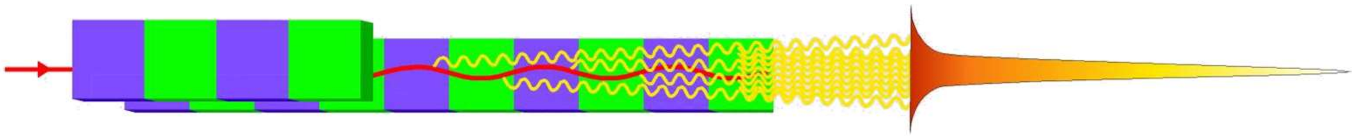


- Synchrotron radiation from a dipole magnet
  - Bremsstrahlung
  - Broad spectrum
  - Wide emission angle



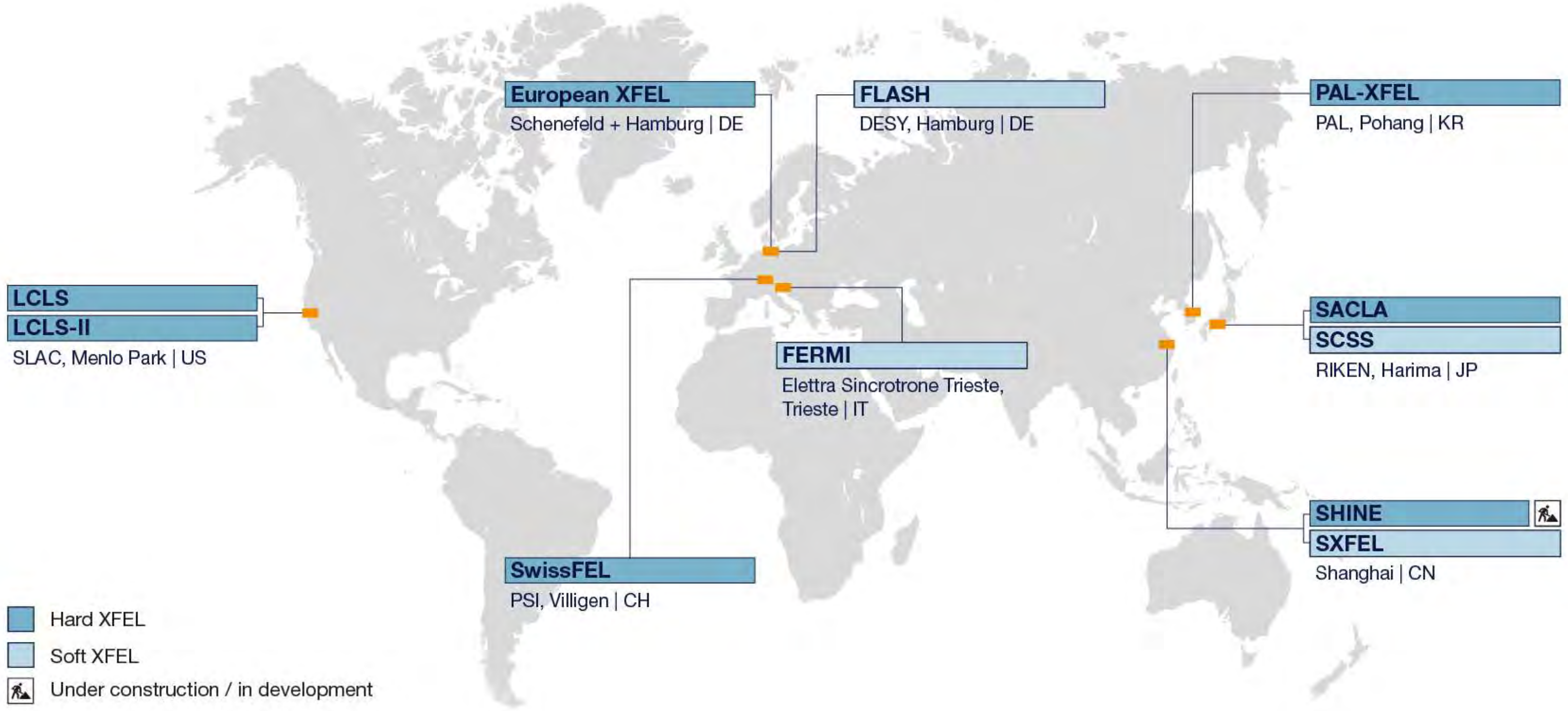
- Undulator radiation
  - Constructive interference of X-rays
  - Tunable narrow spectrum
  - Reduced divergence

- Free-Electron Laser
  - Constructive interference of X-rays **and electrons**



Illustrations: DESY 2006

X-ray free-electron lasers worldwide



Hard X-ray Free-Electron Lasers

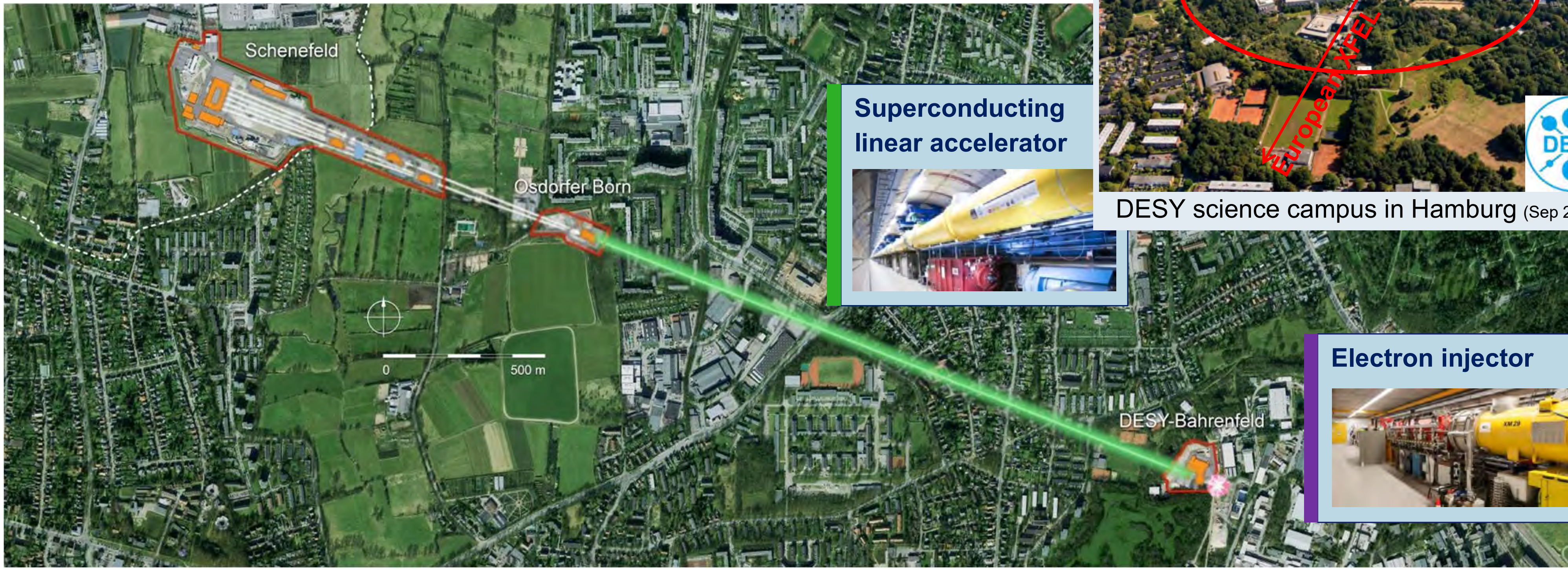
Comparison of some key parameters

	Eu XFEL	LCLS	LCLS II SC	SACLA	SwissFEL	PAL-XFEL	SHINE
Location	Germany	USA	USA	Japan	Switzerland	Korea	China
Commissioned	2016	2009	2020	2011	2016	2016	2025
Flashes per second	27 000	120	1 000 000	60	100	60	1 000 000
Minimum wavelength	0.5 Å	1.5 Å	2.5 Å	0.8 Å	1 Å	0.6 Å	0.5 Å
Electron energy	17.5 GeV	14.3 GeV	5 GeV	8.5 GeV	5.8 GeV	10 GeV	8 GeV
Undulators	3	1		3	1	2	
Experiments	6	5		4	3	3	
Brilliance	5×10^{33}	2×10^{33}	1×10^{32}	1×10^{33}	1×10^{33}	1.3×10^{33}	1×10^{33}

