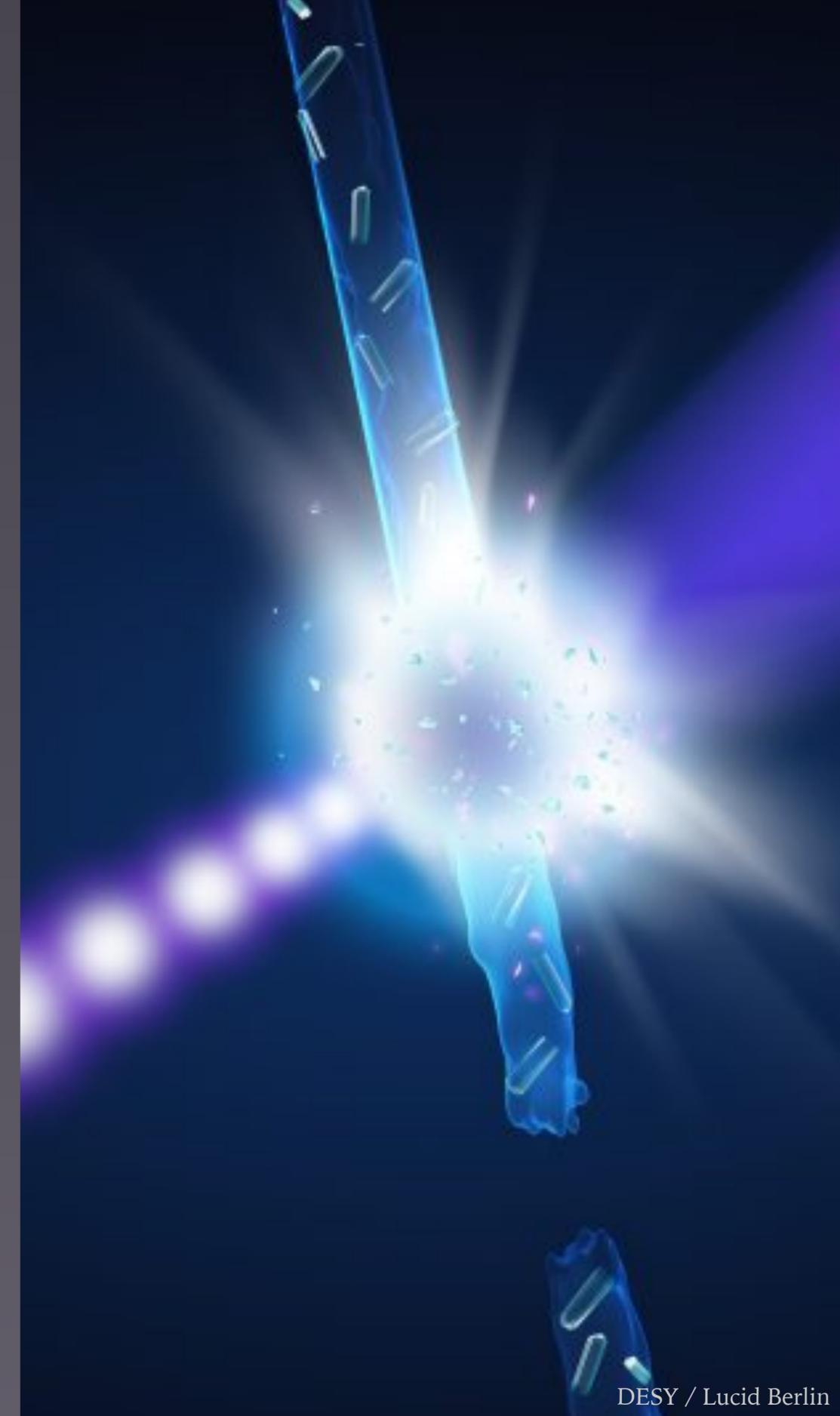


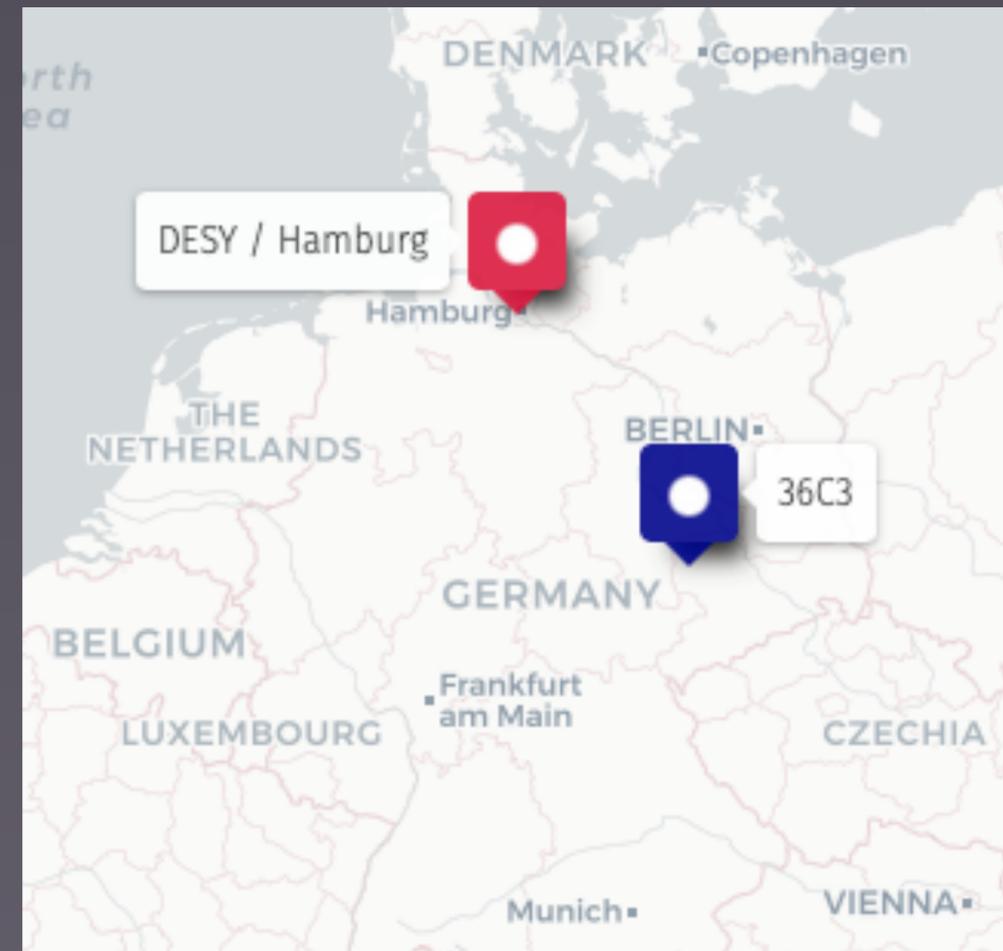
AN ULTRASHORT HISTORY OF ULTRAFAST IMAGING

Caroline Arnold



About Me

- Physicist at Deutsches Elektronen-Synchrotron DESY
- PhD in ultrafast molecular physics
- Pronouns: she/her



What Is the Goal of Ultrafast Imaging?



