LCLS | Linac Coherent Light Source

Probing Ultrafast Dynamics with X-ray FELs

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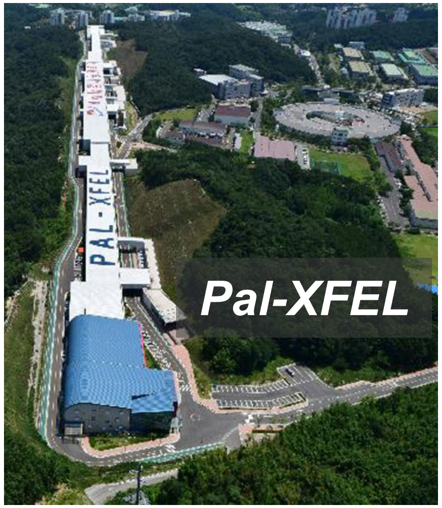
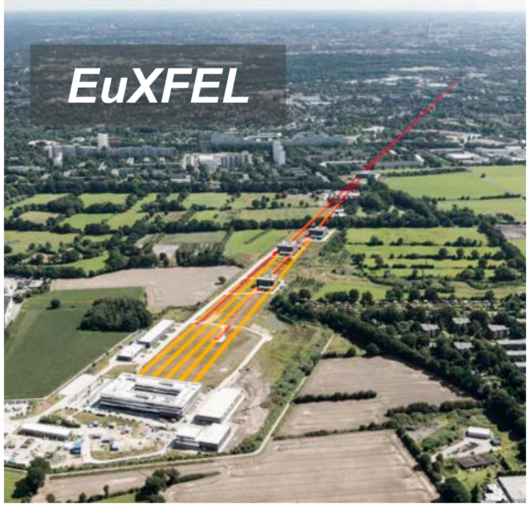
U.S. DEPARTMENT OF ENERGY

Office of Science

SLAC NATIONAL ACCELERATOR LABORATORY

Acknowledgement





Outline

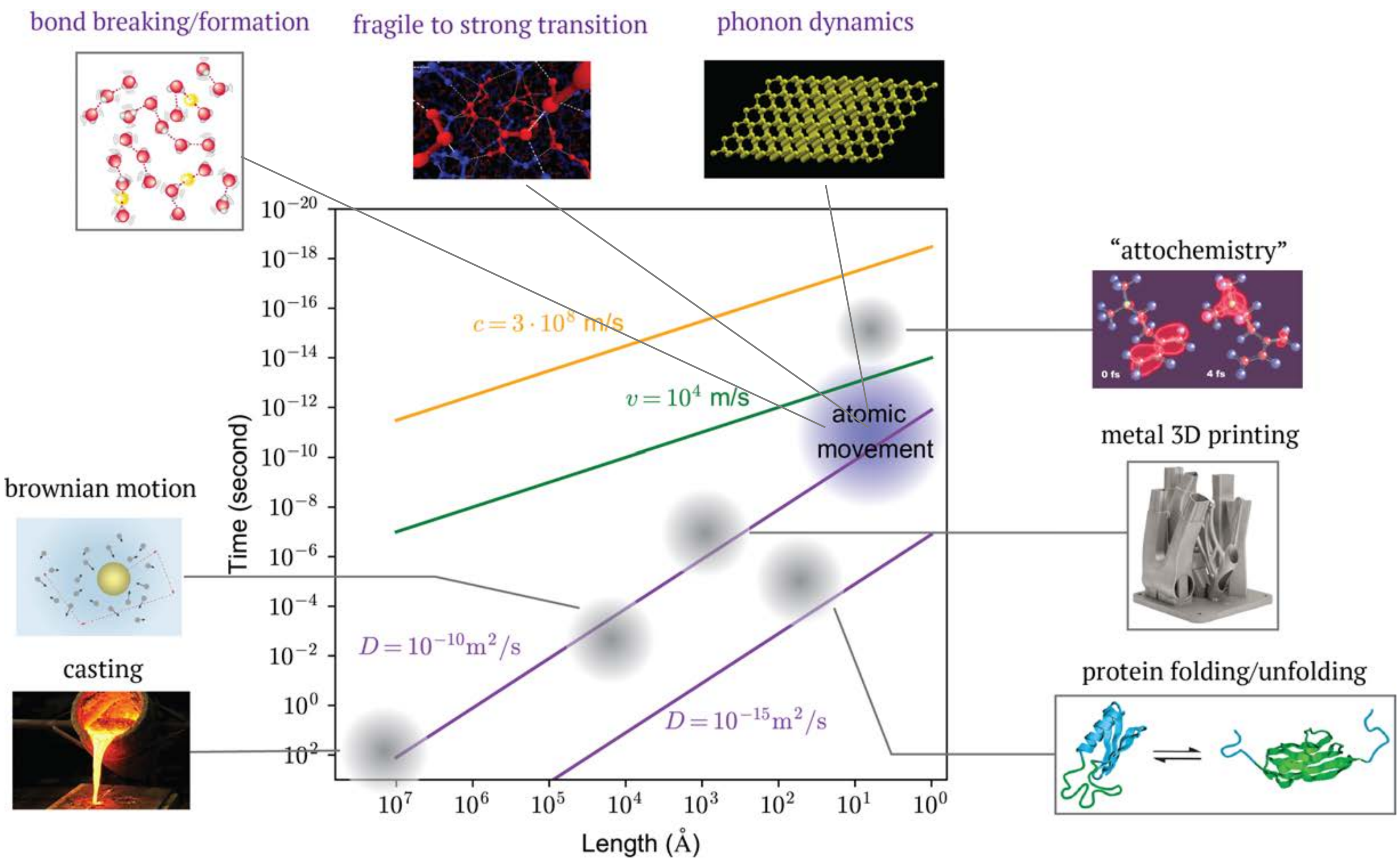
- Why time-resolved, why pump probe?
- Basic elements of an X-ray pump probe experiment
- How to get to femtosecond time resolution?
- Some examples and why they need an X-ray FEL
- Outlook, discussion, Q&As

Introduce the thought process in developing and evaluating a potential x-ray free electron laser experiment in the pump probe format.

image credit: http://www.hummingbirdsplus.org

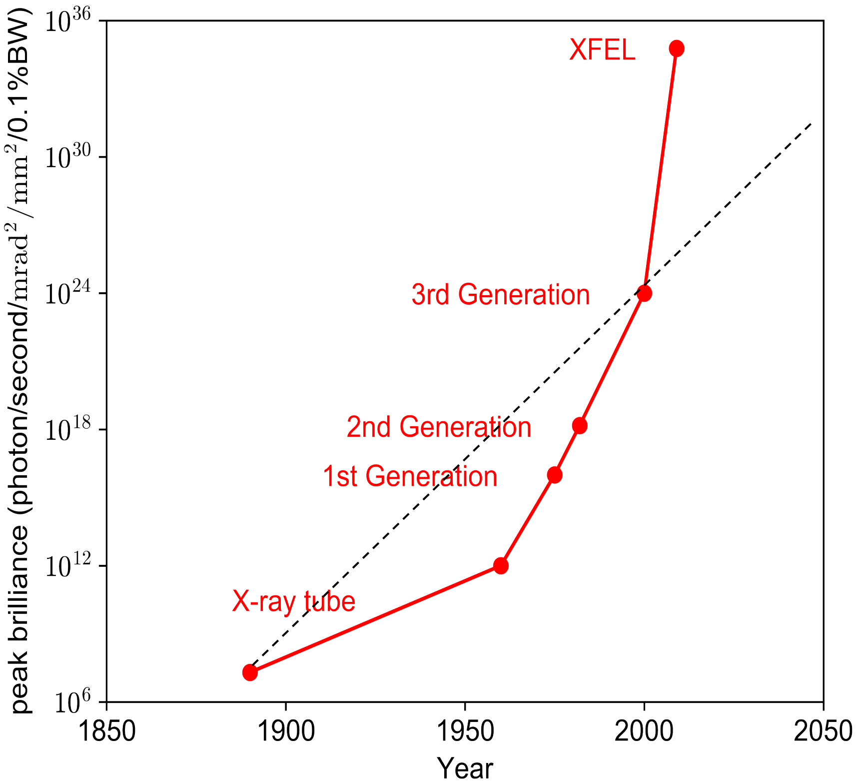

Bright flash light sequence was the key in capturing the ‘freeze motion’ details of the wing motion.
image credit: https://www.kobistudio.com

Dynamics becomes faster as we ‘zoom in’



Credit: Yanwen Sun

A ‘flash light’ for atoms and molecules



LCLS does not make more photons per second than APS. In fact, the average flux is quite a bit lower.

However, the key difference is 120Hz vs ~6.5MHz in 24 bunch mode, and the diffraction limited divergence of 2 microradian or so.

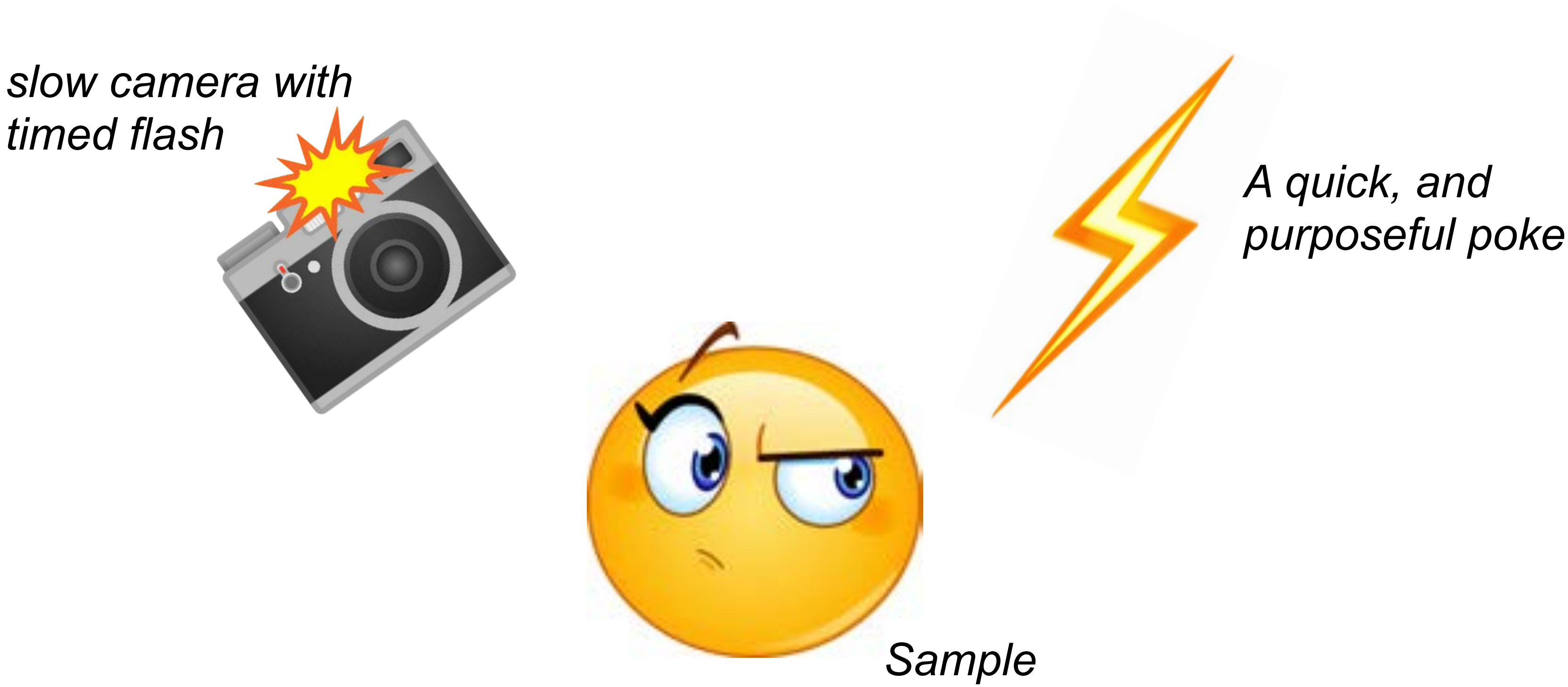
***Beam Parameters**:*

- *> 10^{12} photons in a ~10 fs pulse*
- *0.2 – 25 keV*
- *Fully transversely coherent*
- *Not quite yet fully longitudinally coherent but we have some good ideas to improve it.*

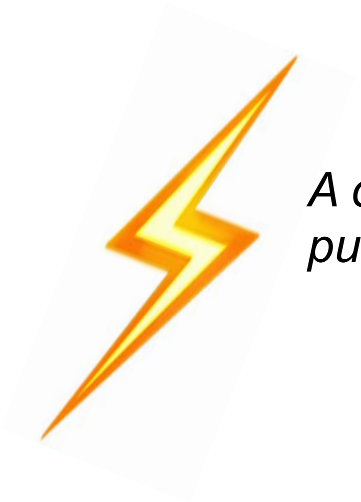
The challenges of going ‘faster’ & ‘brighter’

- Rapid succession of flashes not quite available yet.
- Highest frame rate detectors are still way too slow.
- Too bright a flash can start to disturb your object of interest.