European XFEL—a leading new research facility

The European XFEL electron accelerator and X-ray laser

Total length: 3.4 km

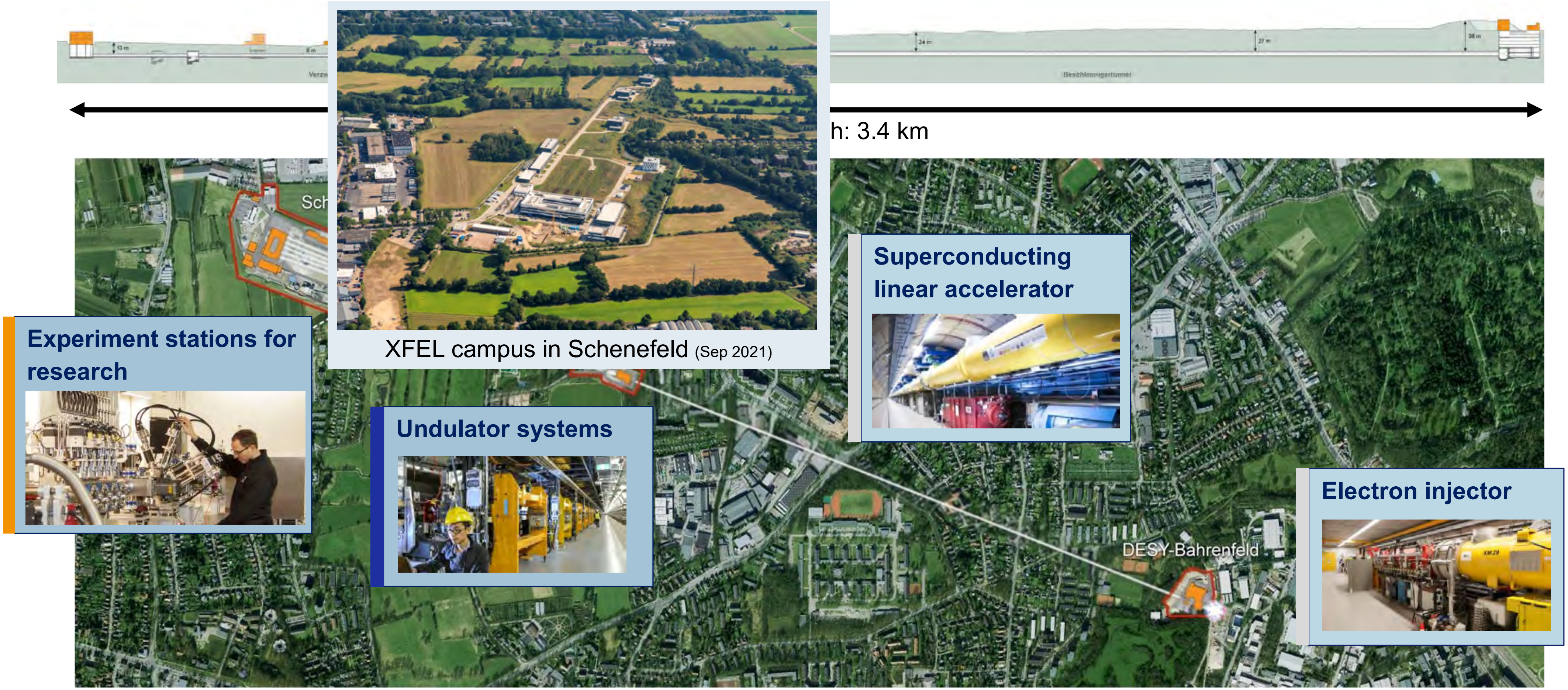


Superconducting linear accelerator

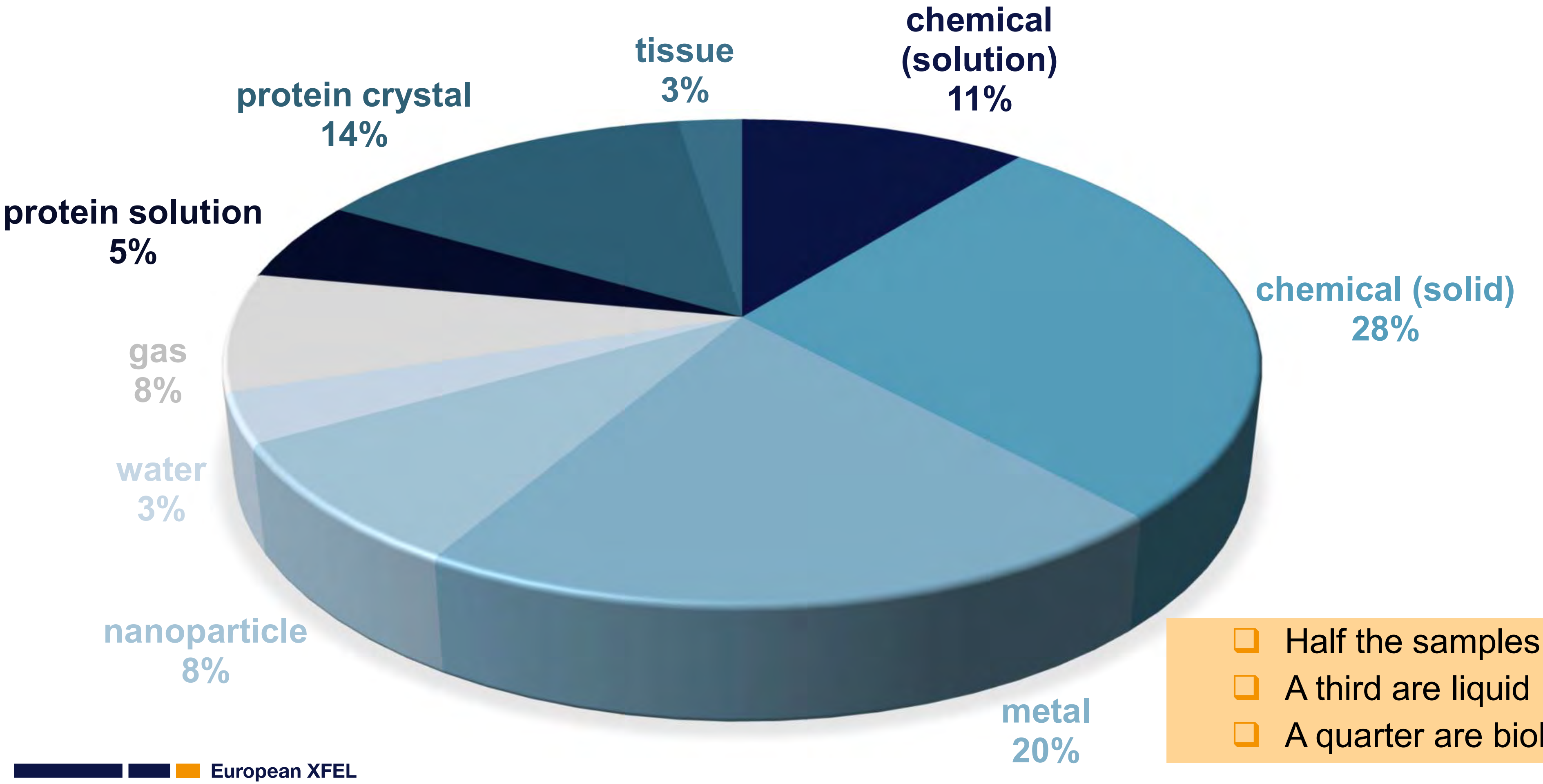
Electron injector

DESY science campus in Hamburg (Sep 2021)