Long exposure time: blurry image

1 s



Short exposure time: clear image

1/100 s

Take clear pictures of moving objects

Movies

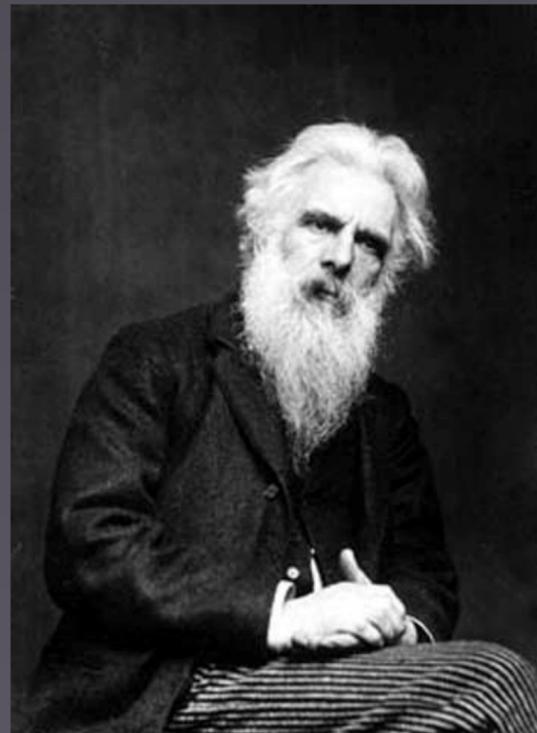


- Most processes in nature are not static
- More insight from time-resolved images

The First Ultrafast Movie

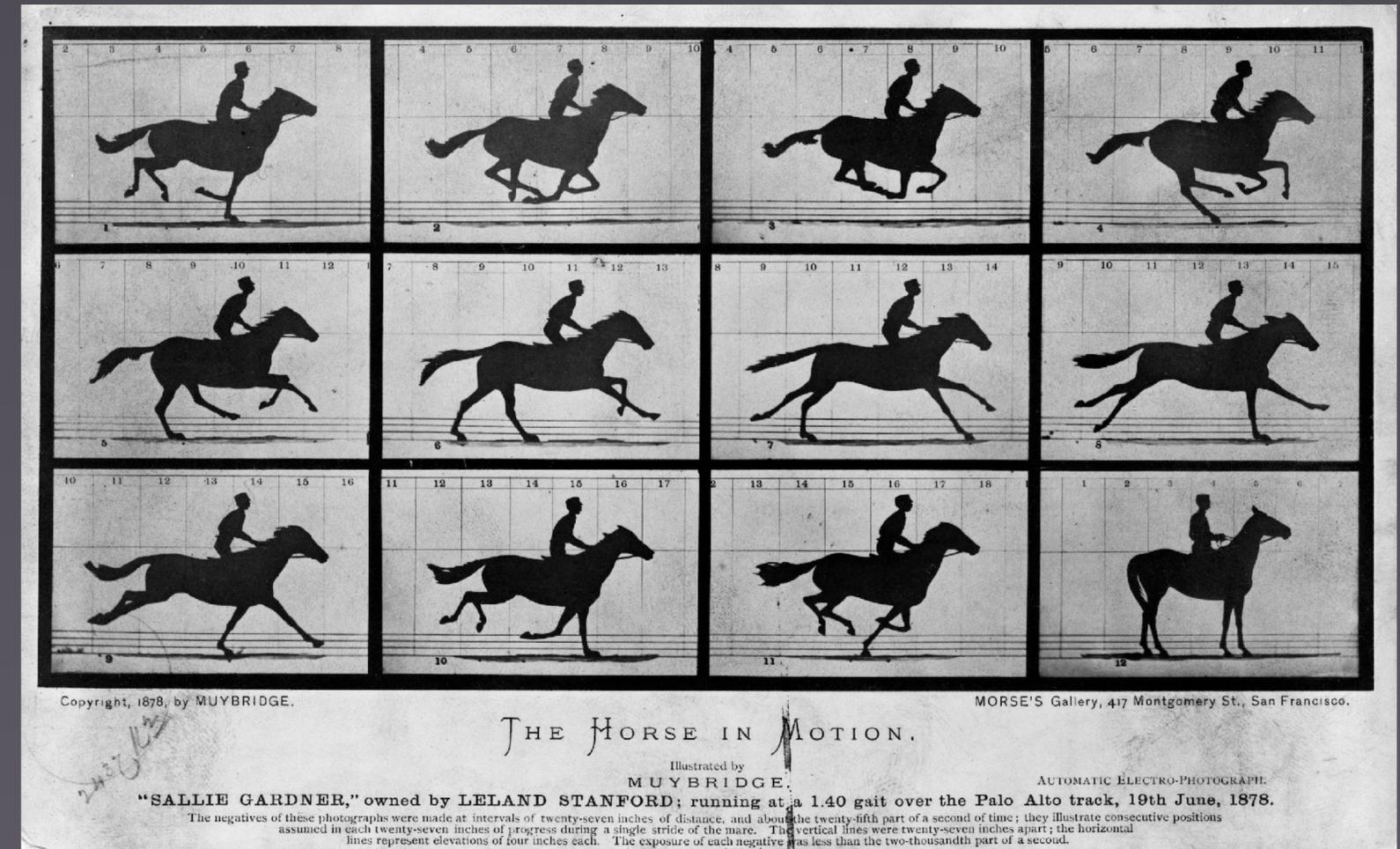
Does a galloping horse ever lift all its legs off the ground?

Eadweard Muybridge (1830 - 1904)



Public Domain, Wikimedia Commons 190376

► Stop-motion technique

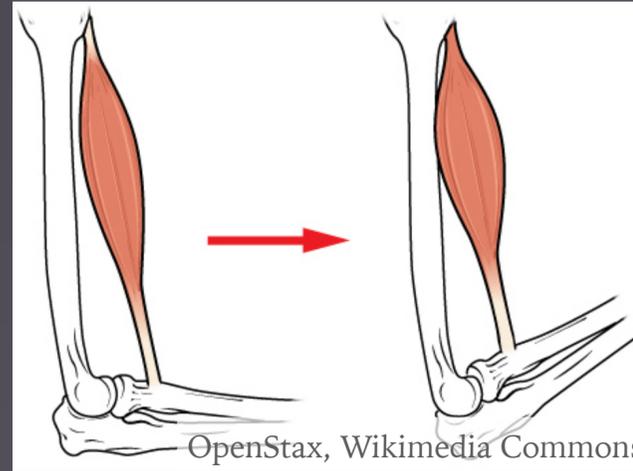
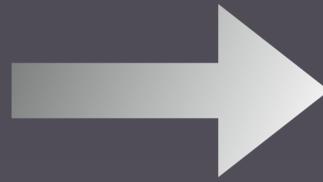