The pump-probe experimental paradigm

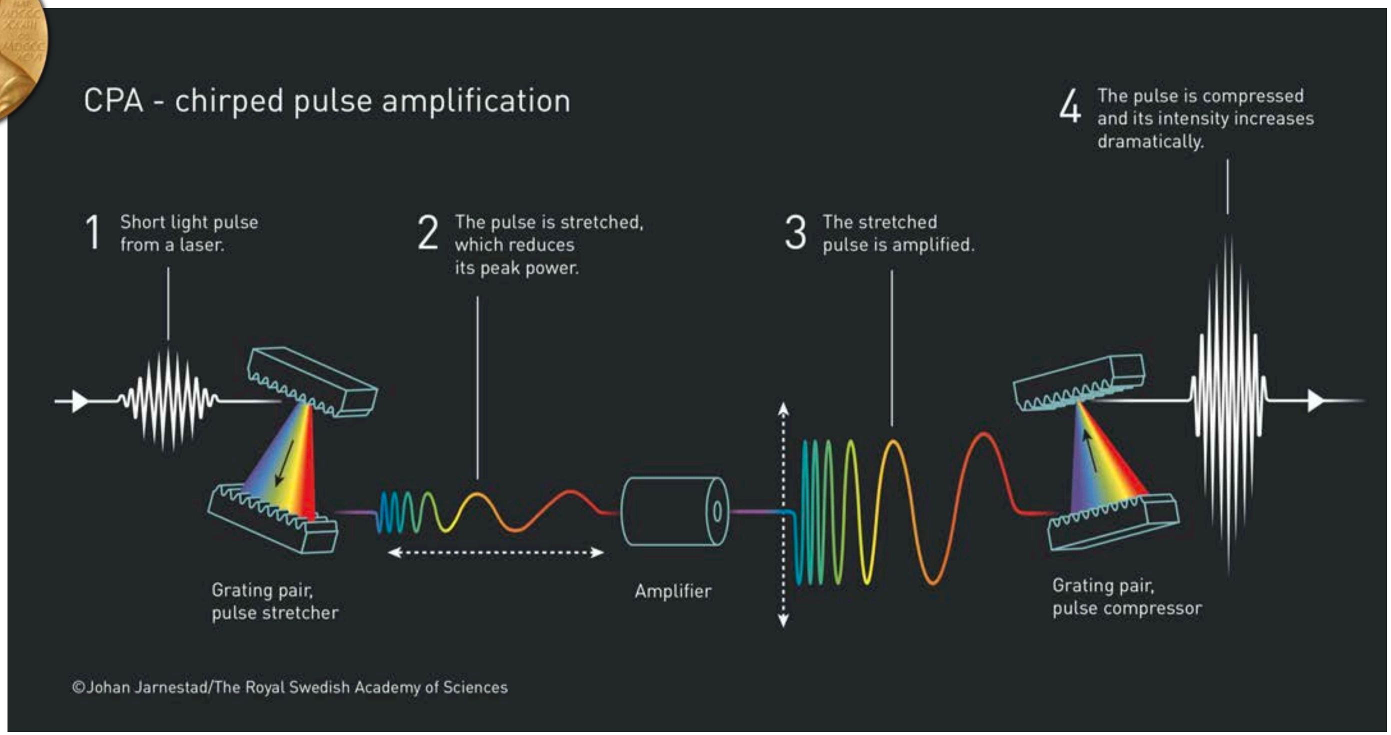


1. Need an ‘actor’ who’s willing to perform the same trick time and time again.
2. Need a prompt ‘poke’, and a brief bright flash.
3. Need to be able to time the flash accurately with respect to the ‘poke’
4. ‘Actor’ needs to return to ‘ground state’ before trying the sequence again, or have a replacement who would react to the ‘poke’ exactly the same way.



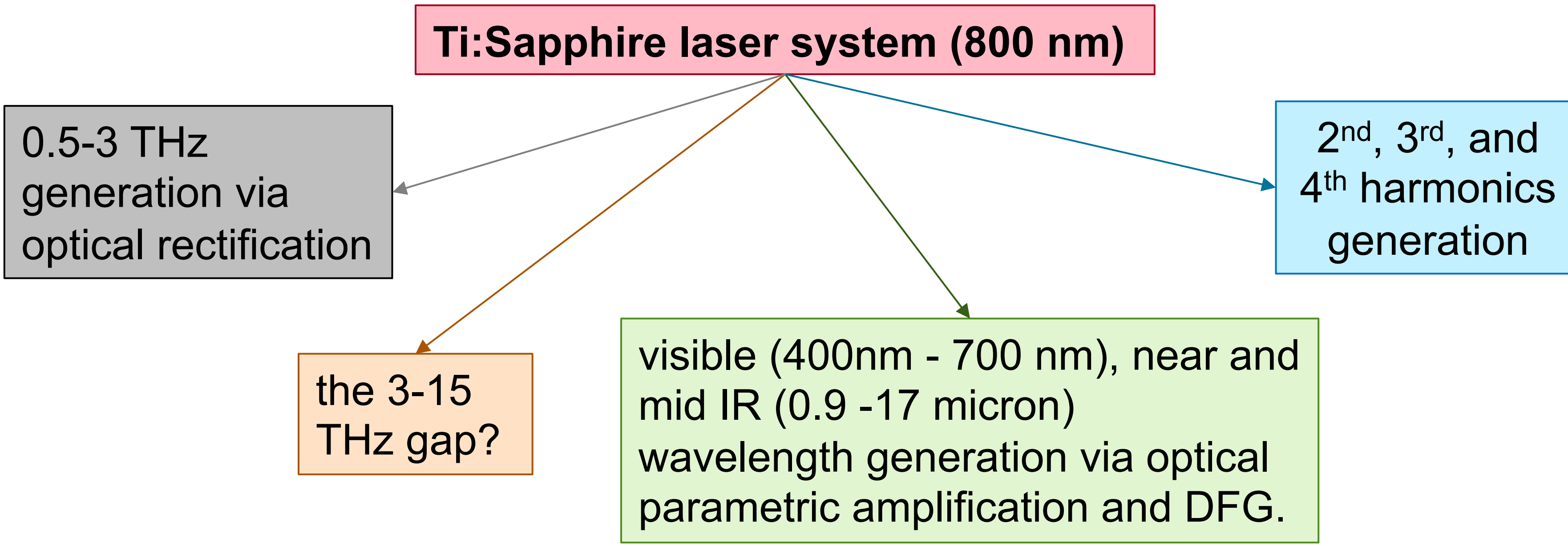
A quick, and purposeful poke

The femtosecond action trigger



Gérard Mourou and Donna Strickland shared the 2018 Nobel Prize of Physics "for their method of generating high-intensity, ultra-short optical pulses".

The femtosecond action trigger



THz: ultrafast polarization switching in ferroelectric materials, strong electric field driven tunneling in material, etc.

Mid-IR: resonance to particular phonons connected to, e.g. E-P coupling in Hi-Tc Superconductivity.

IR: electronic transition in semiconductors, vibrational resonance in materials and molecules, etc.

Visible: life, photo synthesis, electronic transitions in molecules, etc.

UV: bond breaking, photo chemistry, photocatalytic reactions, etc.

X-rays: element and chemical specific excitations, core electron excitations.

slow camera with timed flash

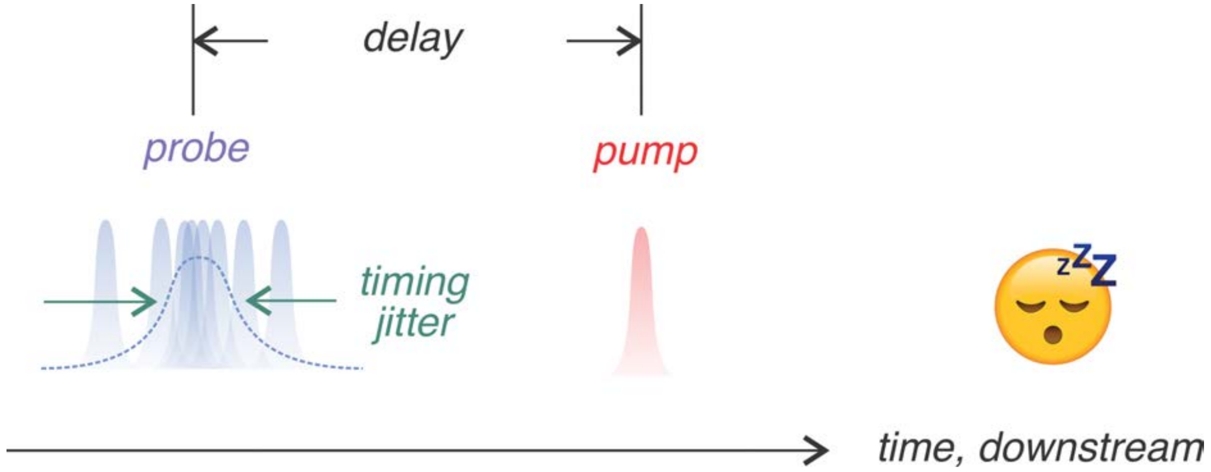
The Camera

- 100+ Hz Mpixel detectors at LCLS, SACLA, PAL-XFEL, SwissFEL.
- 4.5MHz burst mode detector at EuXFEL
- Many oher specialized detectors and new developments.

The Flash

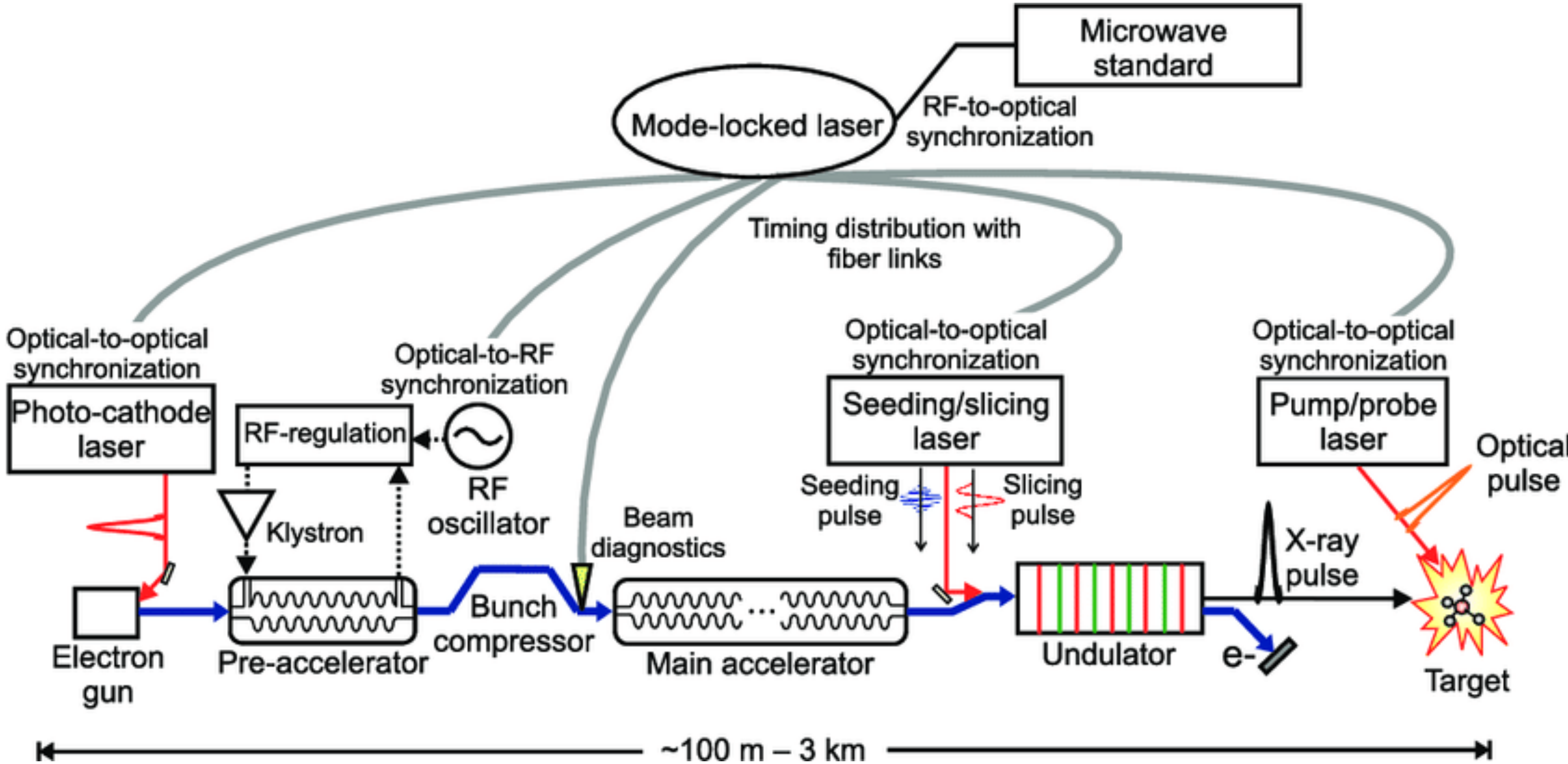
- APS, single bunch, ~ 50ps, 10^9 photons per pulse in white beam mode,1-2%BW.
- X-ray FELs, 1-100 fs, 10^{12} photons, 0.1% BW.
- Need to time up the flash with respect to the excitation.