The European XFEL electron accelerator and X-ray laser



Sample types from user beamtime proposal (recent 1 year)



- Half the samples are solid
- A third are liquid
- A quarter are biological

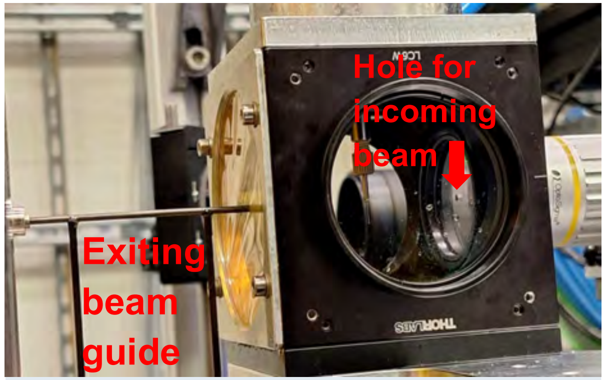
Information was extracted by Huijong Han from proposal A-form sample info.

Many types of samples – What we want to study

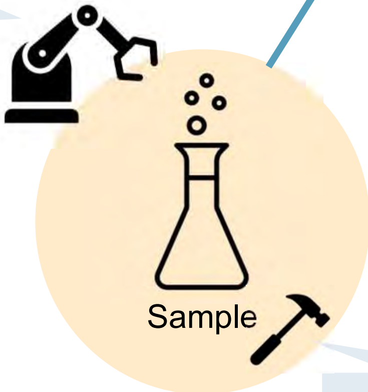
- Samples state of matter:
  - Plasmas
  - Gases
  - Aerosols
    - ▶ Nanoparticles
    - ▶ Droplets
    - ▶ Protein complexes
    - ▶ Viruses
  - Liquids
    - ▶ Liquified gases
    - ▶ Solutions
    - ▶ Suspensions
    - ▶ Complex liquids
      - Lipidic cubic phase
  - Solids
    - ▶ Amorphous
    - ▶ Polycrystalline
    - ▶ Single crystal
    - ▶ Soft matter
- Sample size:
  - Atoms, molecules
  - Nanoparticles
  - Microscopic
  - Macroscopic
- Region of interest
  - Surface
  - Bulk
  - Interface
- Sample conditions
  - Temperature
  - Pressure
  - Humidity
  - Electric fields or currents
  - Magnetic fields
  - Orientation
  - Environment
    - ▶ Vacuum
    - ▶ Helium
    - ▶ Air
    - ▶ Suspended in liquid

Sample Environment – further considerations

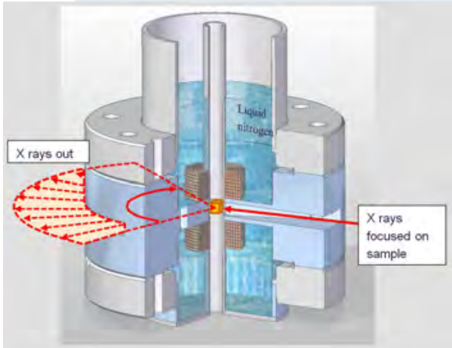
Sample handling
- ✓ Positioning
- ✓ Rotation
- ✓ Stability

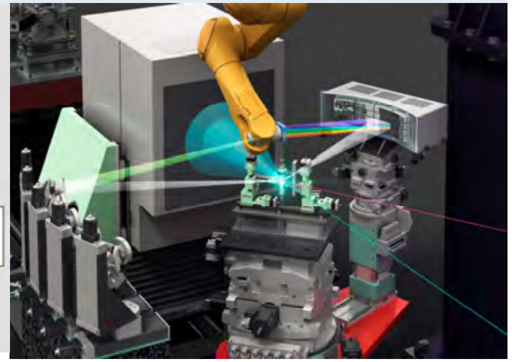


Incoming XFEL Beam
- ✓ Sample-beam alignment
- ✓ Beam profile and size
- ✓ Avoid scattering



Measured signals
- ✓ What? Photons, electrons, ions, current,…
- ✓ Where? Opening angles
- ✓ How? Cutouts, windows, flight paths, mirrors,..



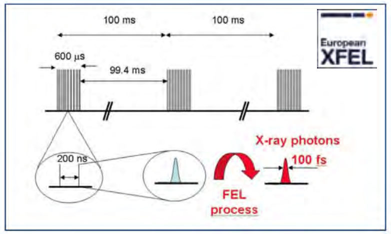


Starting a process
- ✓ Pump lasers
- ✓ Mixing
- ✓ Temperature jumps
- ✓ Electric fields
- ✓ Magnetic fields
- ✓ Compression