The Horse in Motion, Stanford 1878

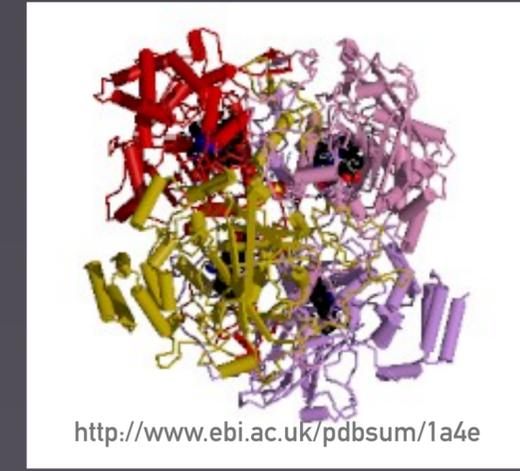
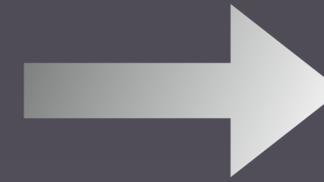
What Is Ultrafast?



second
 1 s



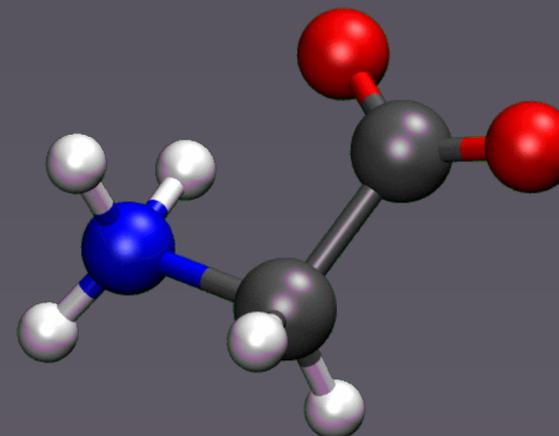
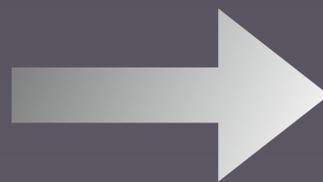
millisecond
 $1\text{ ms} = 0.001\text{ s} = 10^{-3}\text{ s}$



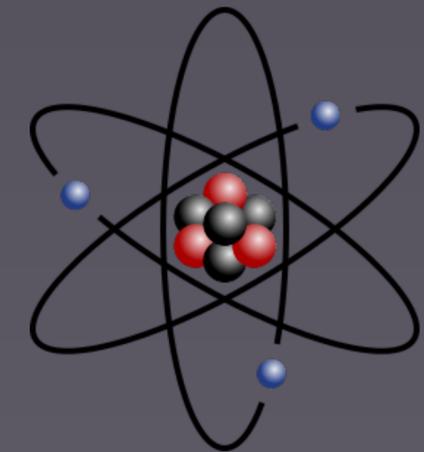
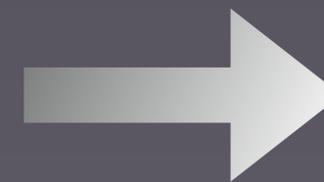
microsecond
 $1\text{ }\mu\text{s} = 0.000001\text{ s} = 10^{-6}\text{ s}$



nanosecond
 $1\text{ ns} = 10^{-9}\text{ s}$



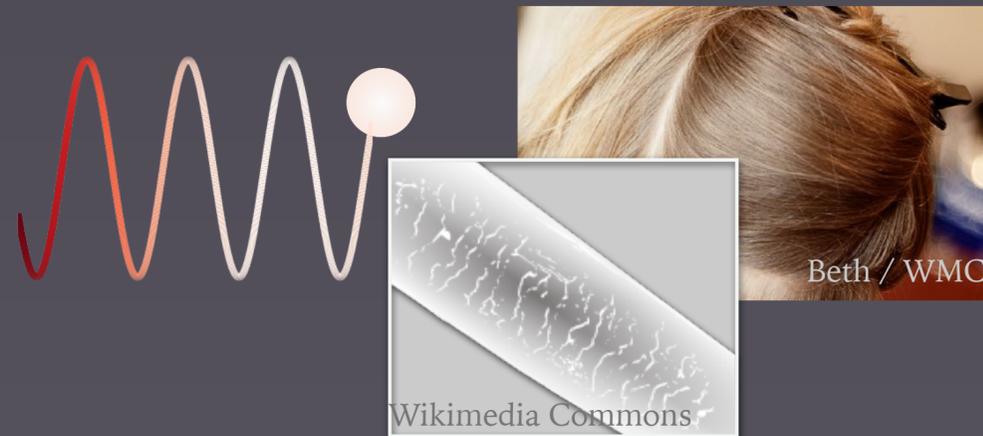
femtosecond
 $1\text{ fs} = 0.0000000000000001\text{ s} = 10^{-15}\text{ s}$



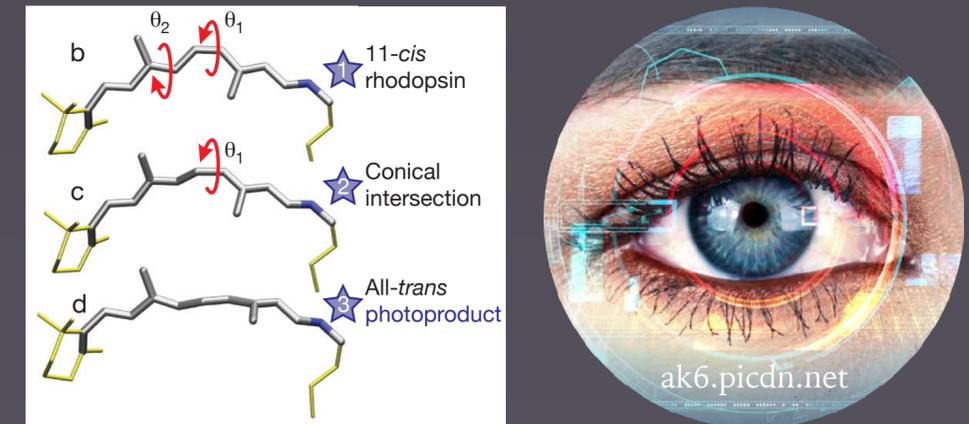
attosecond
 $1\text{ as} = 10^{-18}\text{ s}$

The Femtosecond Time Scale

Light crosses hair in 30 fs



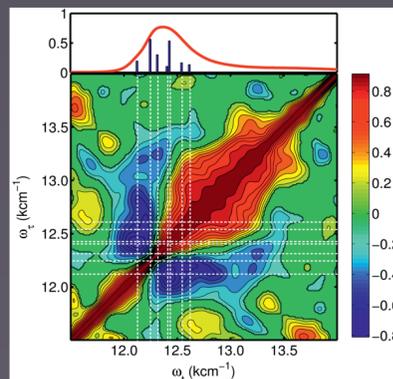
First steps of vision in 200 fs



Polli et al, Nature 467 (2010) Polli et al, Nature 467, 440 (2010)