Timing jitter and time resolution



$$\tau = \sqrt{\tau_{\text{pump}}^2 + \tau_{\text{probe}}^2 + \tau_{\text{jitter}}^2}$$

Timing synchronization & timing jitter



J. Kim and F. Kartner, Laser & Photonics Review **4**, 432 (2010)

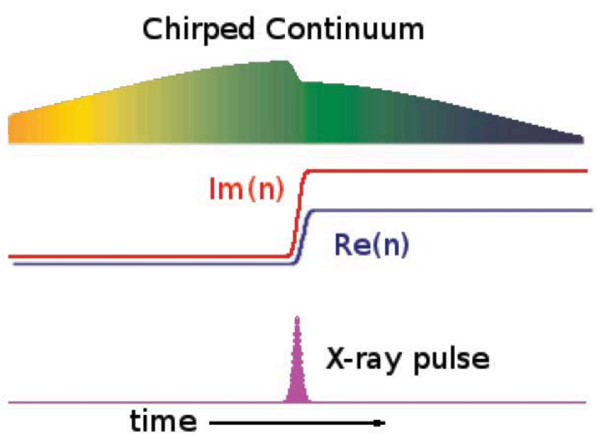
Timing synchronization & timing jitter

- Timing difference between the ‘clock’ signal across the facility.
- Timing synchronization error of the at the electron gun (injector laser)
- Accelerator’s RF amplitude and phase noise leads to jitter in electron beam arrival time.
- X-ray generated by the electron may not necessarily at the center of the electron bunch.
- Vibration of x-ray optics, sample …

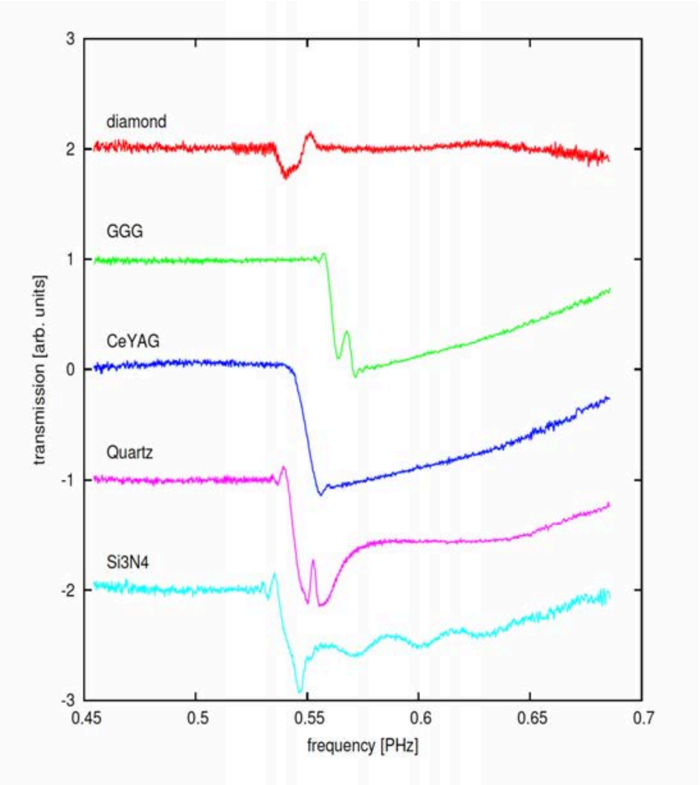
State of the art in synchronization has been moving from 100 fs to 10 fs (RMS) over the past decade.

Shot-to-shot measurement of the timing jitter

Example of spectral-temporal encoding

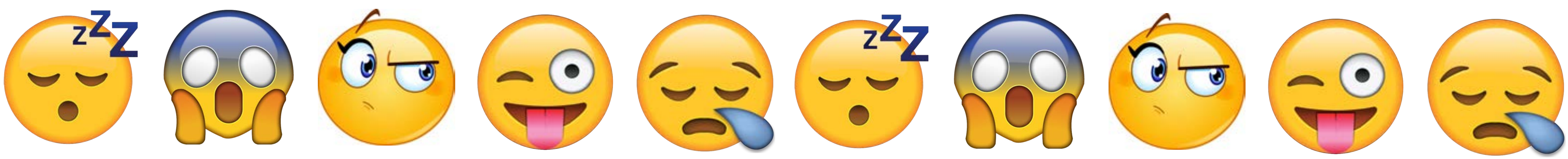


X-ray generate material optical property change is mapped over the spectrum of a chirped pulse.



This, as well as spatial-temporal encoding method, have achieved few-femtosecond scale precision in measuring the shot-to-shot timing jitter.

M. Harmand, *et al.*, Nat. Photonics **7**, 215 (2013), M. Bionta, *et al.*, Rev. Sci. Instr. **85**, 083116 (2014), N. Hartman, *et al.*, Nat. Photonics **8**, 706 (2014)



Sample: you diligent actor

Instantaneous 'heating' and material damage

SLAC

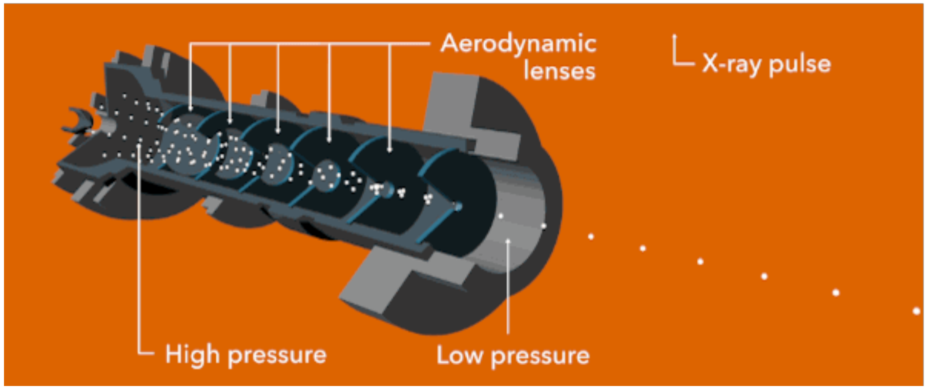
Absorbed dose (eV/atom) under pink beam: 1 mJ @ 10 keV

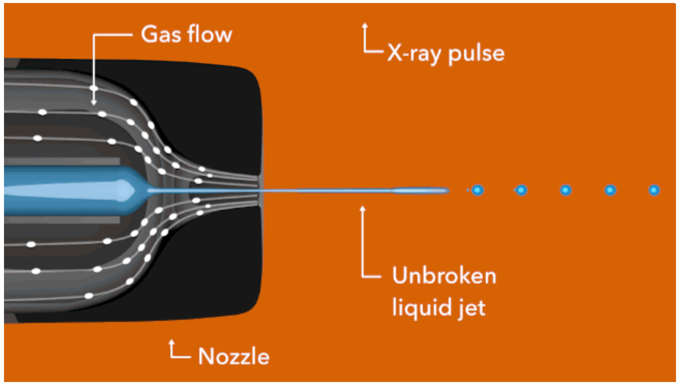
	100 nm	1 um	10 um	100 um
diamond	2300	23	0.23	0.0023
water	2700	27	0.27	0.003
silicon	85000	850	8.5	0.085
iron	870000	8700	87	0.87
gold	2050000	20500	205	2

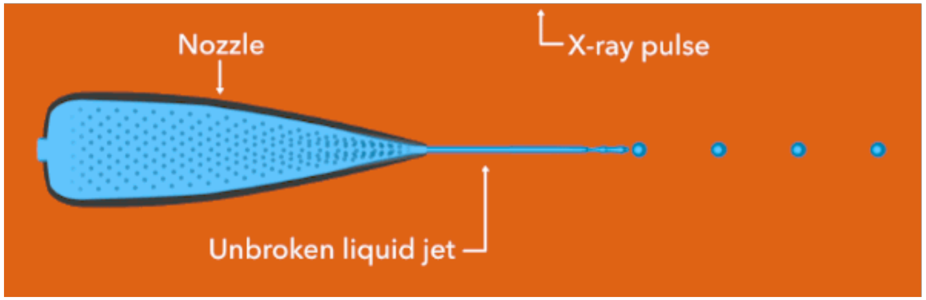
Absorbed dose under mono beam: C(111), 0.5 eV, 0.01 mJ @ 10 keV*

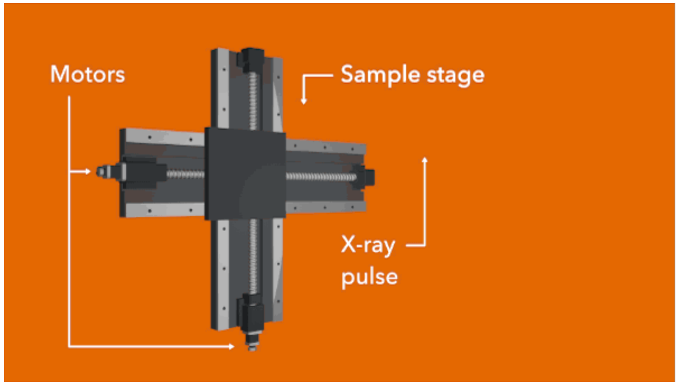
	100 nm	1 um	10 um	100 um
diamond	23	0.23	0.0023	0.000023
water	27	0.27	0.0027	0.000027
silicon	850	8.5	0.085	0.00085
iron	8700	87	0.87	0.0087
gold	20500	205	2.05	0.02

Sample replacement





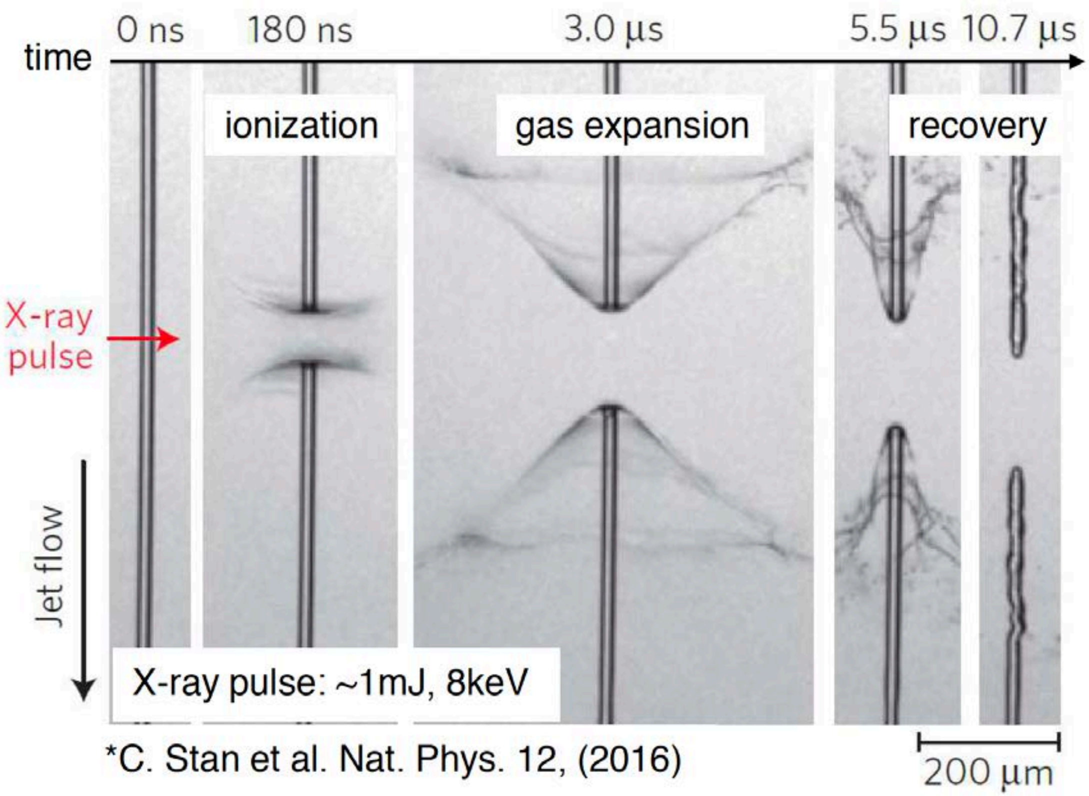




Sample replacement is relatively straightforward for liquid and gas phase samples (assuming you have lots of them), solid target via mechanical translation can be done as well.

https://www6.slac.stanford.edu/news/2015-02-02-5-ways-put-tiny-targets-front-x-ray-laser.aspx

Liquid jet recovery does take some time



*C. Stan et al. Nat. Phys. 12, (2016)

Solid material damage

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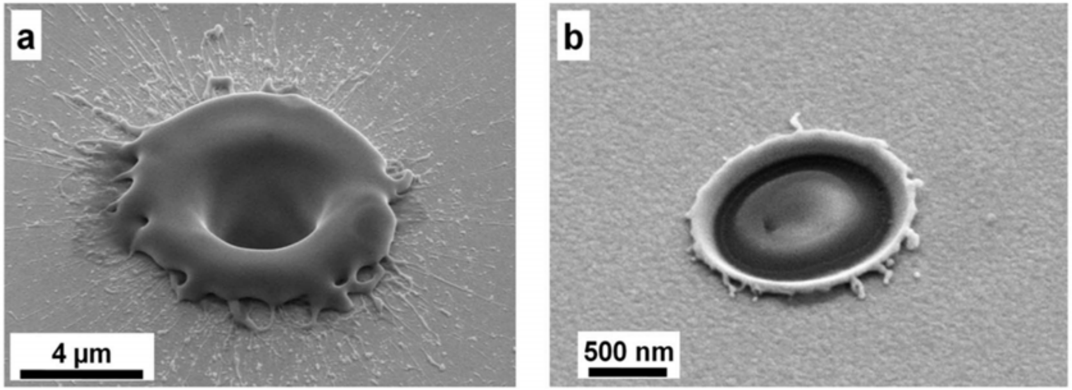


Figure 4 | Imprints created by hard XFEL pulses focused with an Ir-filled diamond FZP.