Specific considerations for XFELs

Pulse Structure and Repetition Rate

XFELs are pulsed sources



4.5 MHz: only 200ns to replace sample

$$\frac{10\ \mu m}{200\ ns} = 50\frac{m}{s} = 50\ 000\ \frac{mm}{s}$$

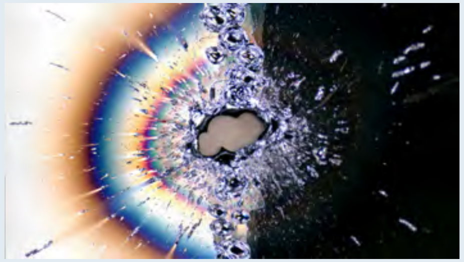
Fast Liquid Jets

180 km/h

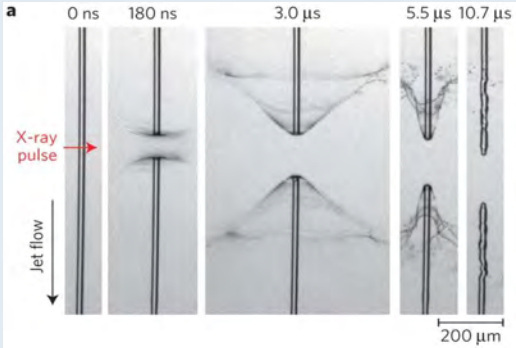
Fast sample delivery

High Power Density

Destroys solids in a single shot

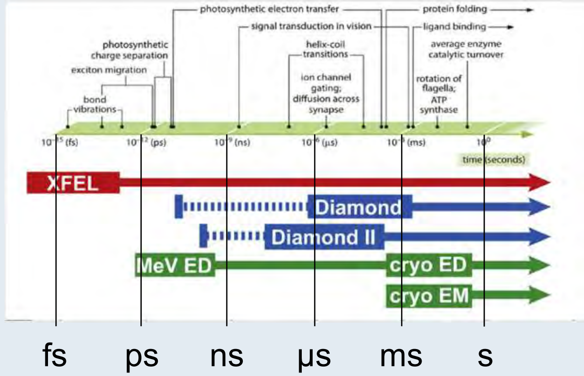


Plasma formation in liquids



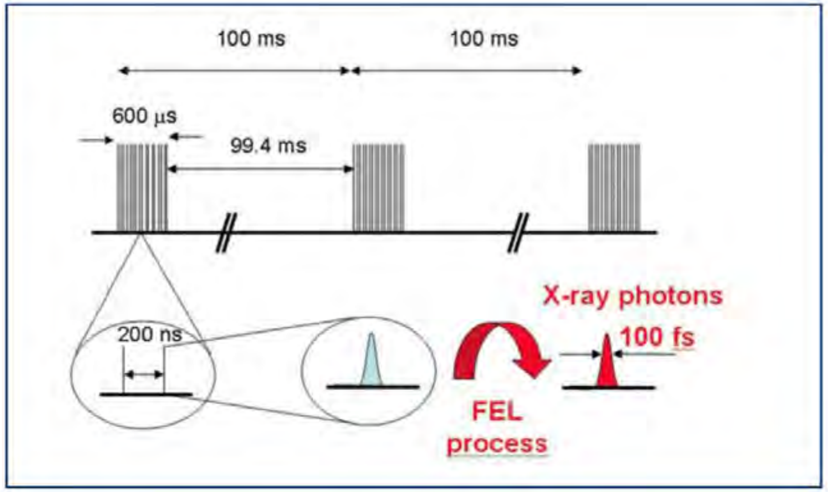
Short pulse length

Pulse length: ~50 fs
Attosecond mode available

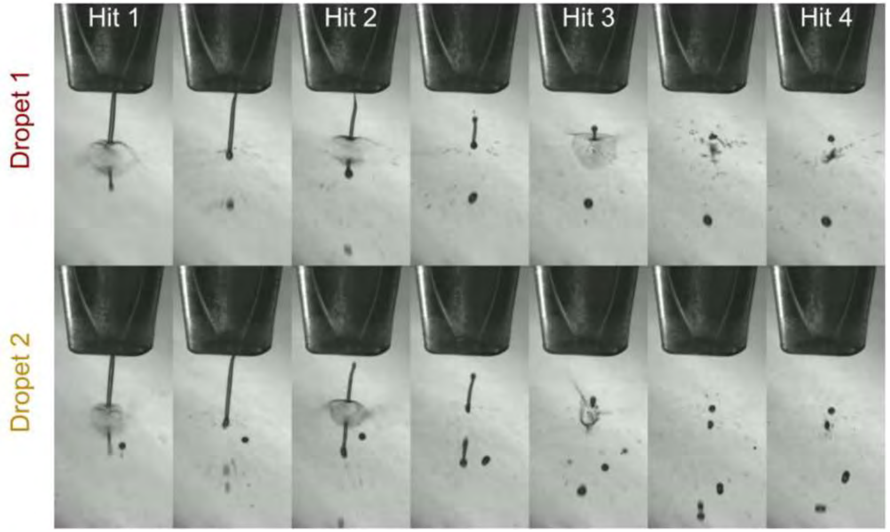


Synchrotron pulse length: ~40 ps

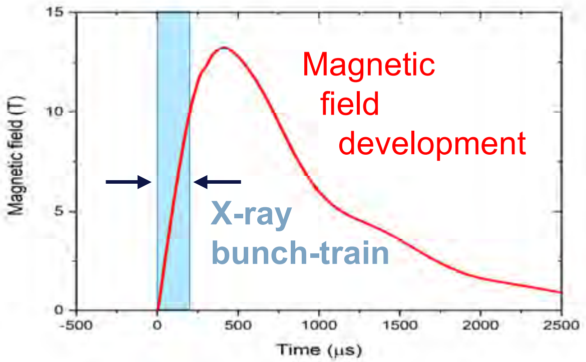
Use of the Bunched Pulse Structure at the European XFEL



Efficient sample delivery



Compact Pulsed Magnet

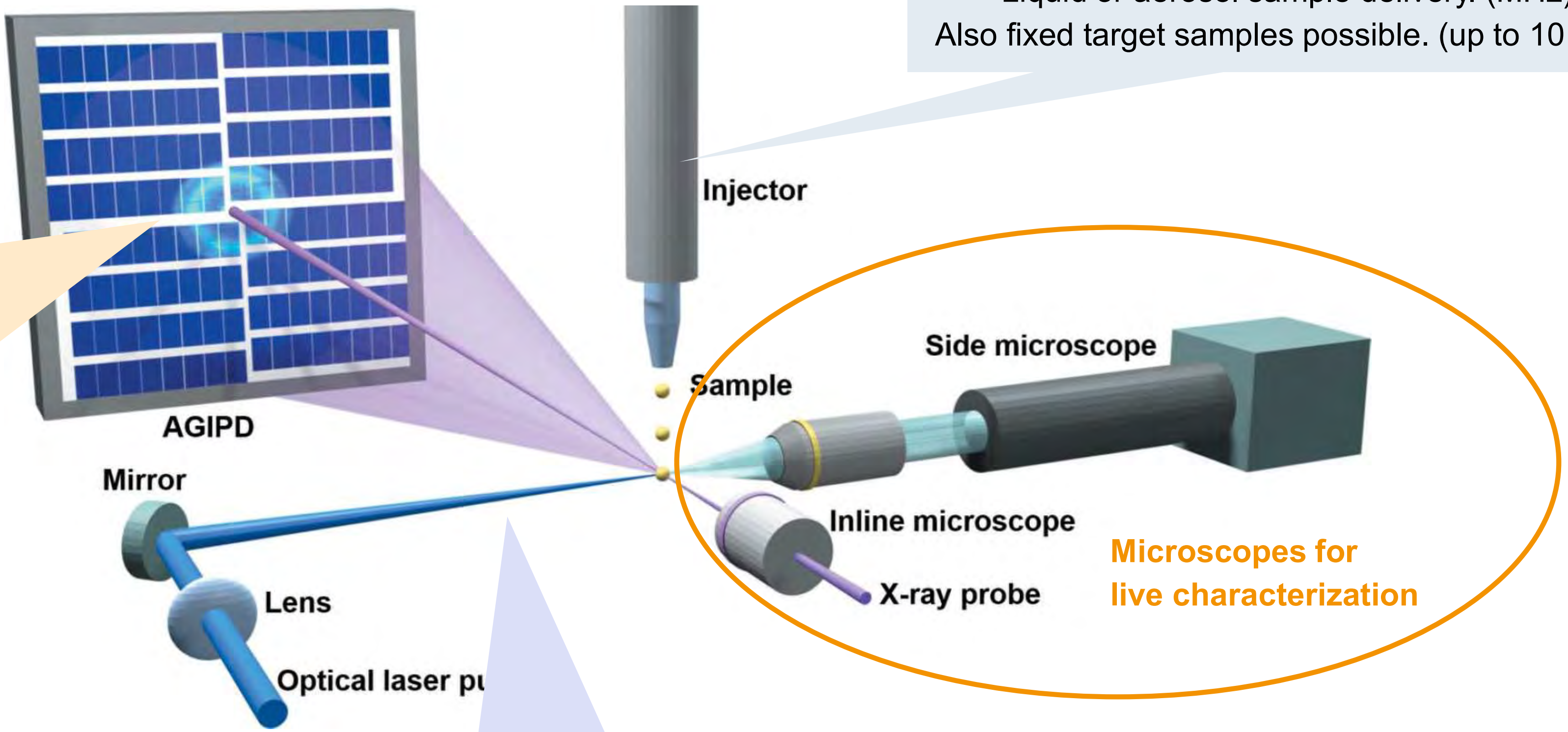


- Drop-on-Demand system (Microfab) at FXE
  - 16 pulses at 47 kHz per bunch train
  - Four elongated droplet each hit four times
  - 160 Hz average repetition rate
  - Reduced sample consumption 1% compared with continuous jet.

Time Resolved Serial Femtosecond Crystallography at XFELs

High **XFEL power density** delivers enough photons for a **full diffraction pattern** per **single femtosecond shot**.