Photosynthesis sets off in 100 fs



Duan et al, PNAS 114 (2017)



Duan et al, PNAS 114, 8111 (2017)

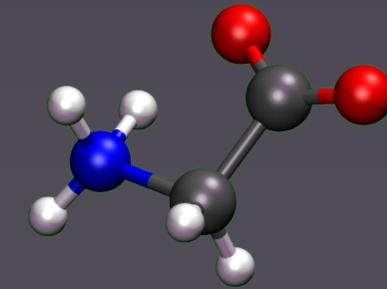


Atomic to
nanometer scale

Time Resolution



Choose the right exposure time



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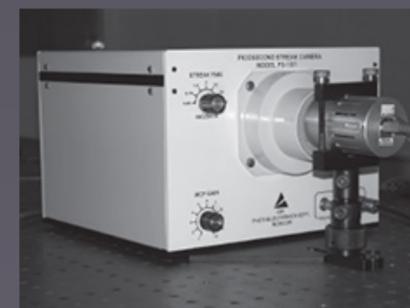
Petr Novak, Wikipedia

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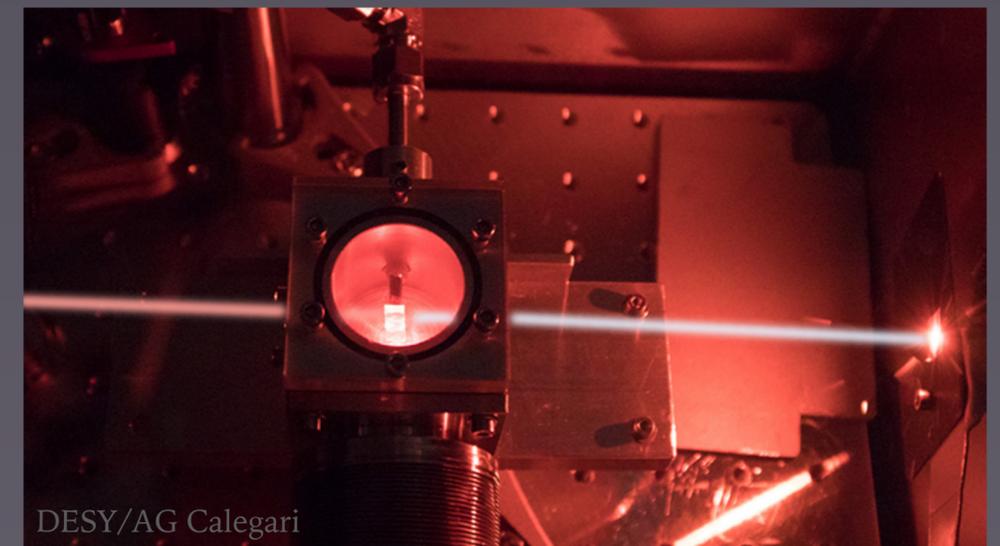
Dmitry Makeev

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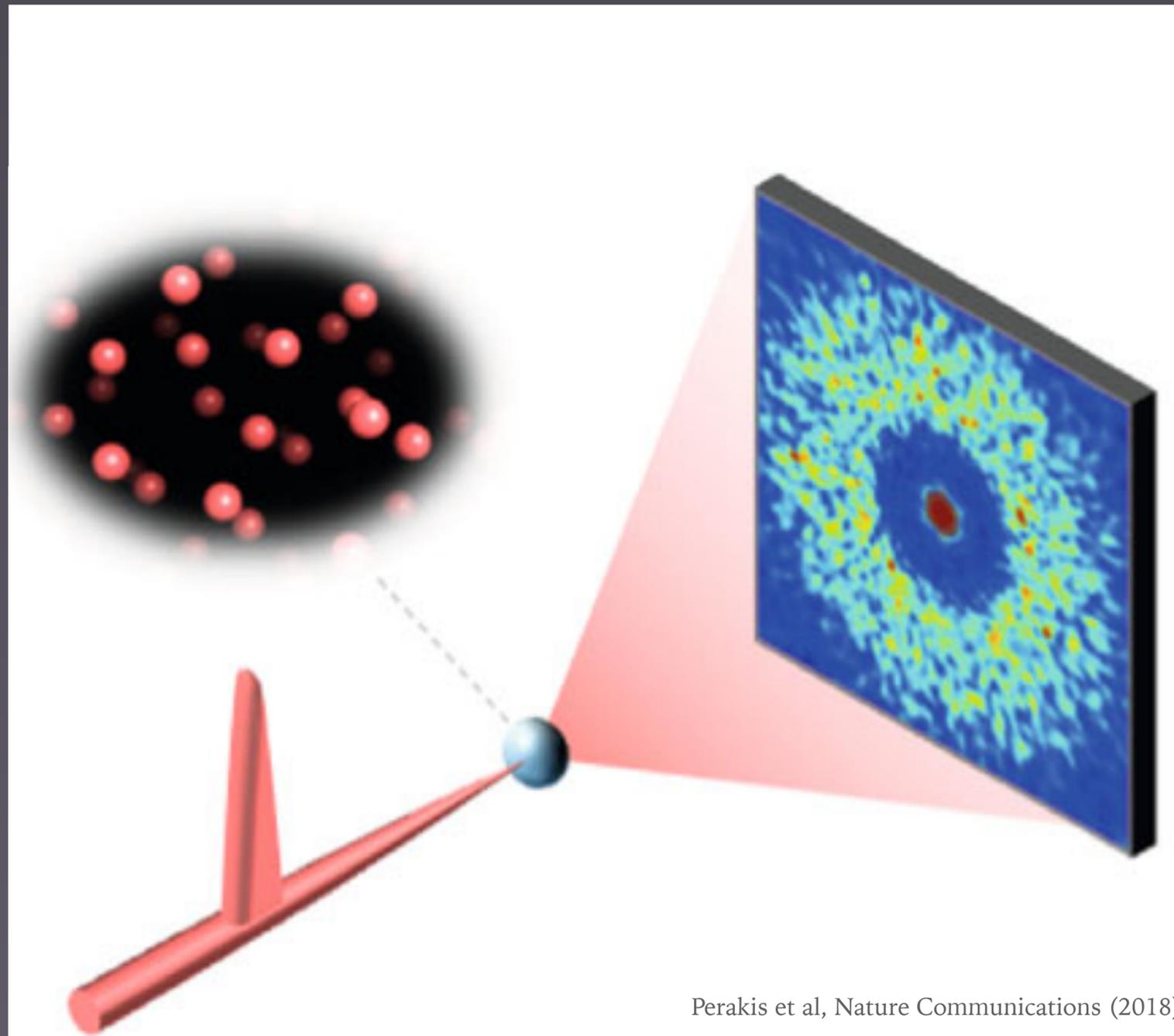
Garanin et al, Quantum Electronics 44 (2014)