From intentional to unintentional damage, this happens regularly across the X-ray FEL facilities.

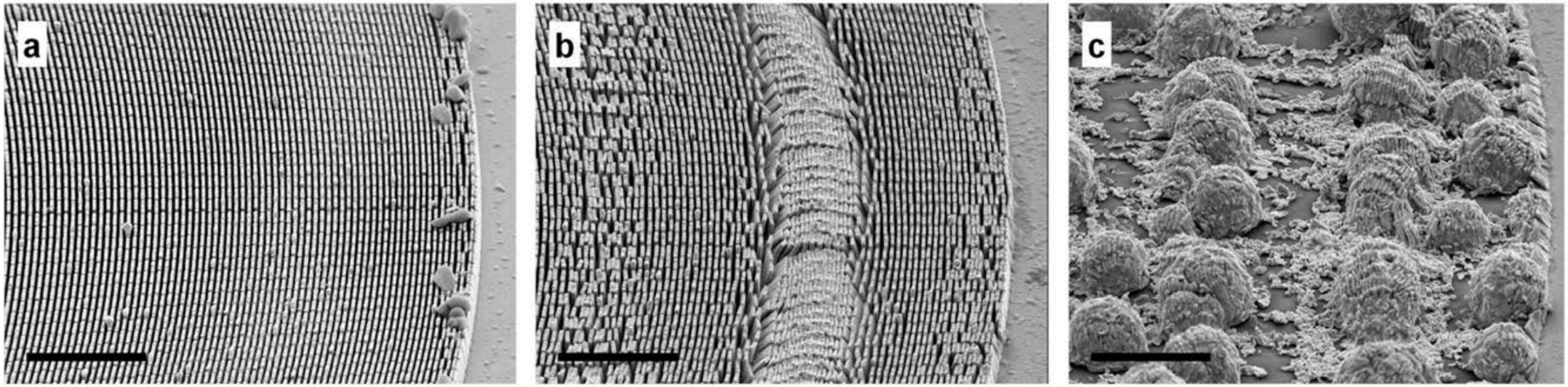


Figure 1 | Gold FZPs damaged by 8 keV LCLS pulses. SEM images of identical devices with 1 µm high structures and an outermost zone width of 100 nm. (a) no irradiation, (b) after 1,000 pulses, (c) after 10,000 pulses. Pulse power: 1.2 mJ, pulse rate: 60 Hz. The scale bars are 4 µm, the view angle is 45°.

C. David, *et al.*, Scientific Reports **1**, 57 (2011)

Instantaneous ‘heating’ and material damage

Absorbed dose (eV/atom) under pink beam: 1 mJ @ 10 keV

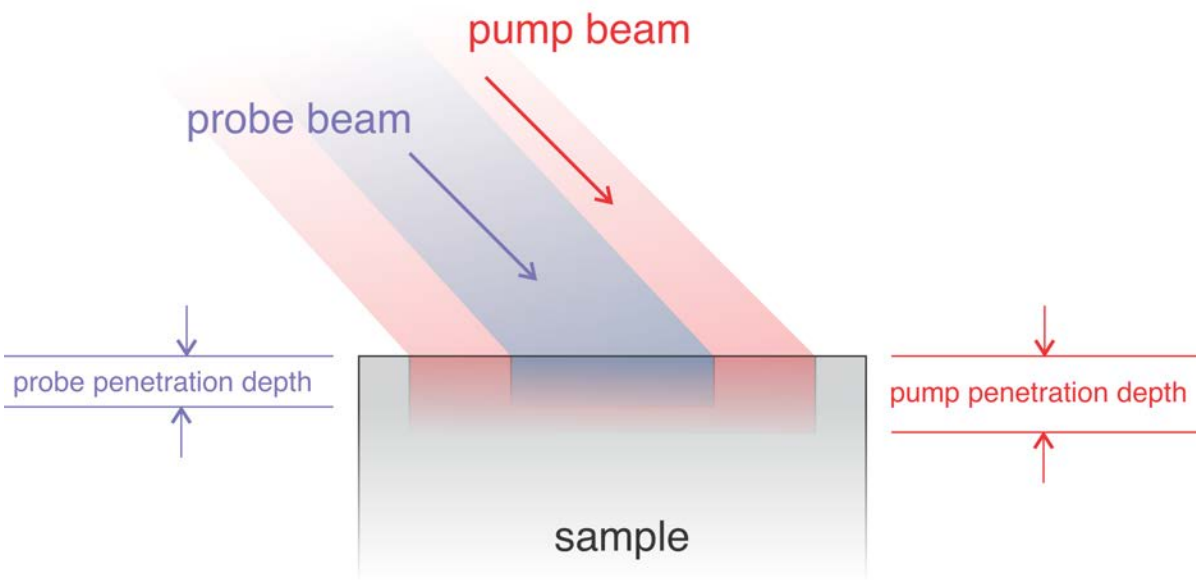
	100 nm	1 um	10 um	100 um
diamond	2300	23	0.23	0.0023
water	2700	27	0.27	0.003
silicon	85000	850	8.5	0.085
iron	870000	8700	87	0.87
gold	2050000	20500	205	2

Absorbed dose under mono beam: C(111), 0.5 eV, 0.01 mJ @ 10 keV*

	100 nm	1 um	10 um	100 um
diamond	23	0.23	0.0023	0.000023
water	27	0.27	0.0027	0.000027
silicon	850	8.5	0.085	0.00085
iron	8700	87	0.87	0.0087
gold	20500	205	2.05	0.02

A few other details

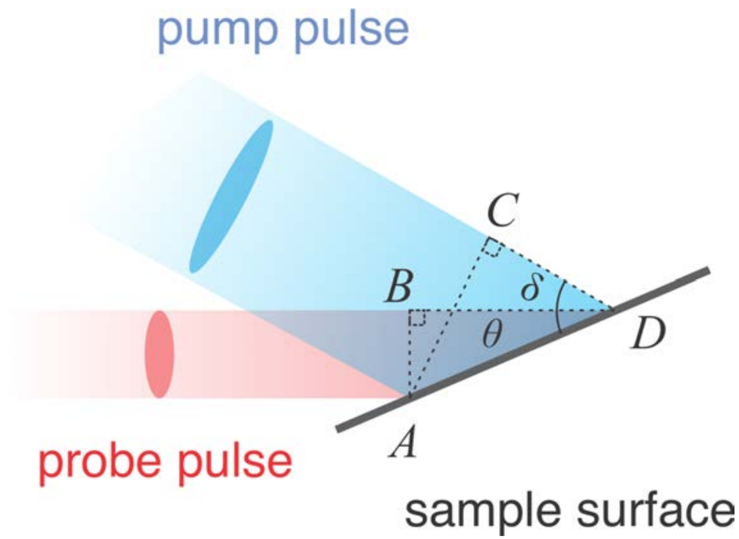
Matching of pump-probe volume



- In order to probe a ‘uniformly excited’ region, the probed-volume needed to be chosen to be smaller that the pumped-volume.
- One needs to consider both the size of the beams, and the beam penetration depth inside the samples.
- For solid bulk samples, grazing incidence/exit angle can be used to minimize x-ray penetration
- Another typical approach/solution is to use small and thin samples.

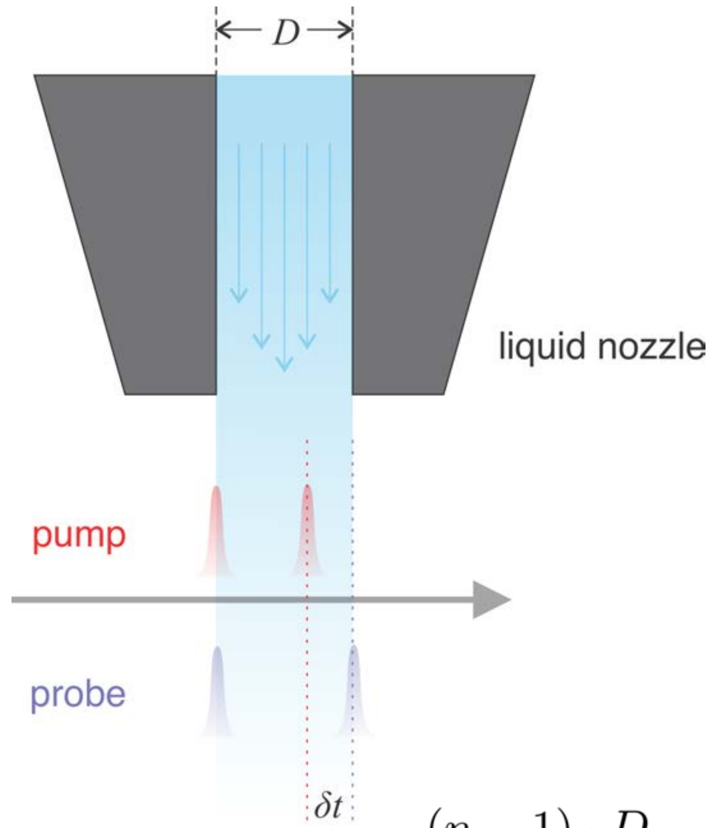
Spatial-Temporal Walk-offs

non-colinear geometry



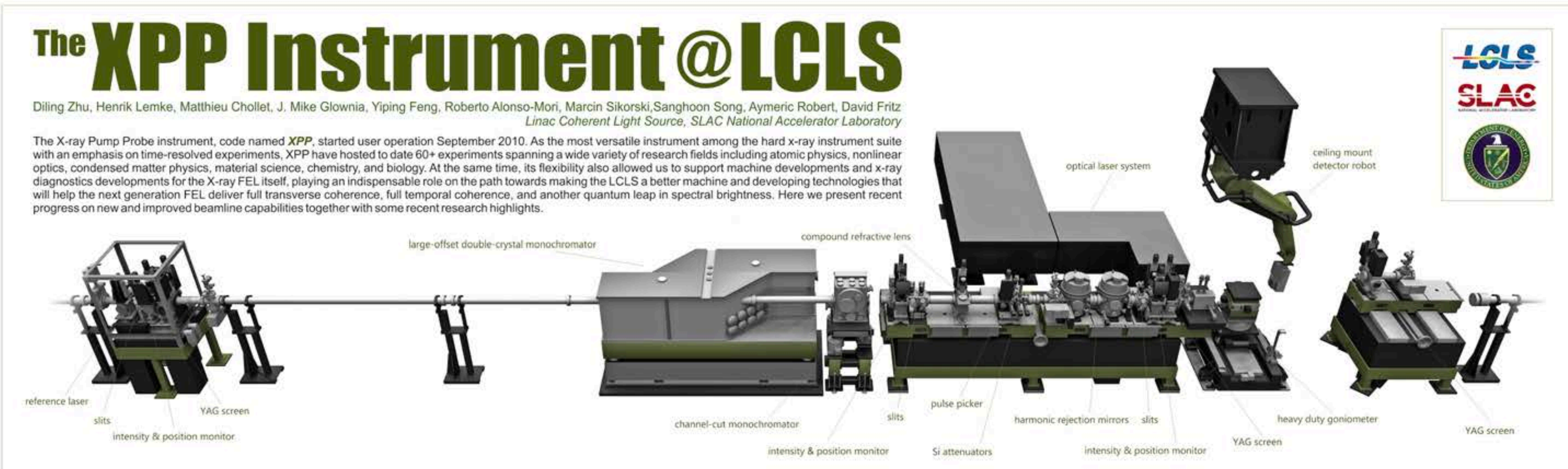
$$\frac{BD - CD}{c} = AB \cdot \frac{\cos\theta - \cos(\theta + \delta)}{c \cdot \sin\theta}$$