Liquid or aerosol sample delivery. (MHz)
Also fixed target samples possible. (up to 10 kHz)

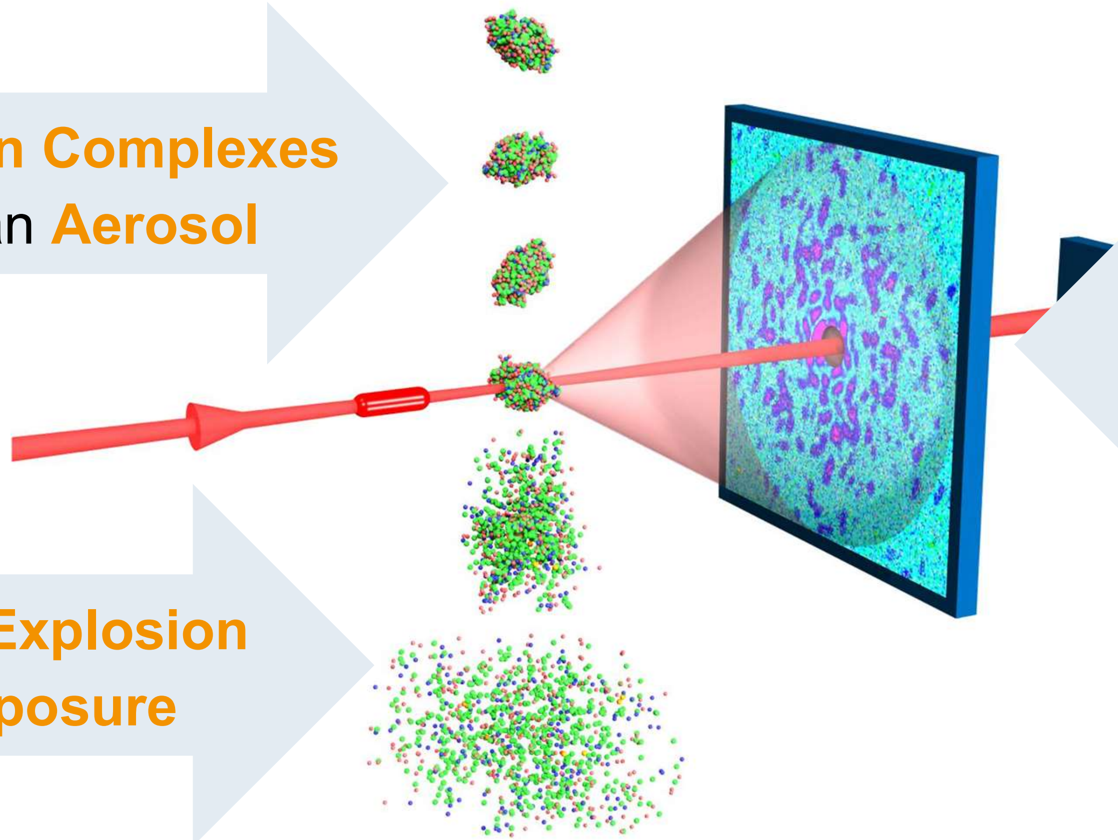


A **femtosecond pump-probe** laser with **adjustable delay** starts biochemical process.

Koliyadu et al, 2022, *J Synchrotron Rad* 29

Diffraction before Destruction
Making use of High Photon Flux and Femtosecond Bunches

Protein Complexes in an **Aerosol**

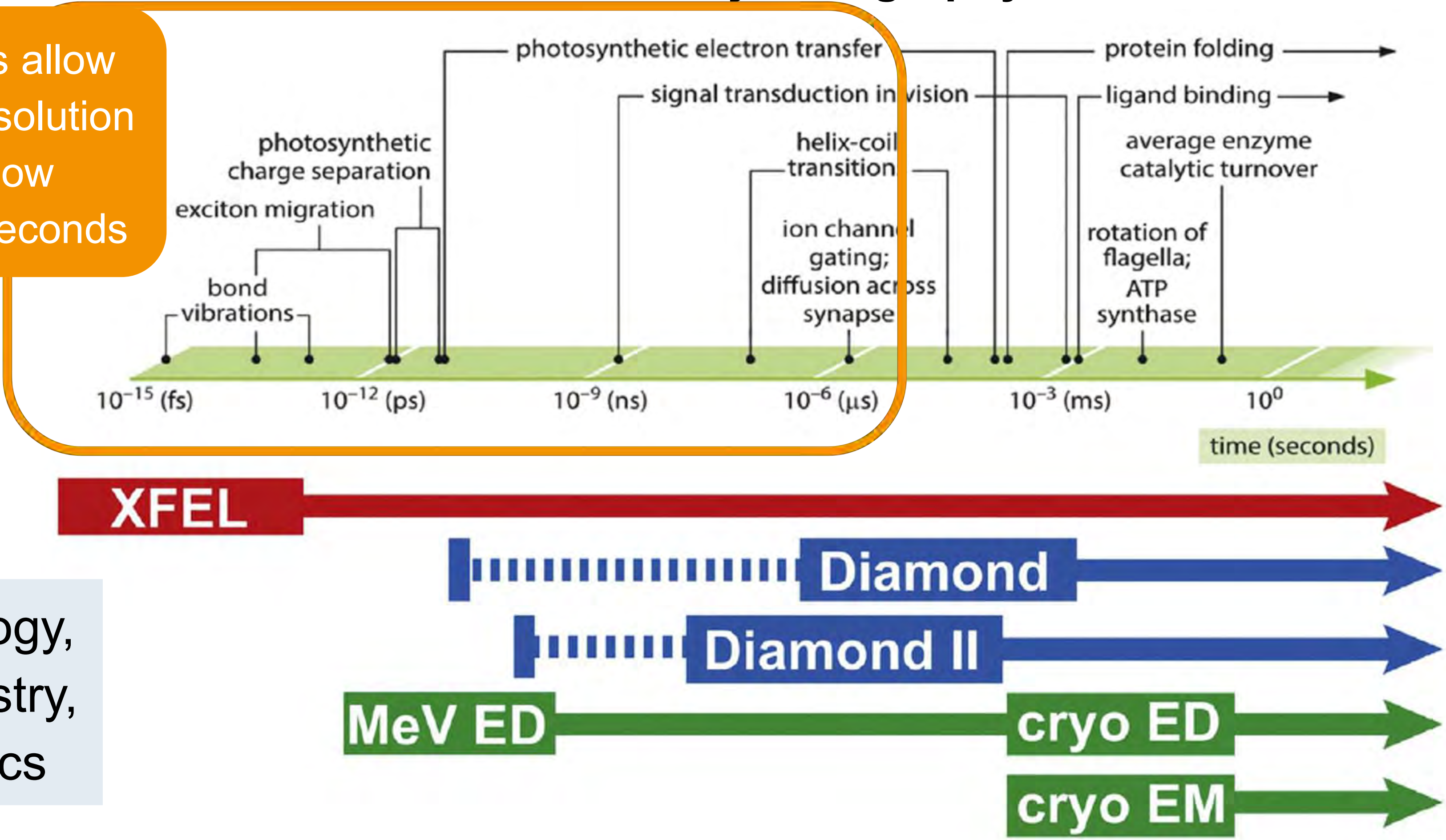


Single Particle Imaging in a Flash

Coulomb Explosion after **Exposure**

Time Resolved Serial Femtosecond Crystallography at XFELs

XFELs allow time resolution below microseconds



In Biology, Chemistry, Physics

Orville, 2020, *Current Opinion in Structural Biology, 65:193–208*

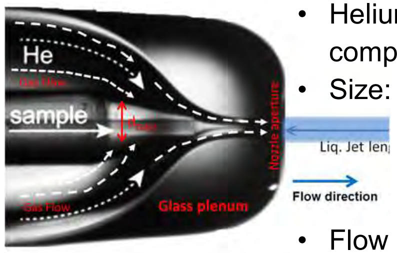
Types of Liquid sample delivery

Jets

Rayleigh jet

- Round nozzle
- Size: Garden hose down to 25 µm
- Flow rate: ~ml/min

Gas Dynamic Virtual Nozzle (GDVN)