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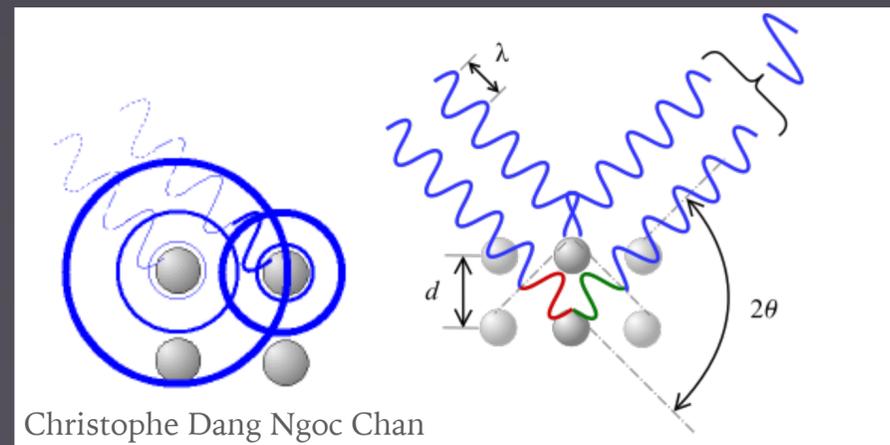


DESY/AG Calegari

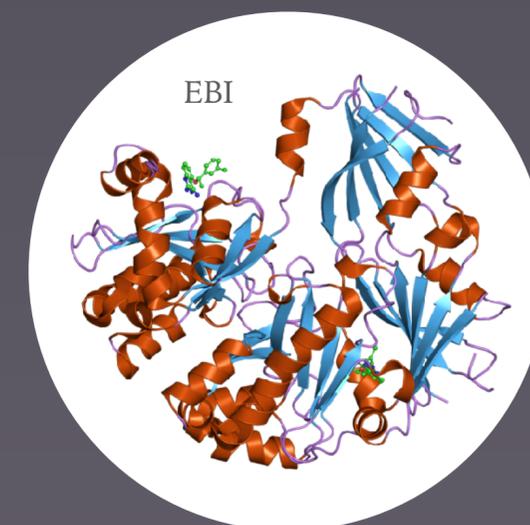
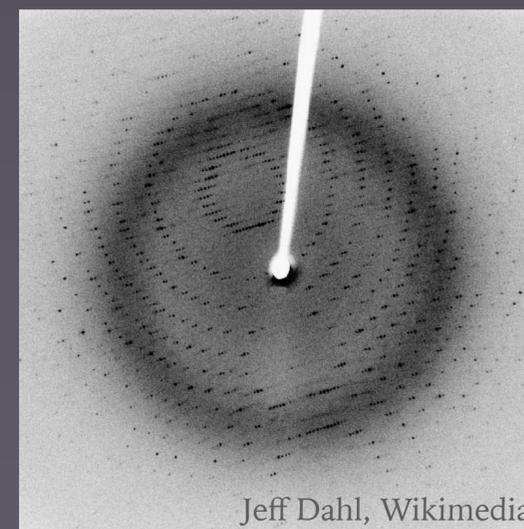
X-Ray Diffraction