group delay



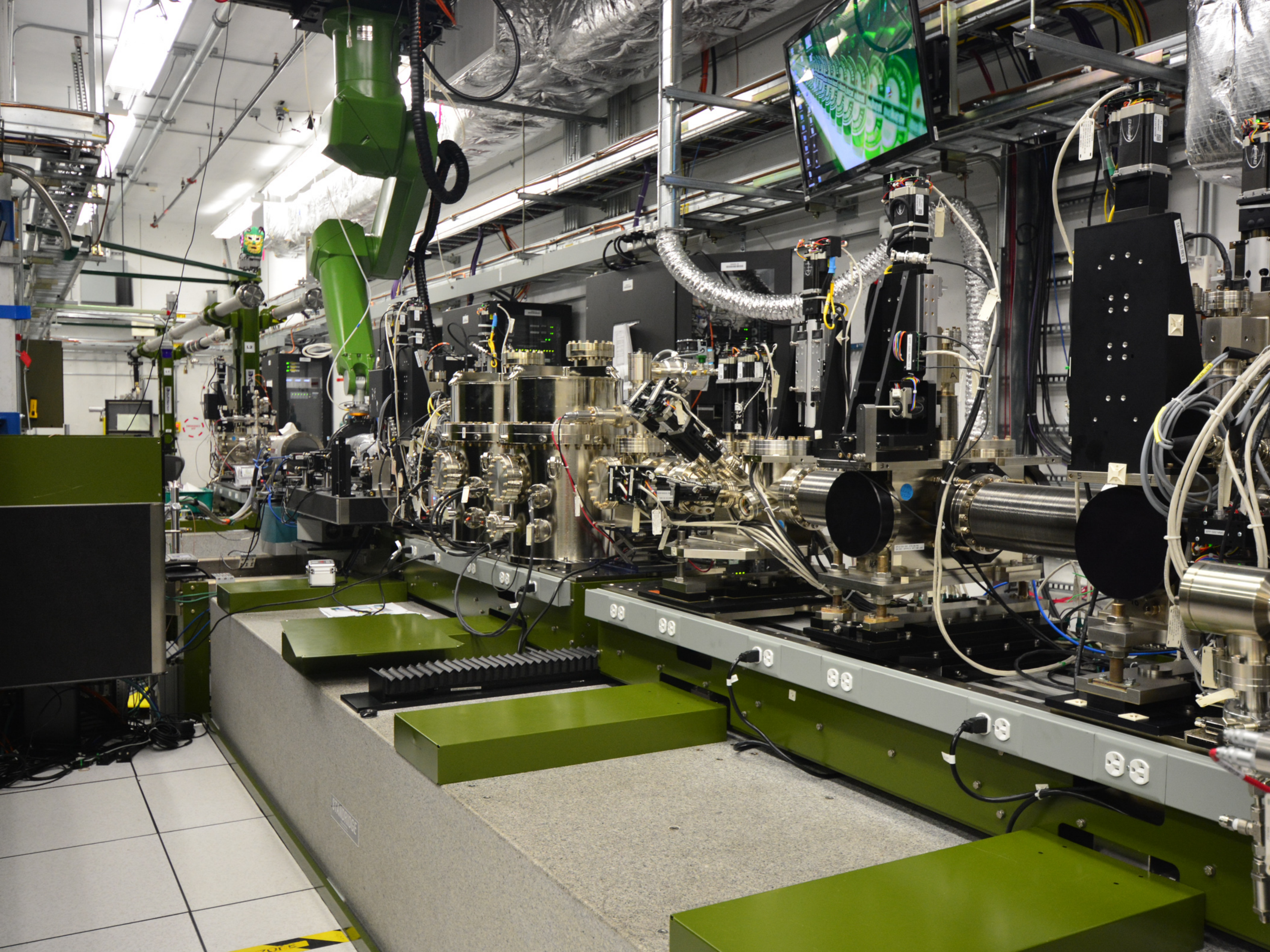
$$\frac{(n-1) \cdot D}{c}$$

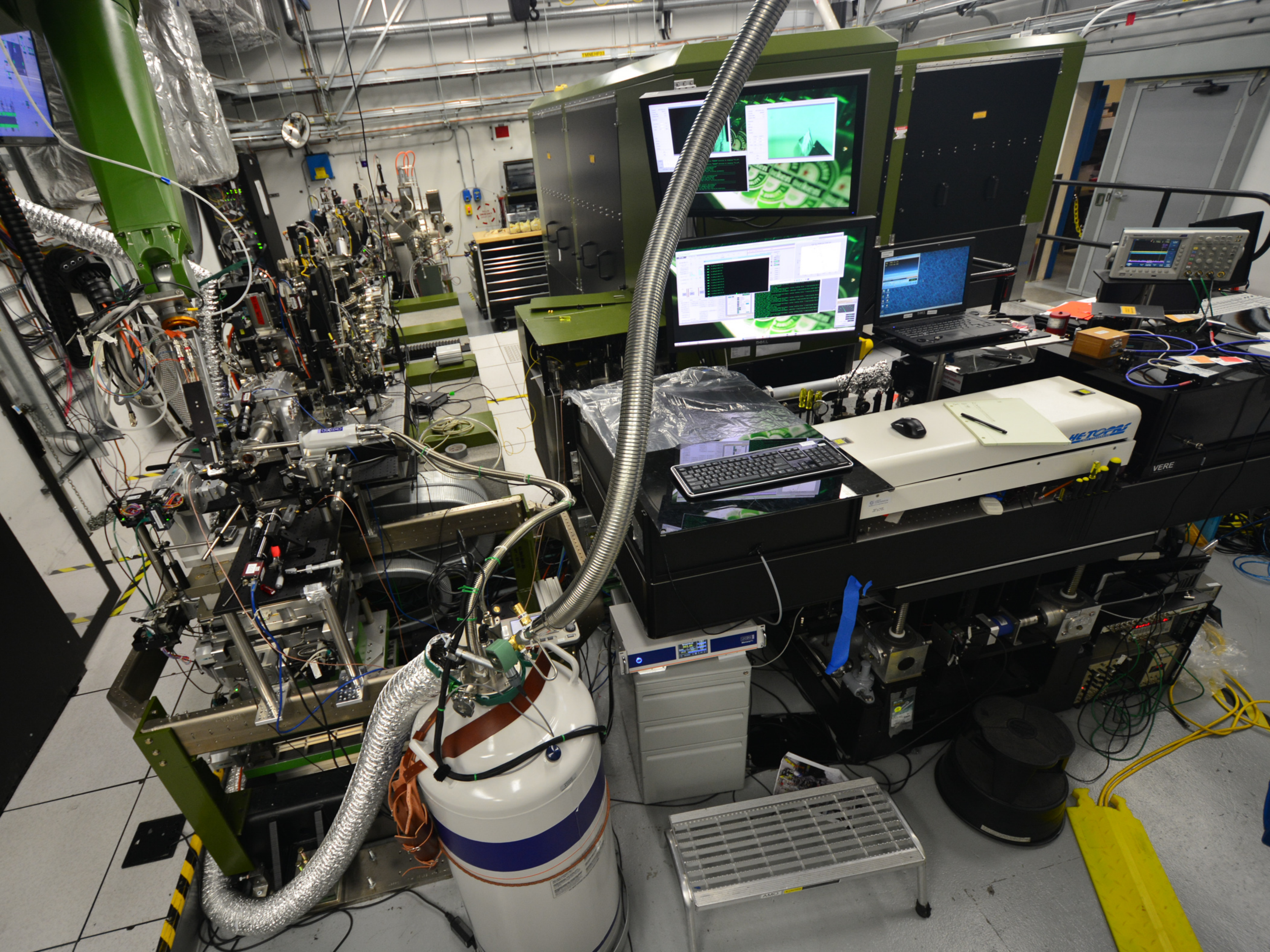
Beamline setup

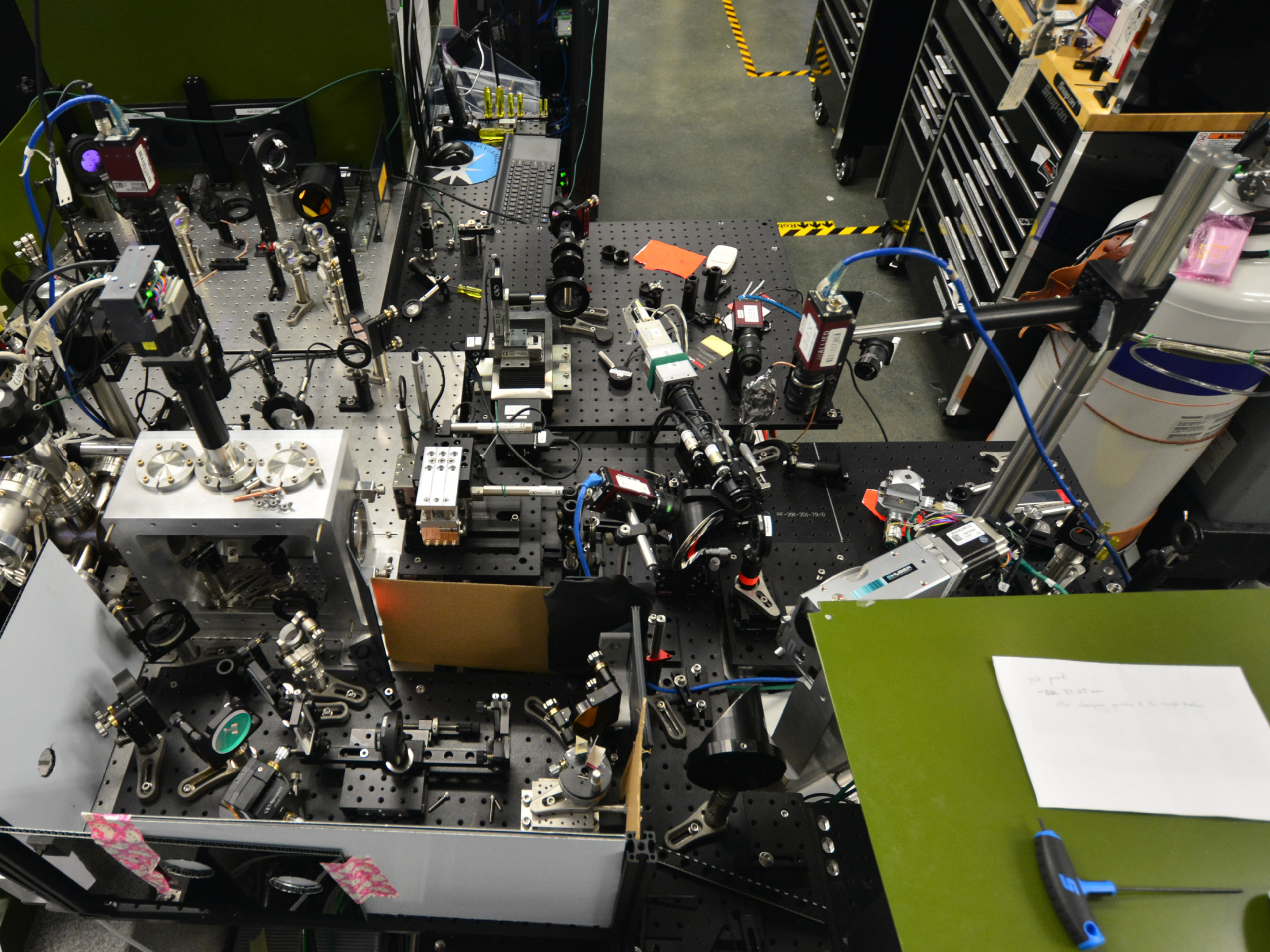


- **Optics and diagnostics to control and characterize the x-ray pulses**: monochromators, focusing optics, intensity diagnostics, profile monitors, slits, harmonic rejection mirrors, etc.
- **Optics and diagnostics to control and characterize the pump laser pulses**: compressors, OPAs, timing diagnostics, pulse duration diagnostics, beam profile diagnostics, beam stabilization system.
- **Sample environment and manipulation**: diffractometers sample chambers, cryostats and cryojets heating platforms for temperature control, liquid jets and gas jets for sample replacement.
- **X-ray Detection System**: scattering detectors, emission spectrometers, etc.