- Helium stream compresses liquid
- Size: <1 µm
- Flow rate: ~10 µl/min

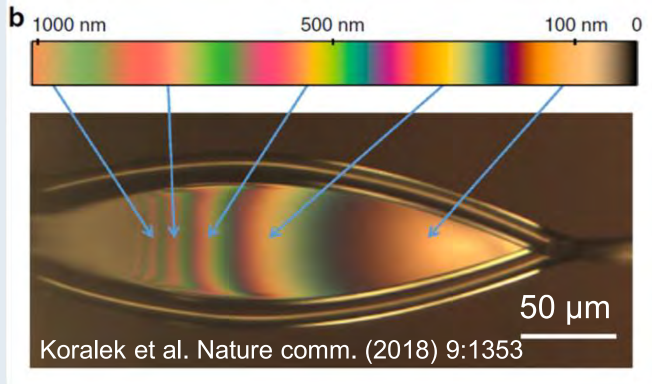
Drops

Jet breakup

Drop-on-demand

- Ink-jet printing technology
- Piezo-driven shock-wave creates a single drop
- < 1 µl/min

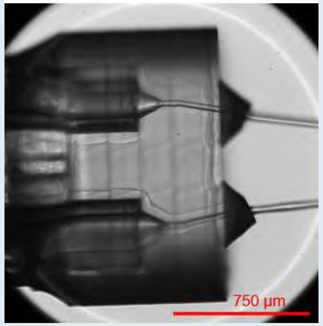
Sheets



Liquid sheet jets

can be created by

- Slit-nozzles
- Gas-dynamic compression of a round jet
- Colliding two round jets



Types of Liquid sample delivery

Rayleigh Jets

+	-
easy	sample consumption
volume	Icing in vac.

Hard X-ray spectroscopy

Jet breakup

+	-
easy	
Rep. rate	Sample consumption

Phase change

Tiny sheets

+	-
Stability	Low volume
Vacuum	
Geometry	

Soft X-ray spectroscopy
Single particle imaging

GDVN

+	-
Vacuum compatible	glogging
Low background	

Serial crystallography

Drop-on-demand

+	-
Sample saving	No vacuum
	Icing/clogging

Serial crystallography

Colliding sheets

+	-
Volume	Sample consumption
geometry	No vacuum

Hard X-ray spectroscopy

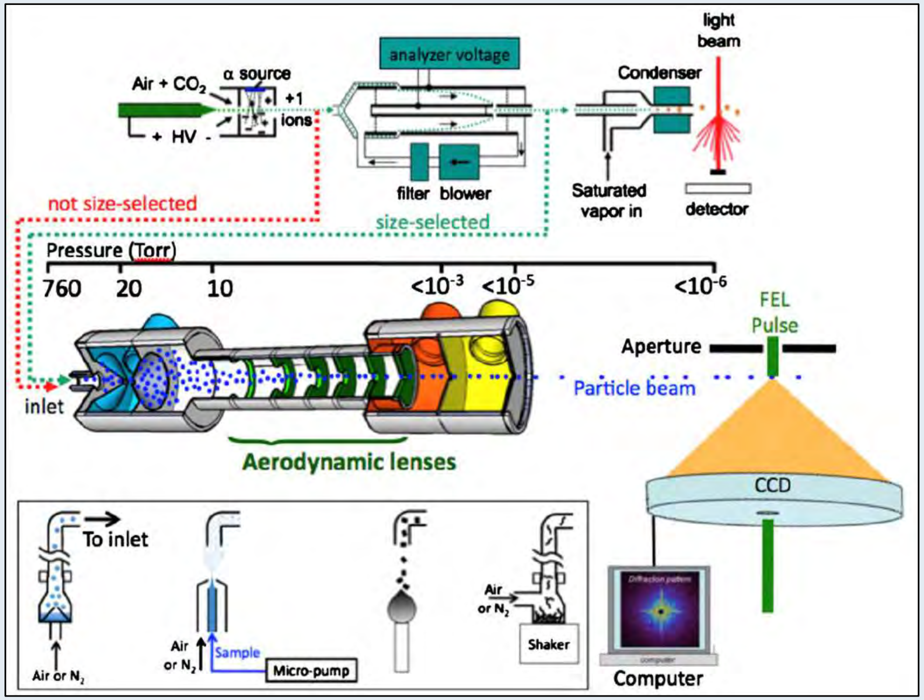
Special types of Liquid sample delivery

High viscosity extruders

Lipid cubic phase

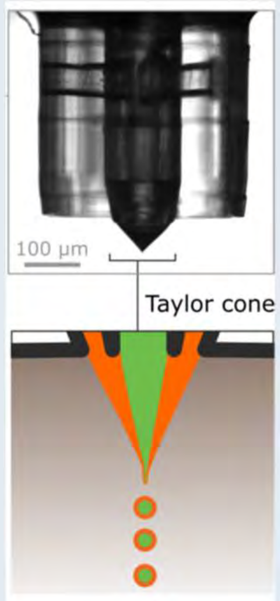
Membrane proteins often crystallize in highly viscous media.

Aerosol sample delivery



M.J. Bogan, et al.: Phys. Rev. ST Accel. Beams 13, 094701 (2010)

Electrospray



Creates **highly charged** droplets by pulling liquid with an electric field.

Two concepts for nozzle handling

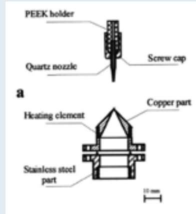
LCLS nozzle shroud 2009

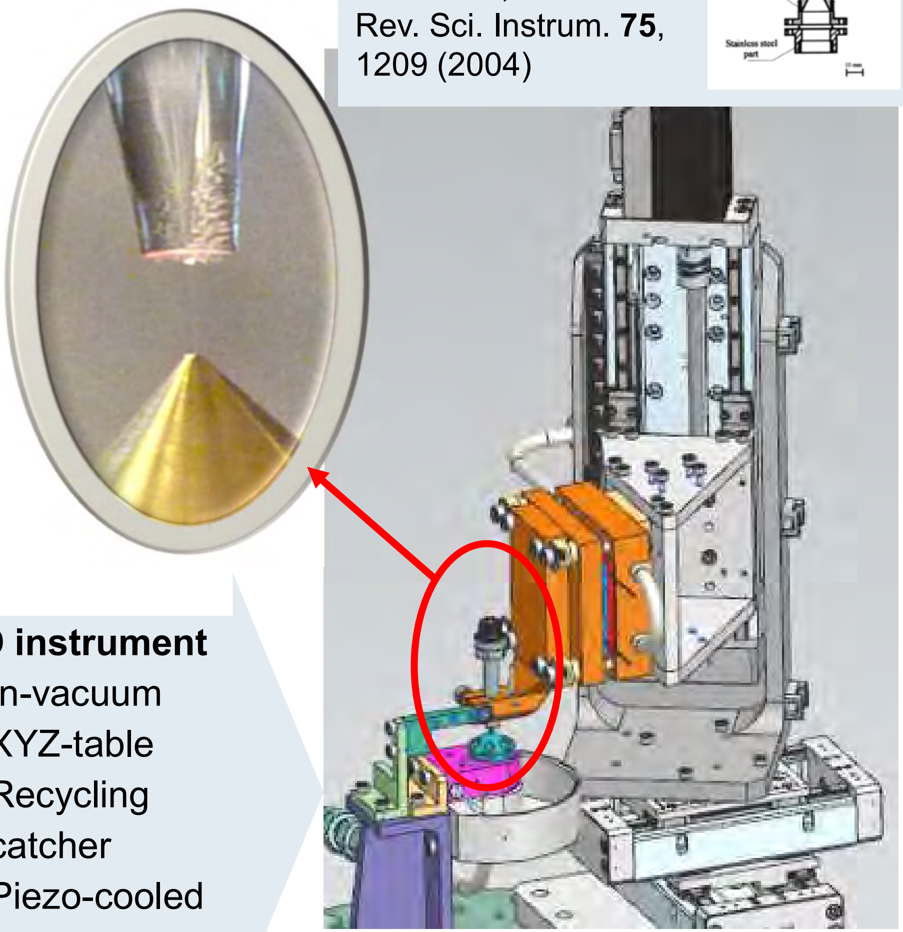
SPB/SFX instrument

- Adjustable shroud with holes
- Top-loaded nozzle rods
- Load-lock
- Pumped from below