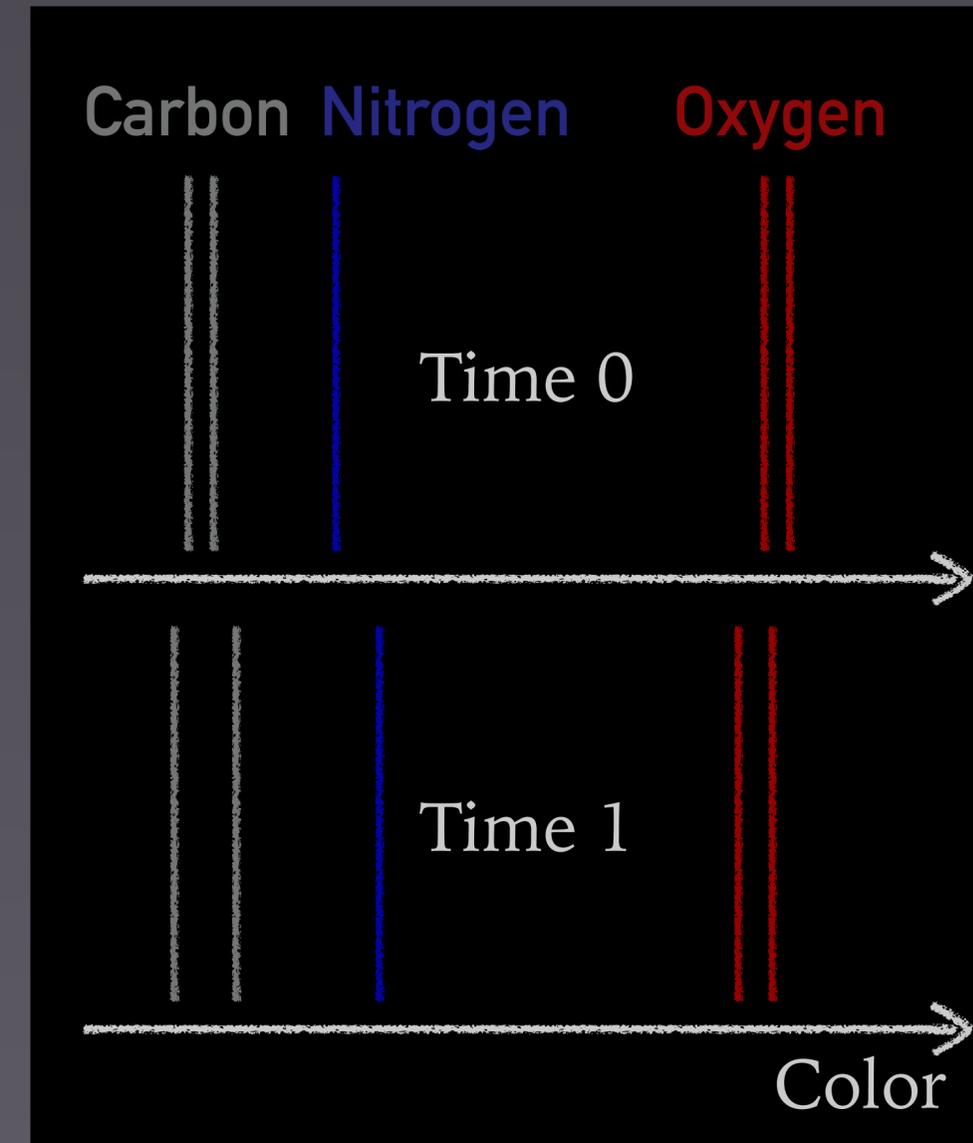
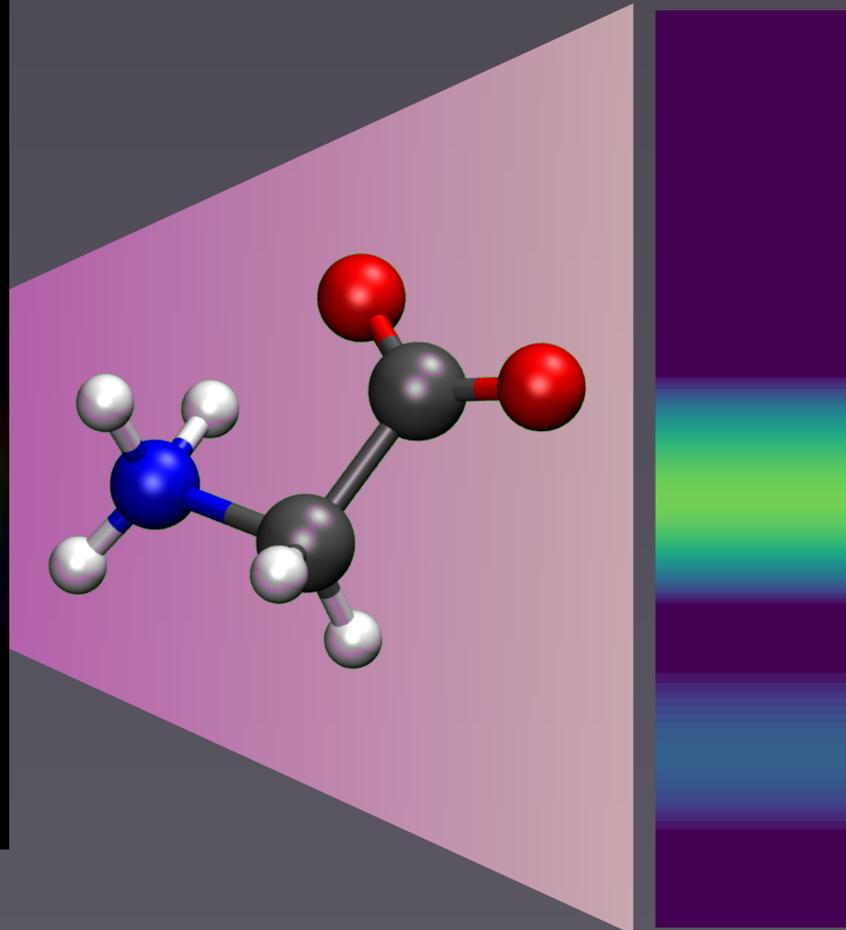
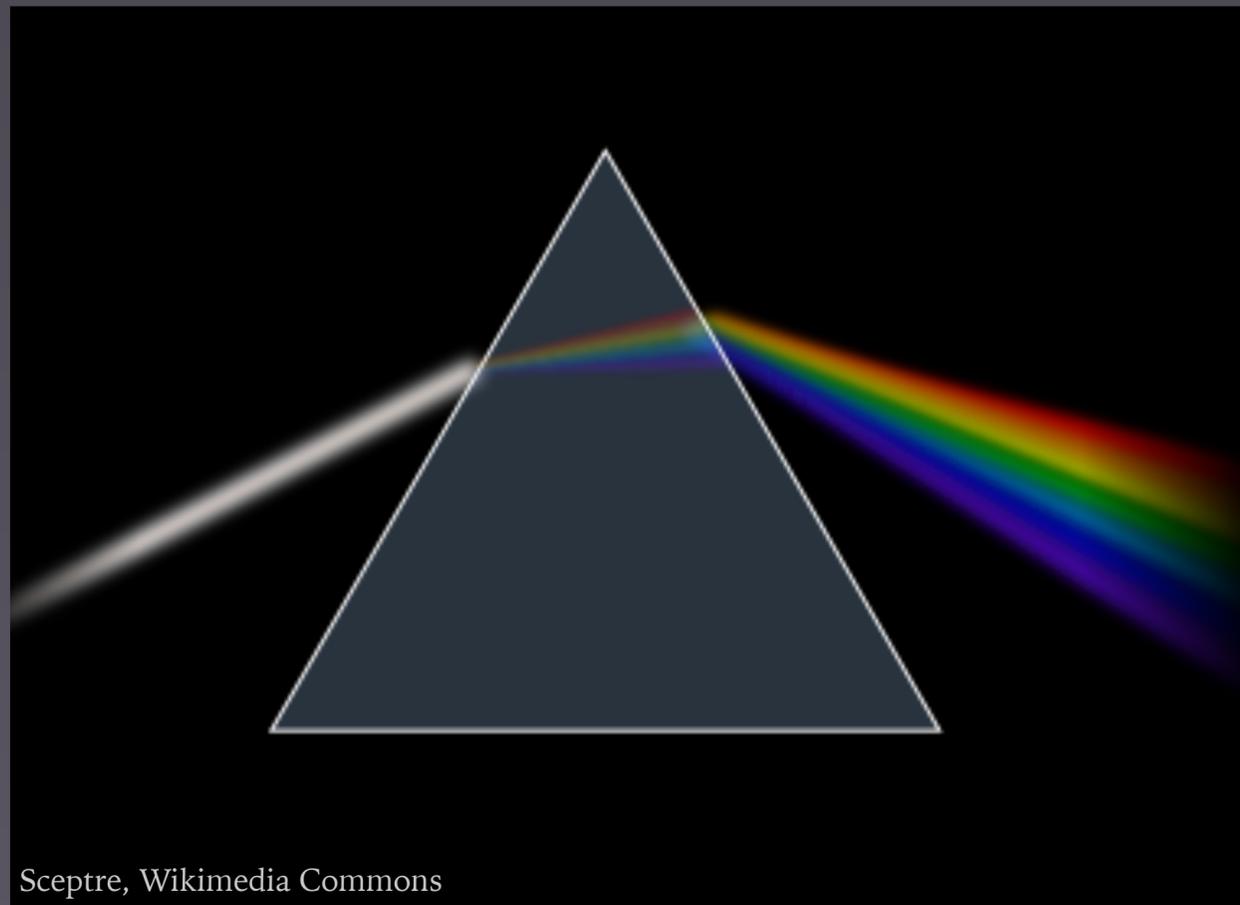
X-rays diffract at crystalline structures



Reconstruction

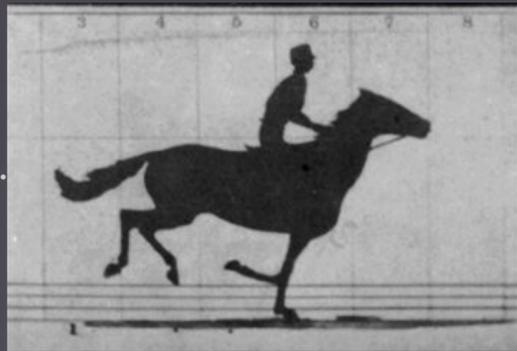


Absorption Spectroscopy



- Molecule blocks certain colors **It's quantum!**
- Characteristic of chemical elements and time-dependent changes
- More probe signals: photoelectrons, fragmentation, ...