CRN #431
rayonix
LISTA
Screws
1" Pedestal Posts, Clamps, Spacers, Adapters
Spanners
Collars
1/2" Post Hardware
Samplers
Beam Splitters




VERE


Snap-on
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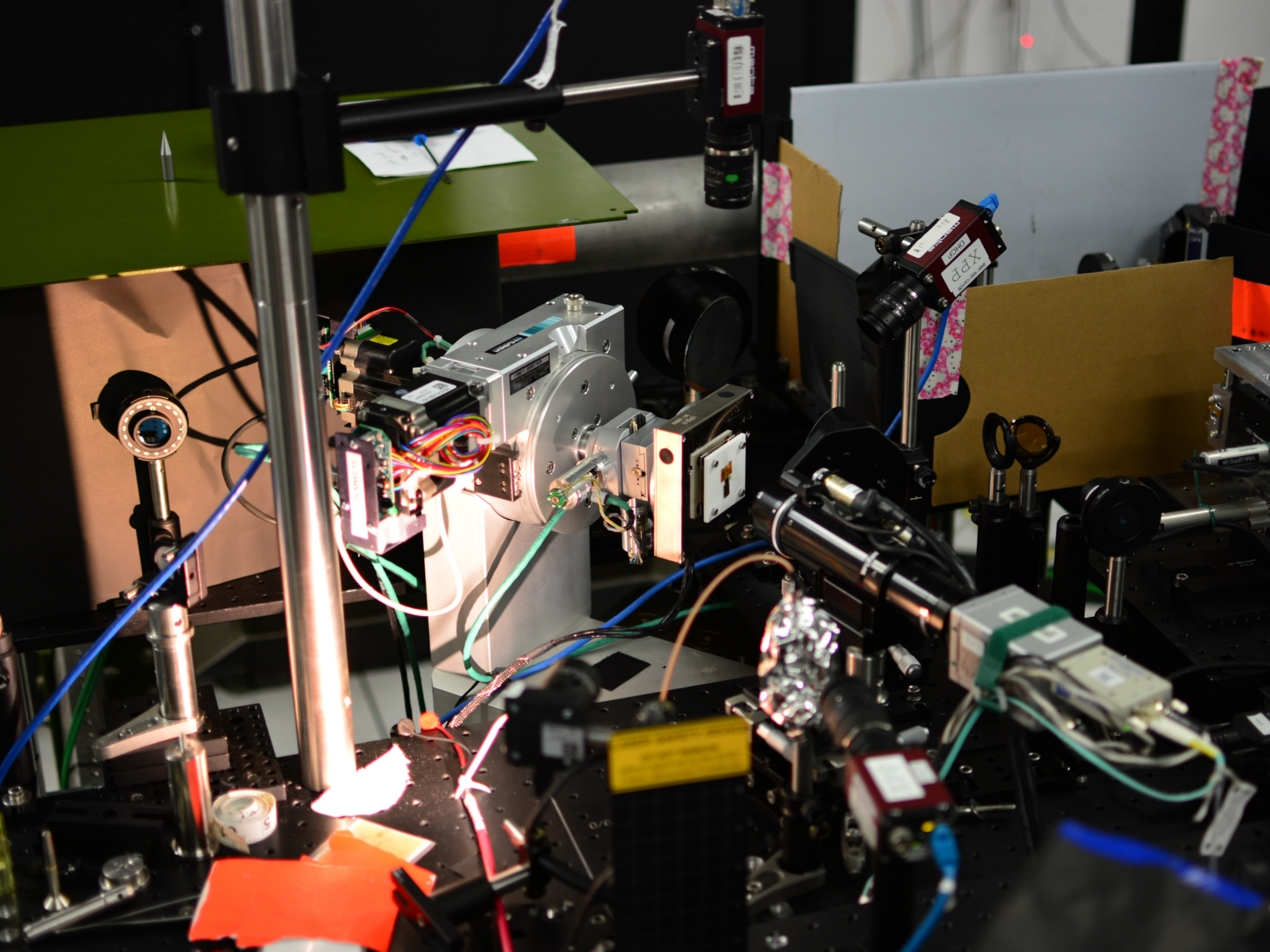

DHCP
XPD

Experiment cheatsheet

SLAC

- **X-ray beam characterization**: are we going to damage the sample? Below the damage threshold, how much signal should I expect? Or do we have enough sample to 'circulate'?
- **Laser beam characterization**: are we going to damage the sample? How much energy am I putting into the sample per unit area? Are we in the linear excitation regime?
- Are both beams hitting the sample?
- Are the two beams sufficiently overlapped in space on the sample?
- Are the two beams sufficiently overlapped in time? What time resolution do we expect with the current setup?

Let’s look at some concrete examples next

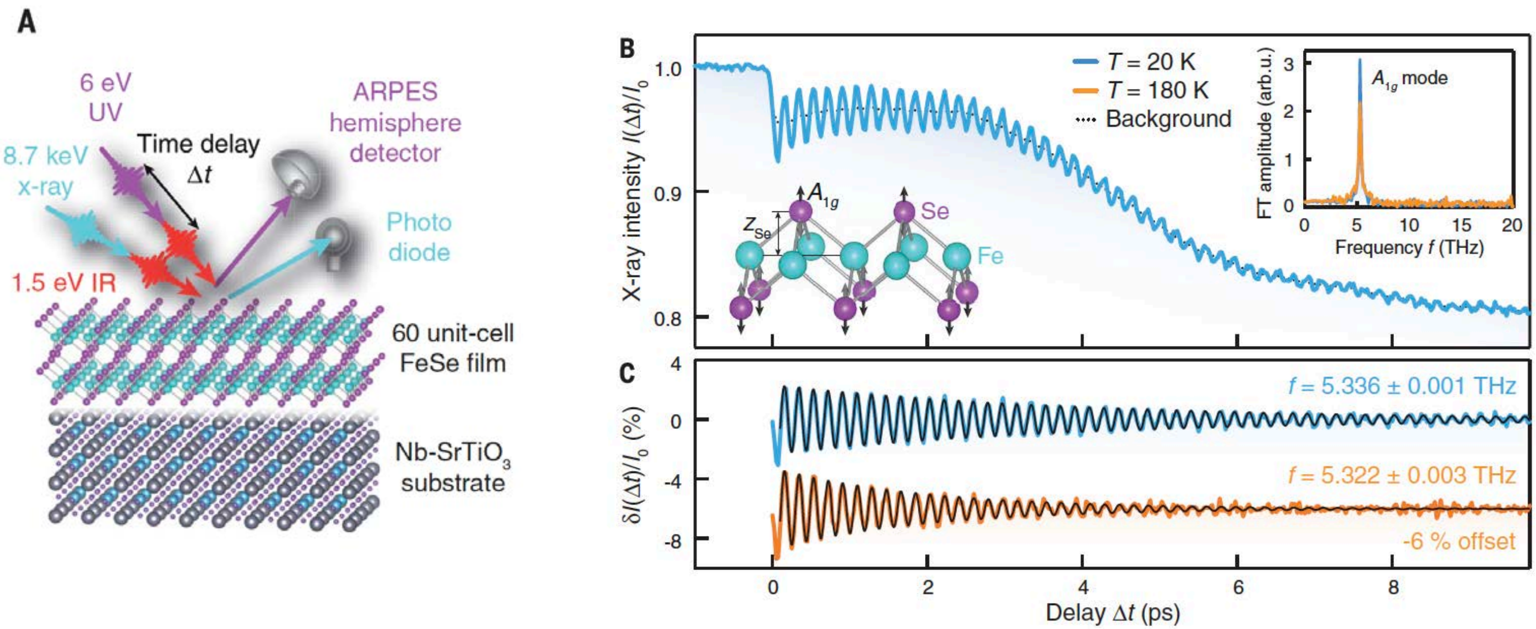
For each experiment, I’ll ask the same question:

Why do we need the x-ray FEL for the experiment?

Typical answers are:

1. Femtosecond time resolution
2. Spatial coherence
3. Pulse energy
4. To mitigate sample damage
5. To capture ‘rare’, and/or ‘transient’ state with the highest quality.

Example: Electron phonon coupling in FeSe



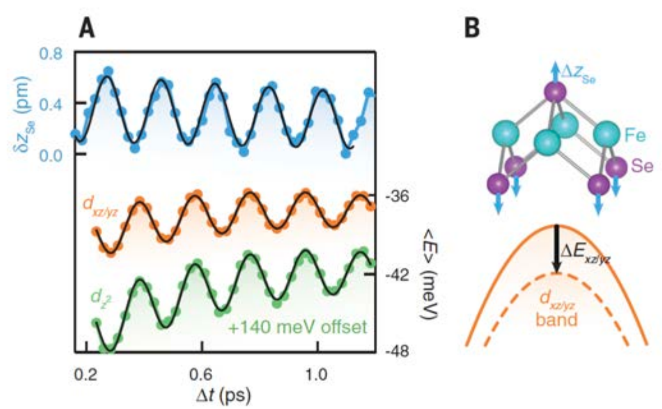
Pump: 40 fs 800 nm IR pulses

Probe: 8.7 keV, attenuated mono beam

Sample: FeSe thin film on Nb-STO substrate

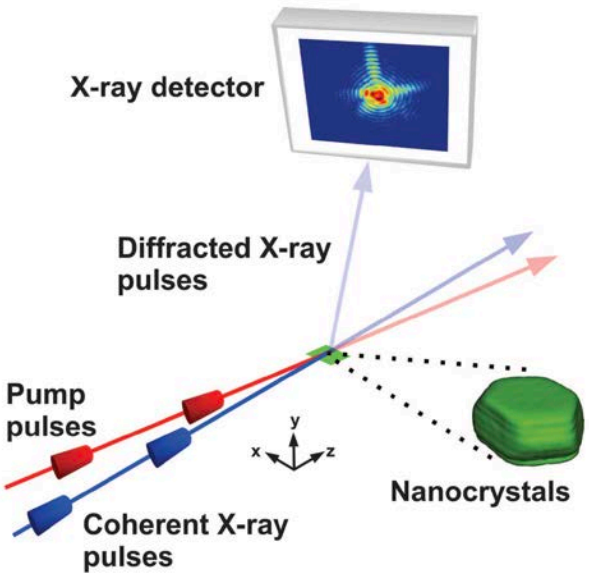
Action: coherent lattice motion and associated electronic structure variation.

Method: Bragg diffraction and ARPES



S. Gerber, *et al.*, Science **357**, 71 (2017)

Example: Acoustic modes in nanocrystal Au



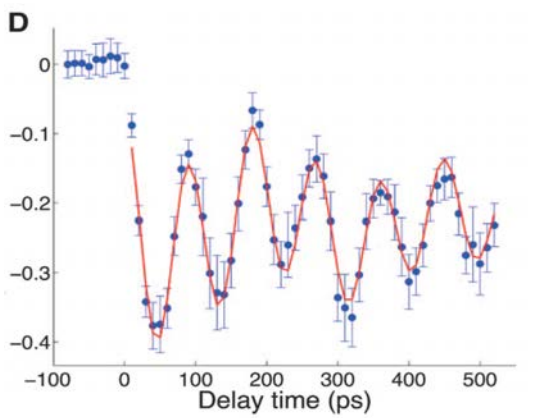
Pump: 50 fs 800 nm IR pulses

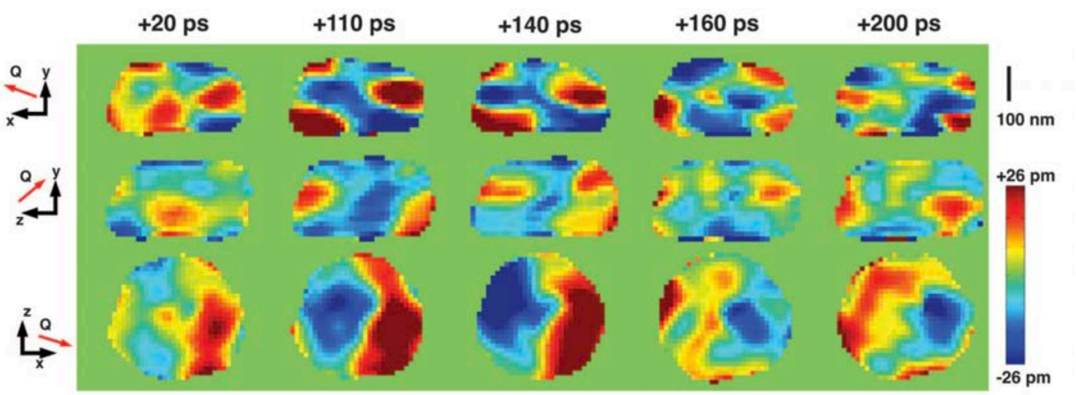
Probe: 9.2 keV, attenuated mono beam

Sample: A single Au nanoparticle

Action: nanoscale thermal acoustic responses within a single nanoparticle

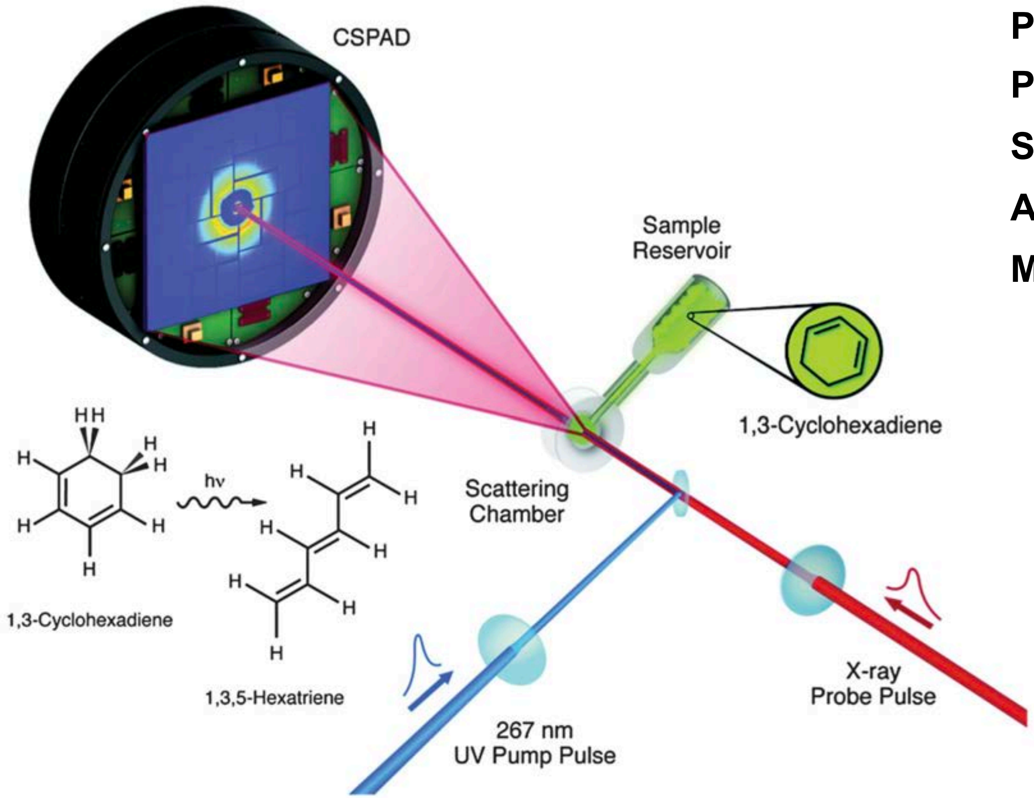
Method: Bragg coherent diffraction imaging (BCDI).