A versatile liquid-jet setup for the European XFEL
J. Schulz et al.
J. Synchrotron Rad. (2019). **26**, 339–345

“Göttingen” Jet
A. Charval, E. Lugovoj, M. Faubel, B. Abel
Rev. Sci. Instrum. **75**, 1209 (2004)



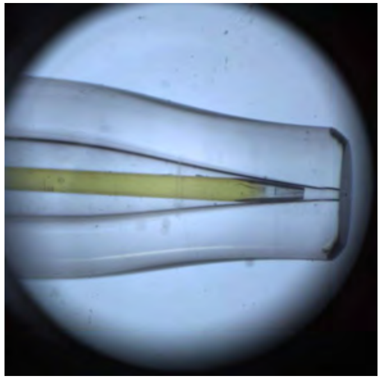


MID instrument

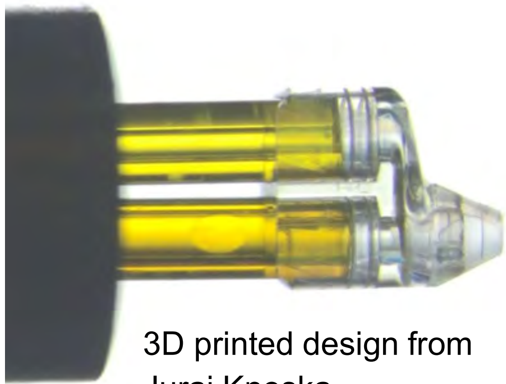
- In-vacuum XYZ-table
- Recycling catcher
- Piezo-cooled nozzle tip

Nozzle production methods

- Nozzle production is time consuming
  - For a long time GDVNs were produced by hand
    - ► Grinding nozzles demands skill and patience
    - ► The reproducibility is limited, every nozzle is different
  - Since 2018, we use a Nanoscribe 3D printer
    - ► This makes the tips reproducible
    - ► Assembly still requires a steady hand
  - Microfluidic chips have the potential for mass production
    - ► Laser etching in glass
      - Dan DePonte (SLAC) developed a standard
      - A hand full flat sheet devices commercially available
      - These nozzles are in use at our SCS instrument
  - Injection moulding
    - ► Many materials possible: plastic, ceramics, metals
    - ► Project at European XFEL in preparation

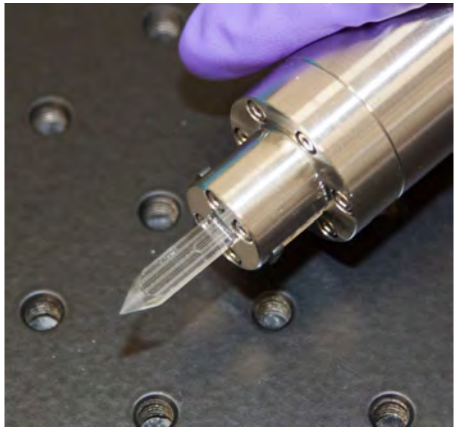


Hand grounded Glass nozzle



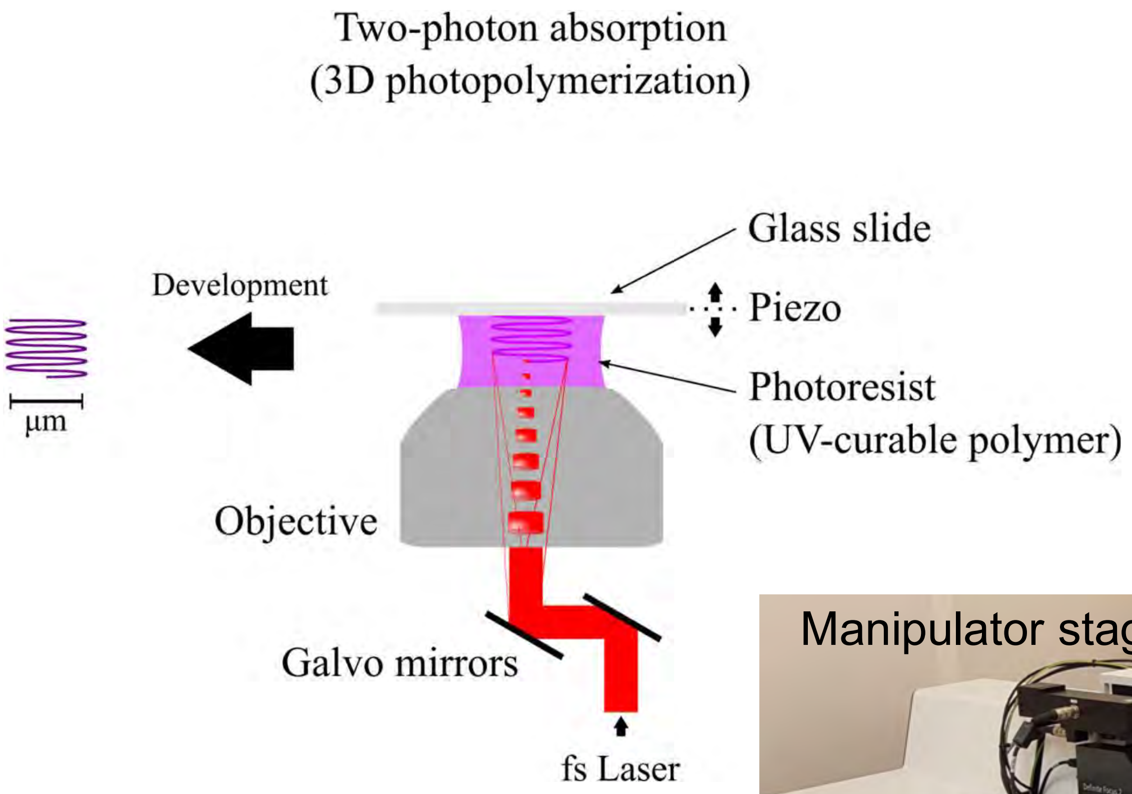
3D printed design from Juraj Knoska, Michael Heymann

Our current preference!