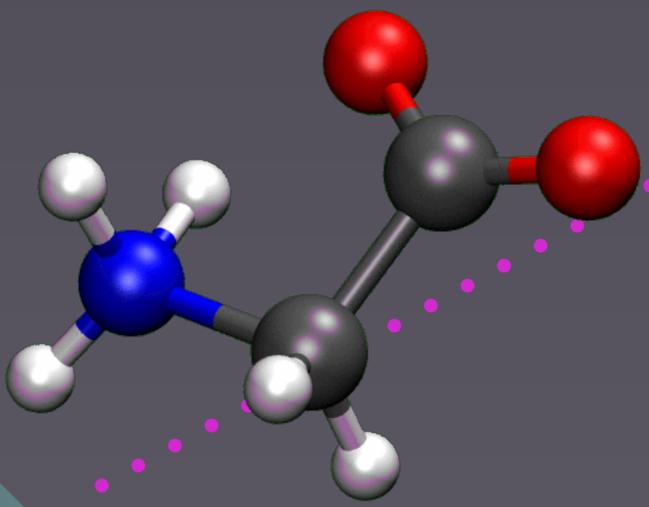
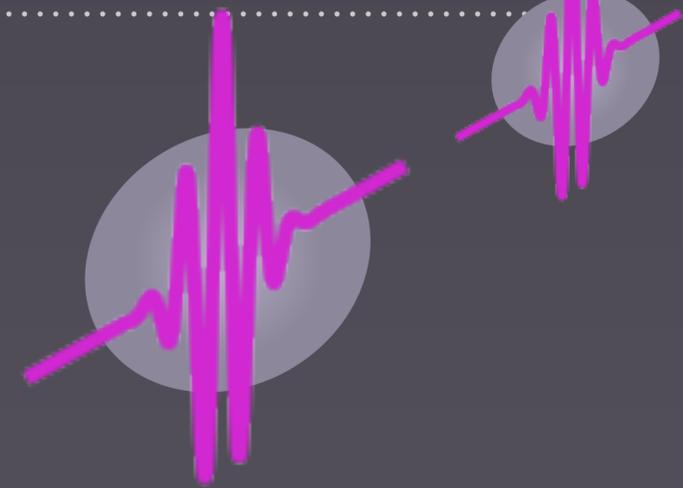
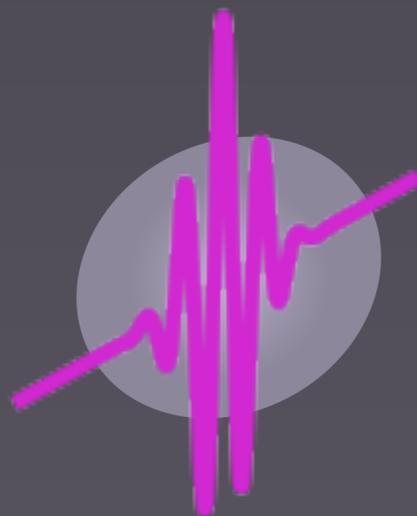
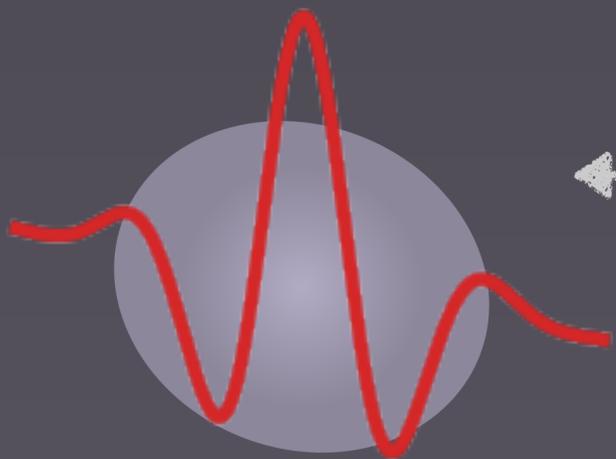
Molecular Movie