J.N. Clarke, *et al.*, Science **341**, 56 (2013)

Example: UV induced ring opening reaction



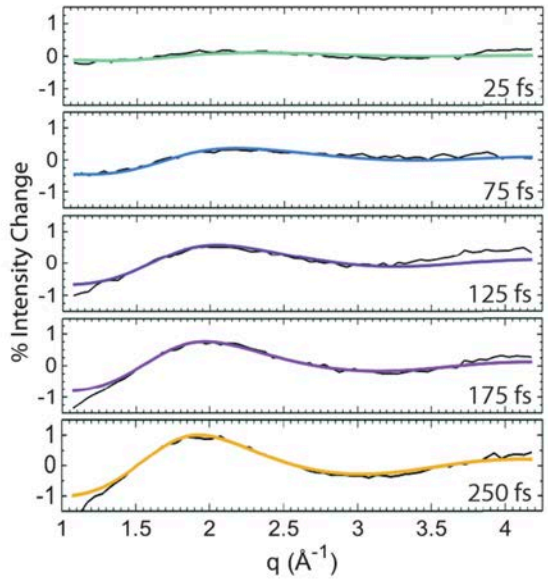
Pump: 50 fs 267 nm UV pulses

Probe: 8.3 keV pink beam

Sample: Cyclohexadiene in a gas cell

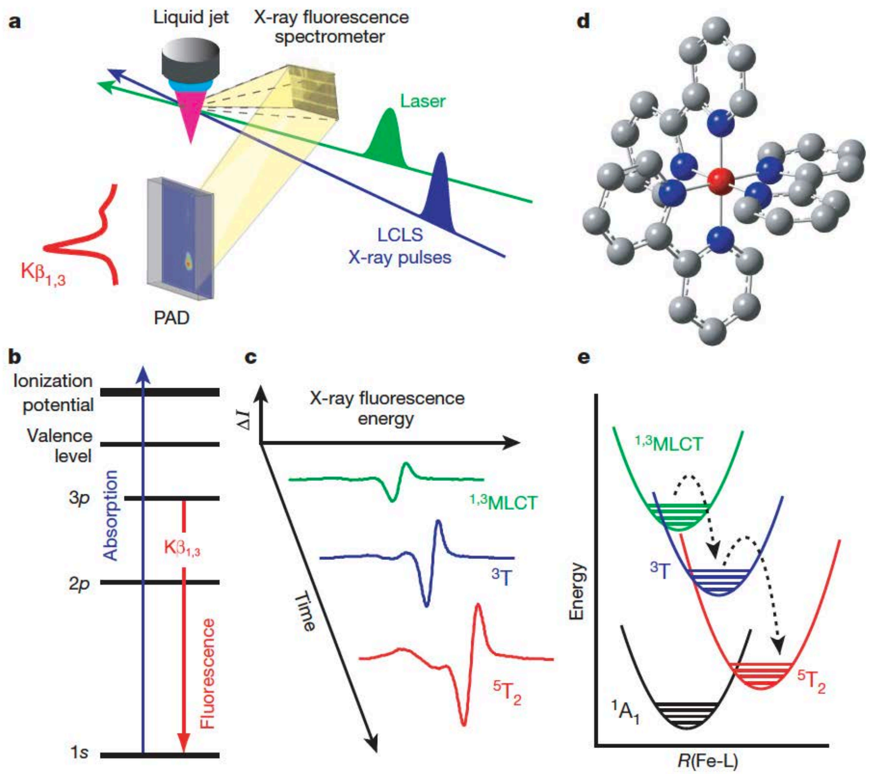
Action: bond breaking and ring opening

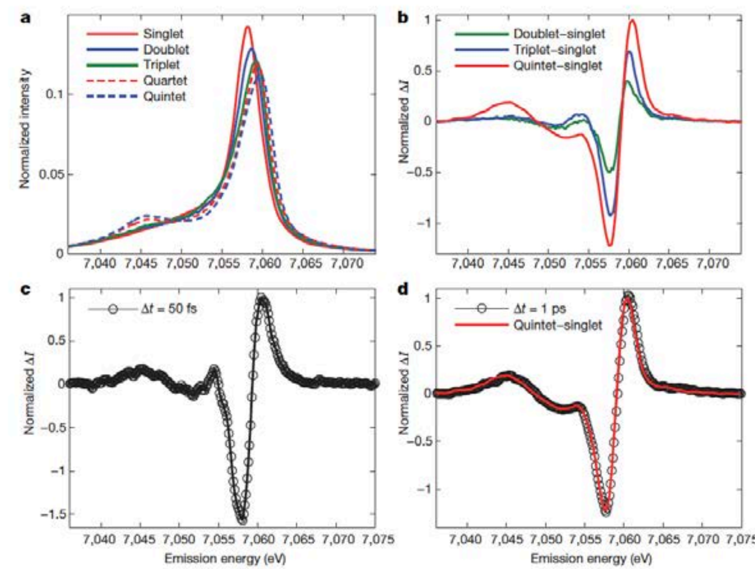
Method: X-ray wide angle scattering



M. Minitti, *et al.*, PRL **114**, 255501 (2015)

Example: Electron & spin relaxation in molecules





Pump: 70 fs, 520 nm

Probe: 8.0 keV, pink beam

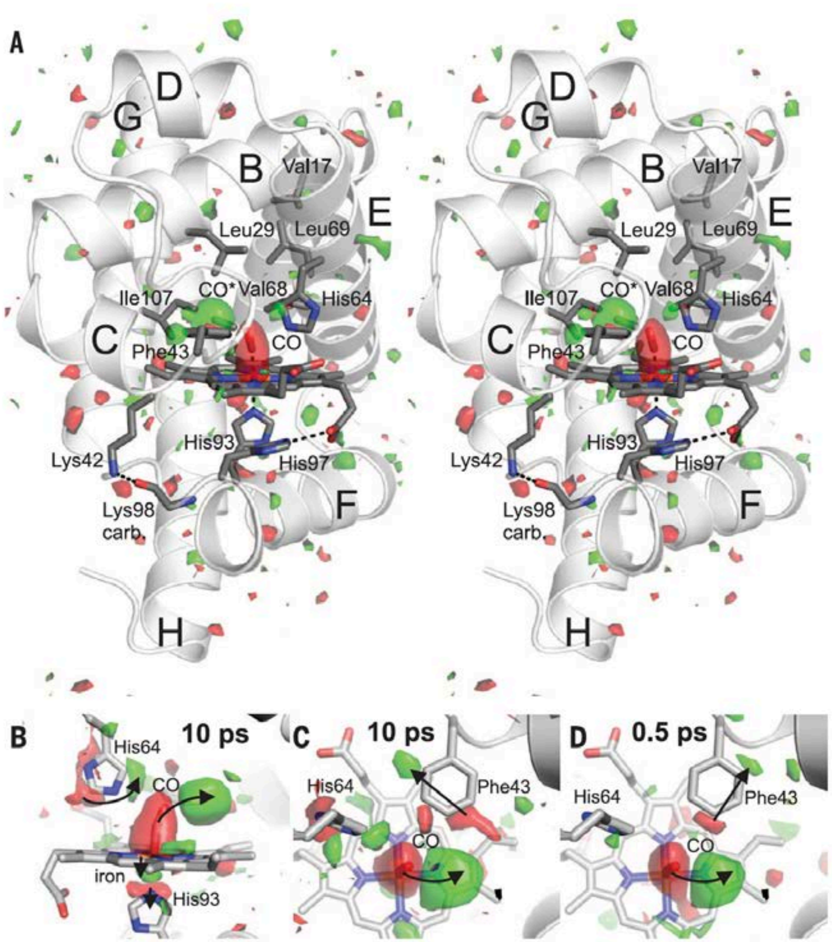
Sample: polypyridyl iron complexes

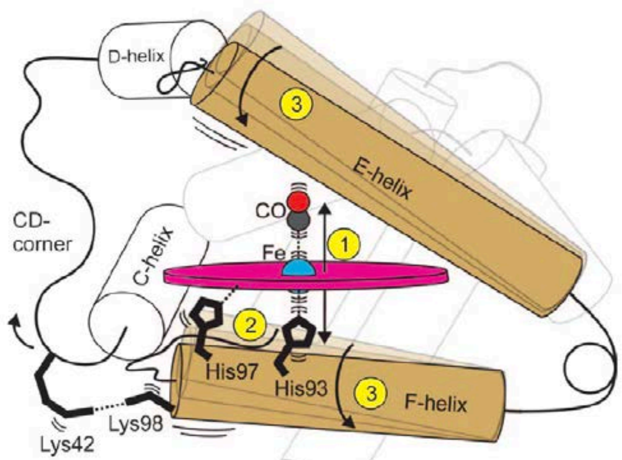
Action: metal to ligand charge transfer

Method: X-ray emission spectroscopy

W. Zhang, *et al.*, Nature **509**, 346 (2014)

Example: Protein structural change





Pump: 100 fs 532 nm pulses

Probe: 6.7 to 6.9 keV, pink beam

Sample: horse heart Myoglobin-CO microcrystal

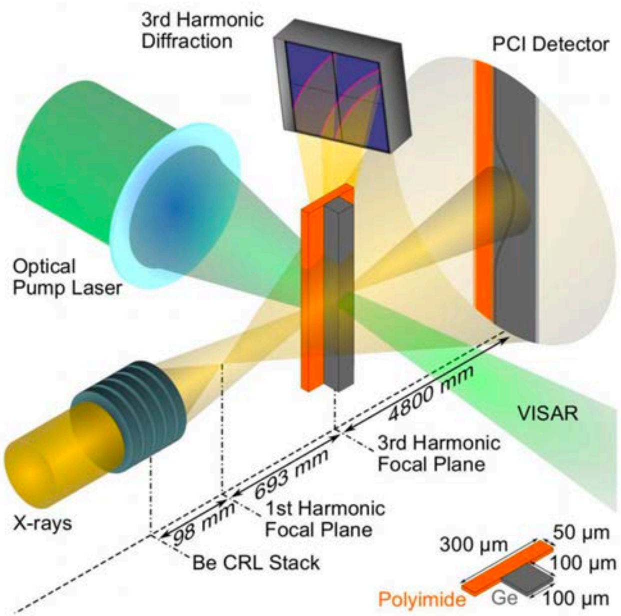
Action: structural changes following the photolysis of the Fe-CO bond.