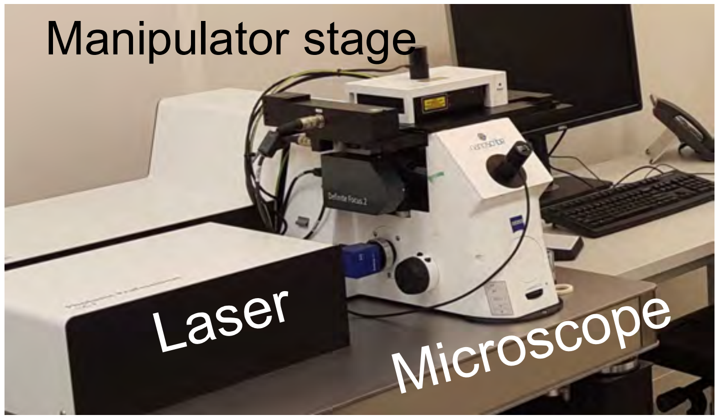
Glass device from Femtoprint developed by Rita Graceffa

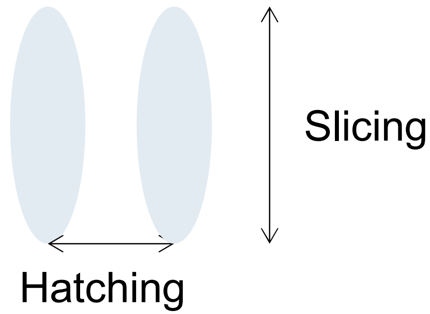
Nanoscribe 3D printer



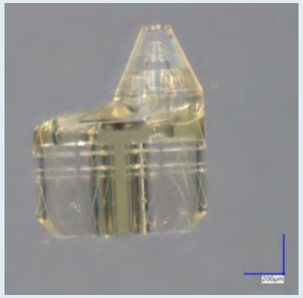
https://support.nanoscribe.com

Objective	63x	25x	10x
Resolution in *x*, *y* (width) [µm]	0.3	0.5	1.5
Resolution in *z* (height) [µm]	1	3	15
Hatching distance [µm]	0.2	0.5	1
Slicing distance [µm]	0.3	1	5
Print time [mm³/h]	0.4	0.8	6.6
Print field [Ø µm]	200	400	Ø 1000

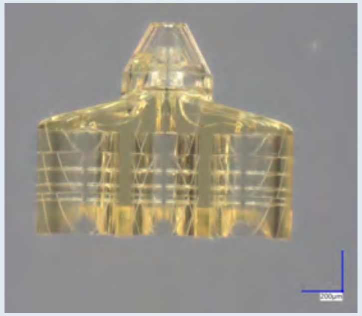




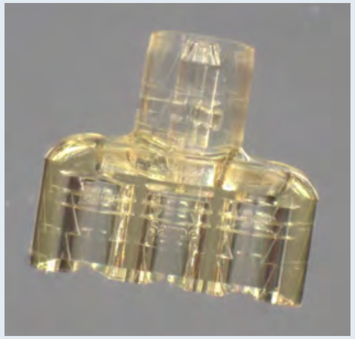
Range of 3D-printed nozzle devices



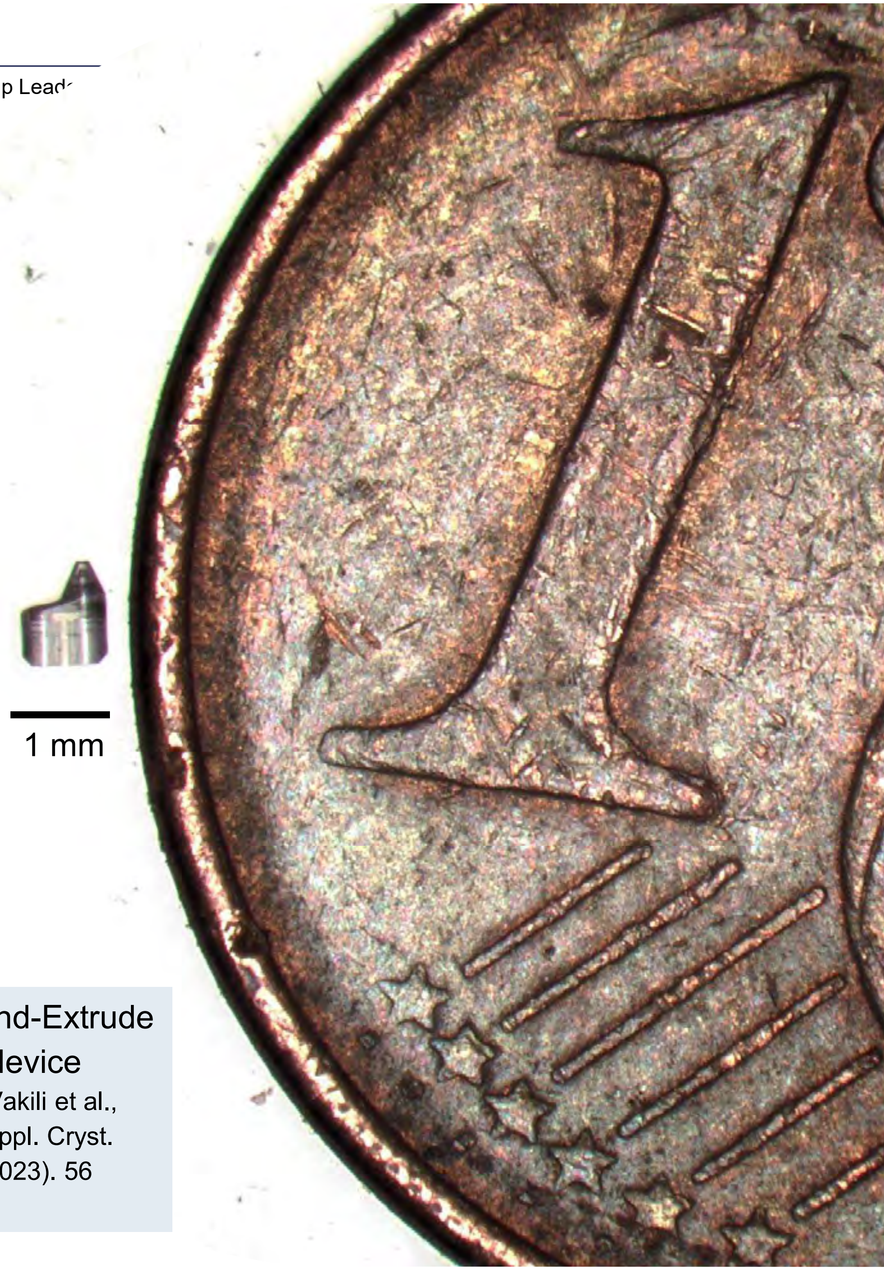
Gas Dynamic Virtual Nozzles (GDVNs)
Juraj Knoska et al.
Nature Comm. 11:657 (2020)



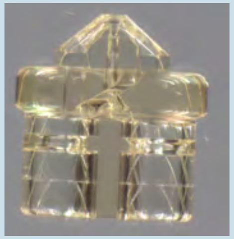
Double Flow Focussing Nozzles (DFFNs)
Juraj Knoska et al.
Nature Comm. 11:657 (2020)



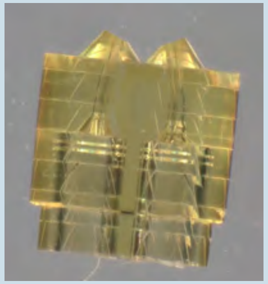
Coaxial Helium Electrospray (CHeES)
S. Rafie-Zinedine *et al.*,
J. Synch. Rad. 32 (2025)



Many different sizes:
Liq:Gas:Dist
30:30:30
50:60:60
75:60:75
100:90:100
150:100:150
180:145:180



Gas driven sheet
T. Gallo et al.
J. Synch. Rad. 31 (2024)



Colliding Rayleigh Jets

Flat Sheet Nozzle

Mix-and-Extrude device
M. Vakili et al.,
J. Appl. Cryst.
(2023). 56

Warm thank you to…

- Agnieszka Wrona
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- Kristina Lorenzen
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- Sandra Plett
- Christina Schmidt
- Vasilii Bazhenov

European XFEL

Our world wide collaborators

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  - Alte Meents
  - Holger Fleckenstein
  - Dominik Oberthür
  - and teams
- LCLS
  - Ray Sierra
  - Dan Deponte
  - Marc Hunter
  - and Teams
- ASU
  - Rick Kirian
  - Alexandra Ros
  - Petra Fromme
  - and teams
- Swiss FEL
  - Emma Beale
  - and team
- PAL-XFEL
  - Jaehyun Park
  - and team
- MPI Heidelberg
  - Ilme Schlichting
  - Bob Shoeman
  - Bruce Doak
  - and team