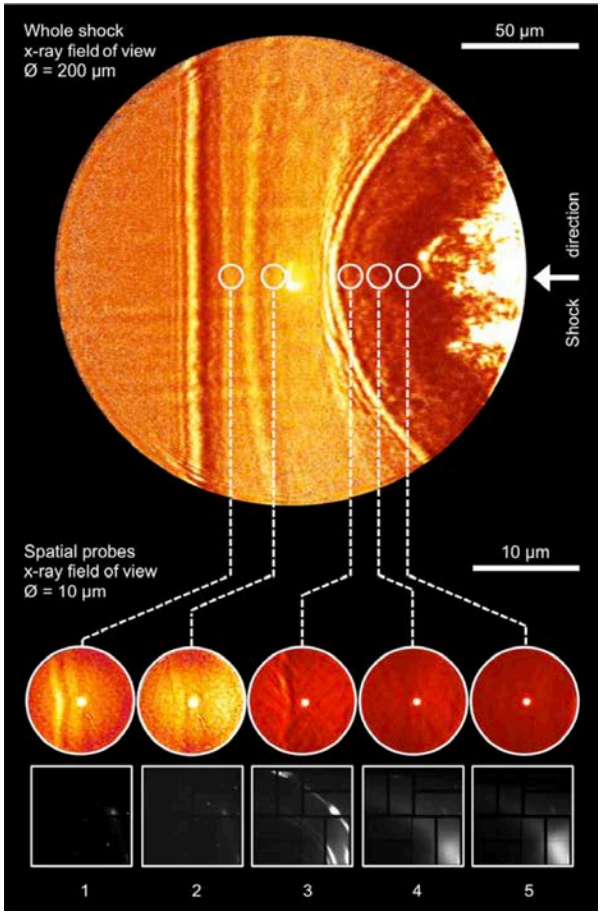
Method: X-ray protein crystallography

T. Barends, *et al.*, Science **350**, 445 (2015)

A few more not-so-fast examples

Example: Material structure under shock wave





Pump: 15 ns 527 nm, 2J

Probe: 8.2 and 24.6 keV pink beams

Sample: single crystal silicon

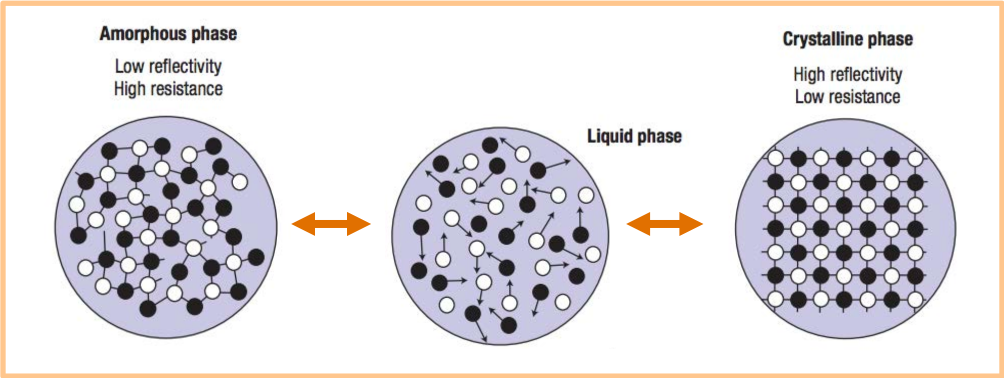
Action: elastic-inelastic response under shock compression

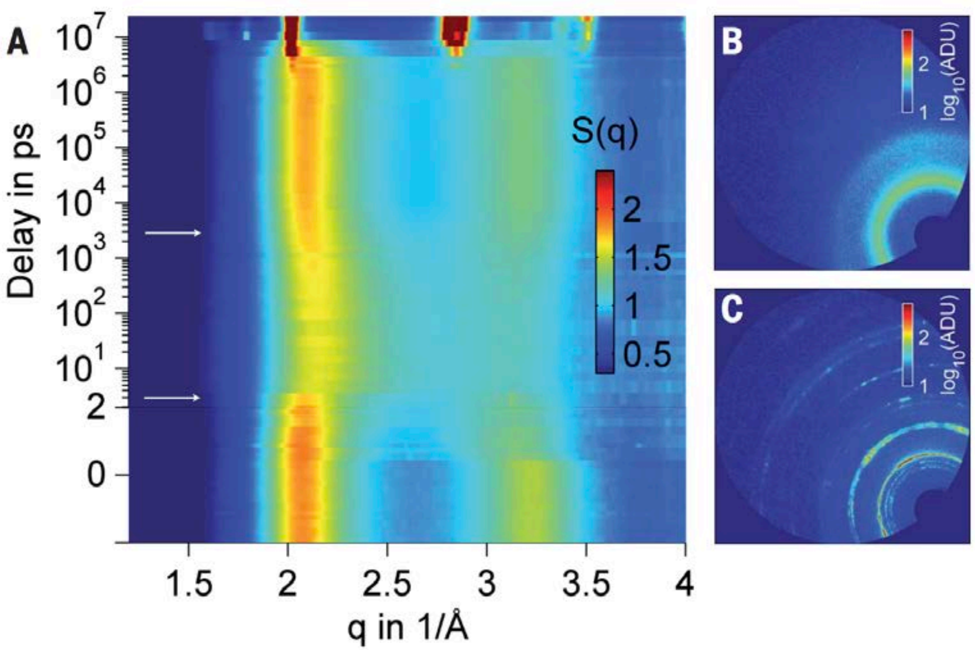
Method: phase contrast imaging, wide angle scattering, powder diffraction

F. Seiboth, *et al.*, APL **112**, 221907 (2019)
S. Brown, *et al.*, Science Advances **5**, 8044 (2019)

Example: Phase change material

The inner working of the materials behind CD, DVD, and Blueray discs.





Pump: 50 fs 800 nm IR pulses

Probe: 9.5 keV pink beam

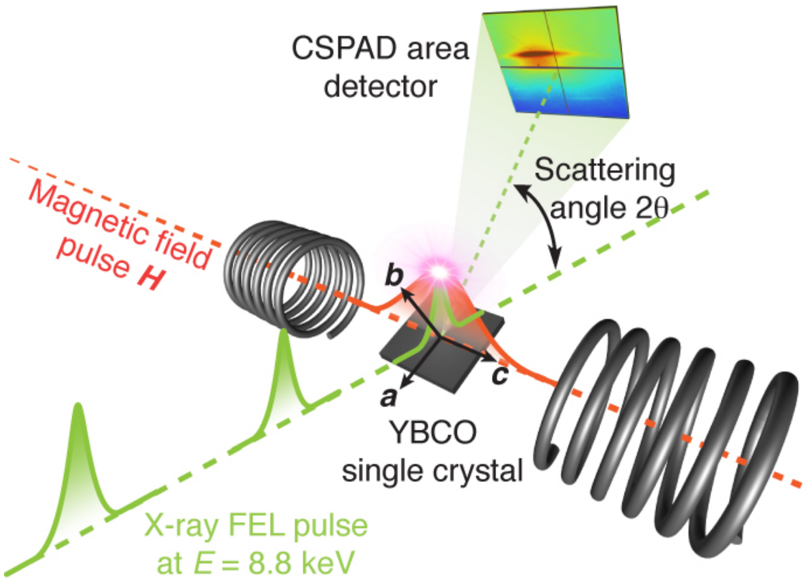
Sample: GaSbTe thin film

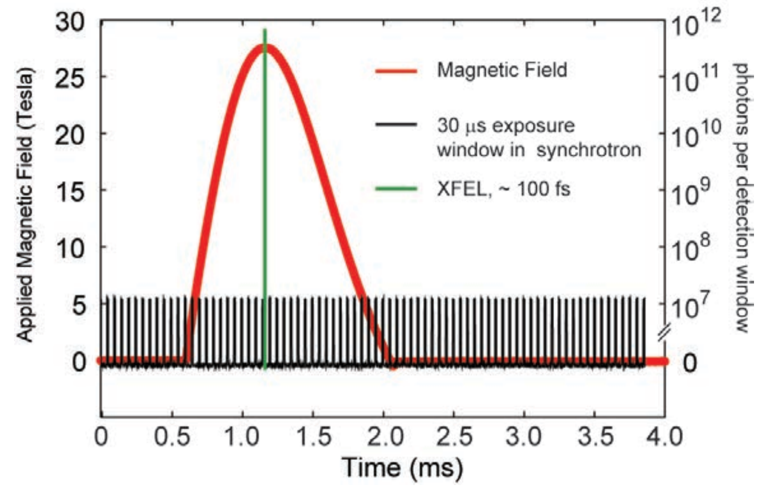
Action: amorphous to liquid to crystalline structural transition

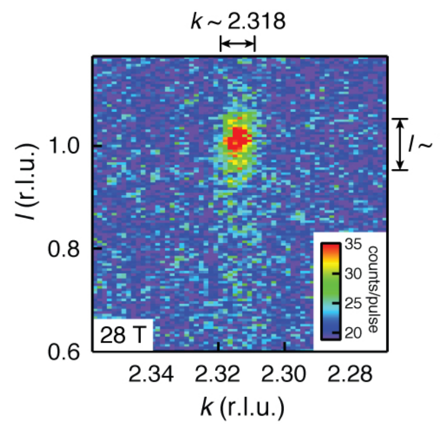
Method: X-ray wide angle scattering, powder diffraction.

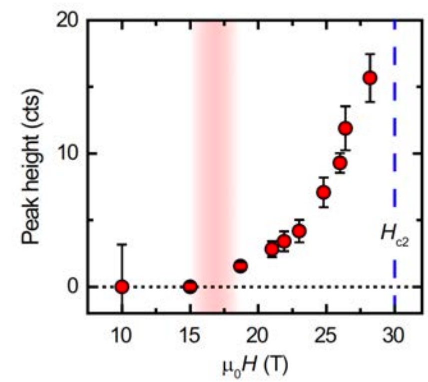
M. Wuttig, et al., Nat. Materials **6**, 824 (2007)
P. Zalden, *et al.*, Science **364**, 1062 (2019)

Example: YBCO in pulsed high magnetic field









Pump: 1 ms, 30+ Tesla magnetic field pulse

Probe: 8.9 keV pink beam

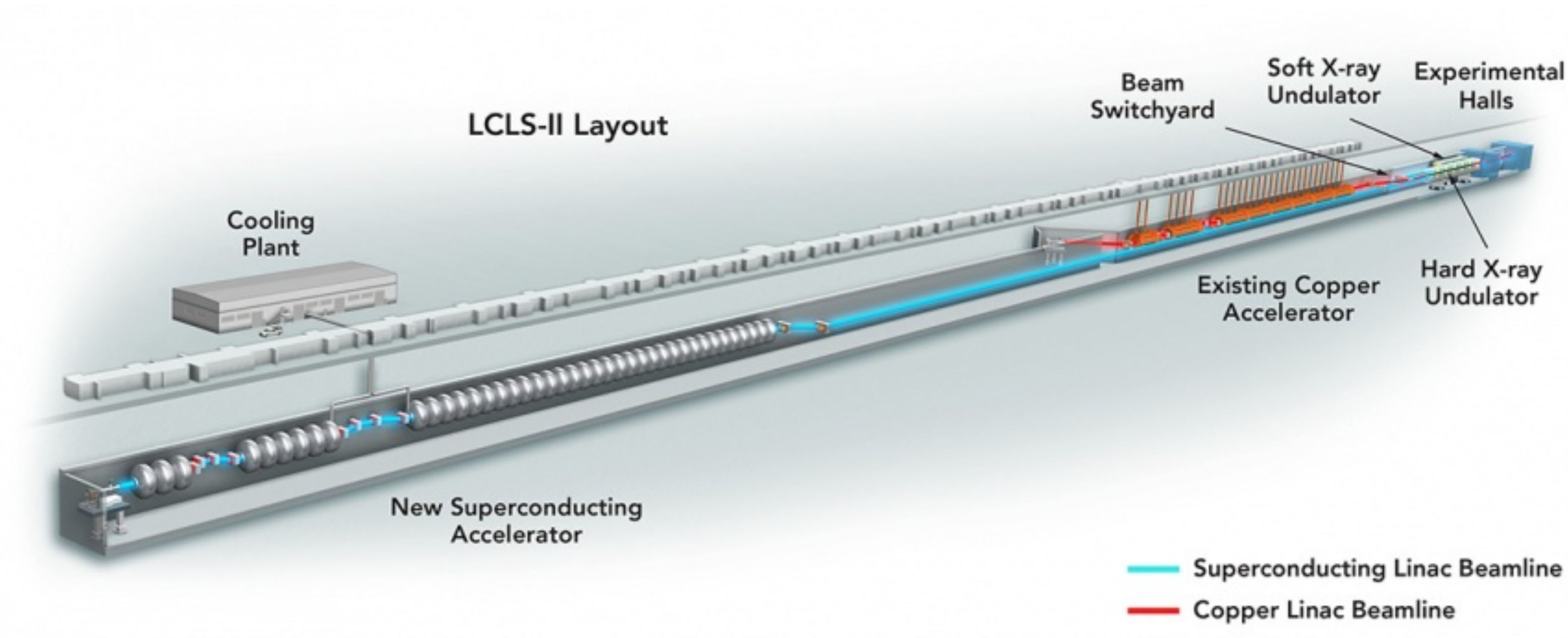
Sample: single crystal YBCO

Action: appearance of a 3D charge density wave order when super conductivity is suppressed by the fied.

Method: single crystal diffraction

S. Gerber, *et al.*, Science **350**, 949 (2015)
H. Jang, et al., PNAS **113**, 14645 (2016)

LCLS-II & HE: prospects and challenges



The new FEL will be driven by a superconducting linear accelerator operating at ~1MHz compared to the current 120Hz machine. Is everything going to be 10,000 times better?

Q&A, Discussion

Adding time axis to various x-ray techniques

SLAC

What has been done so far?

X-ray Techniques	Status
Imaging, microscopy (scanning, full field)	A little
Tomography	No
Absorption spectroscopy (XANES, EXAFS)	Yes
Coherent diffractive imaging	Yes
Small & wide angle scattering	Yes
Powder diffraction	Yes
Emission spectroscopy, fluorescence	Yes
Diffraction, crystallography	Yes
Surface scattering	A little
inelastic scattering (resonant, non-resonant, Raman)	Yes
What's msissing?	

A few take home messages

- X-ray free electron lasers are for now the brightest sub-nanometer wavelength ‘flash light’ that you may get your hands on.
- Pump-probe methodology using X-ray FEL gives the x-ray characterization methods you are familiar with a new dimension: femtosecond time axis.
- The key to make an experiment work is spatial temporal overlap.
- The short intense pulse can be used to freeze and capture other transient phenomena too, to overcome radiation damage, or to maximize ‘image’ quality of relatively rare events.
- Compare to the history of synchrotron experiments (~10^4 beamline-year), the field of x-ray FEL is still in its infancy, at about ~20 beamline-year. There are plenty of unchartered territories that are still waiting to be explored.