Trigger pulse

Probe pulse

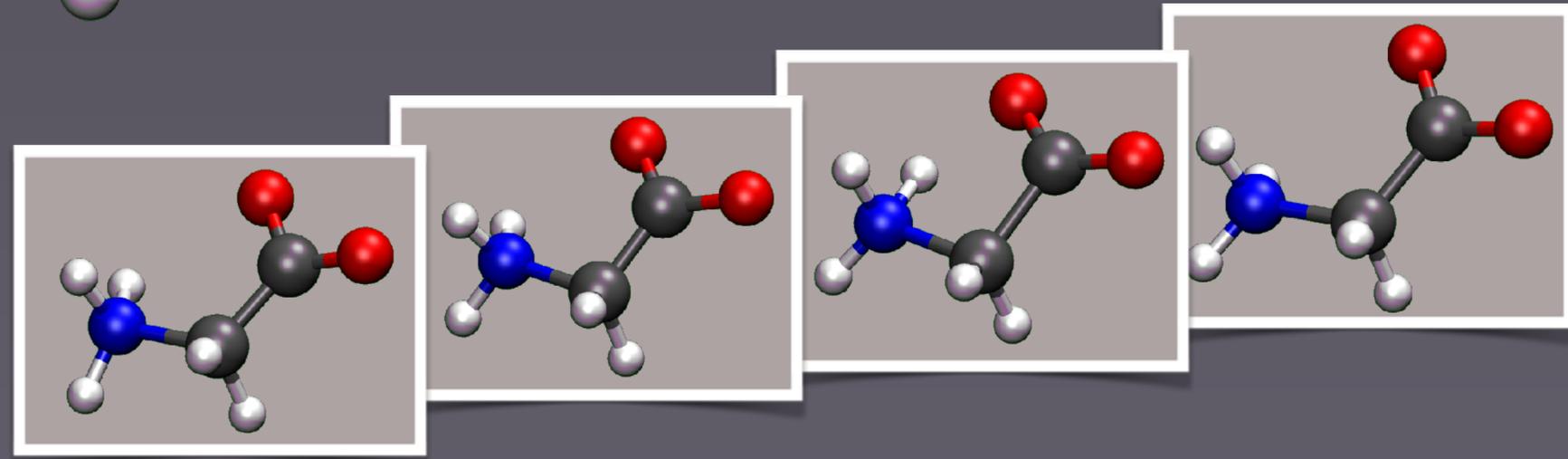
Time delay



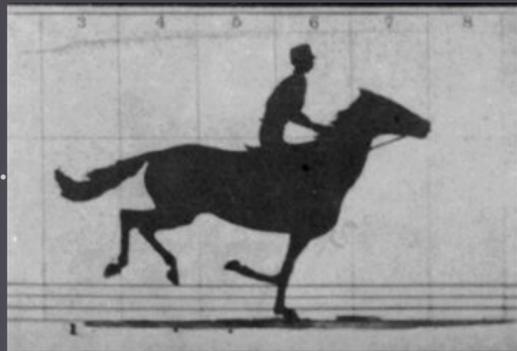
Detector

Reconstruction

```
> ./reconstruct_molecule
```



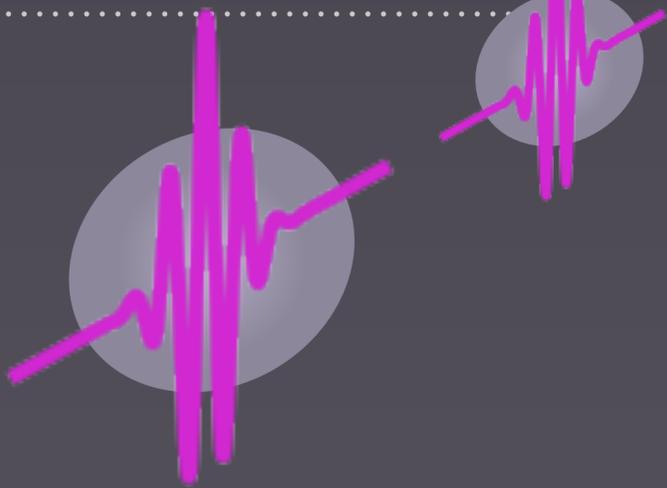
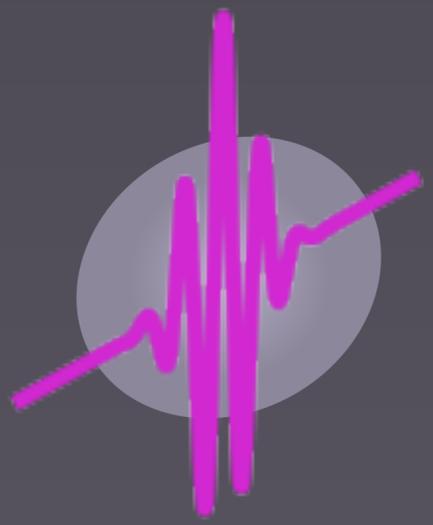
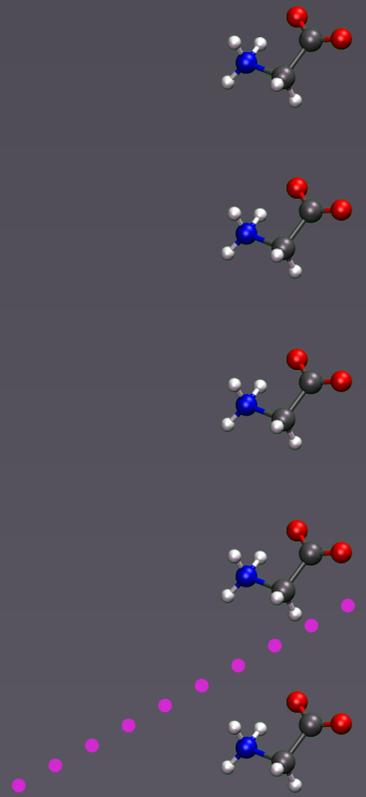
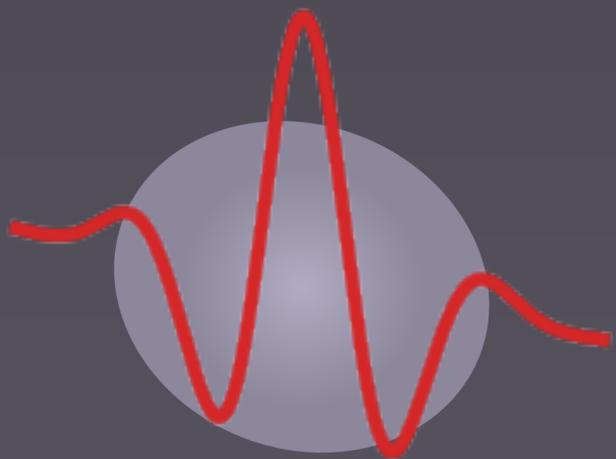
Molecular Movie



Trigger pulse

Sample stream

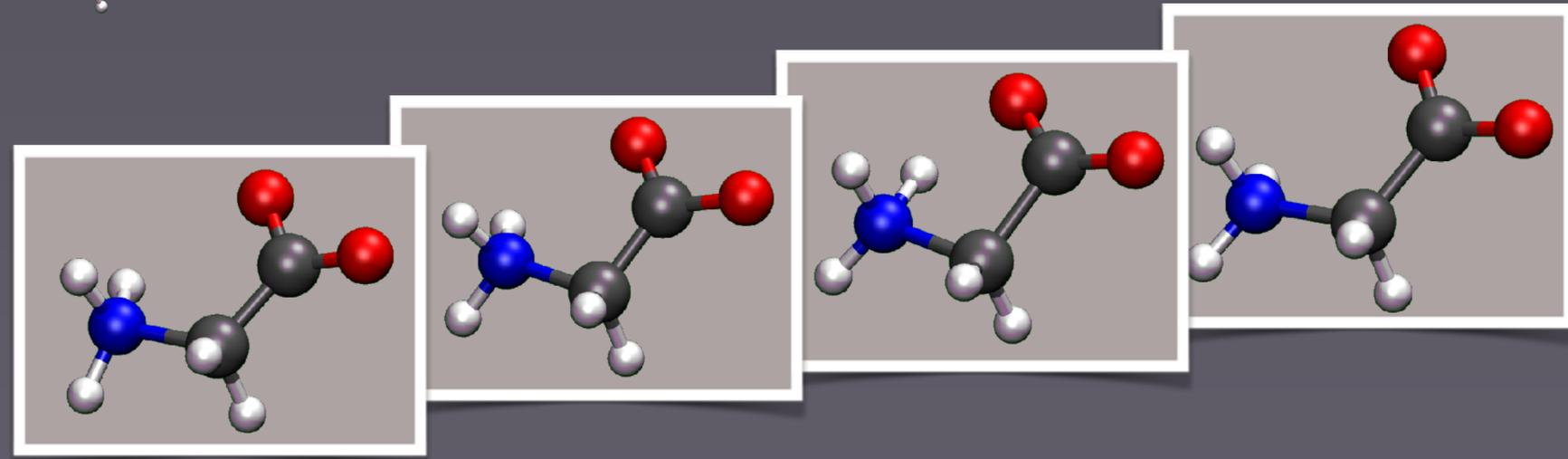
Probe pulse



Detector

Reconstruction

```
> ./reconstruct_molecule
```



Requirements for Light Sources

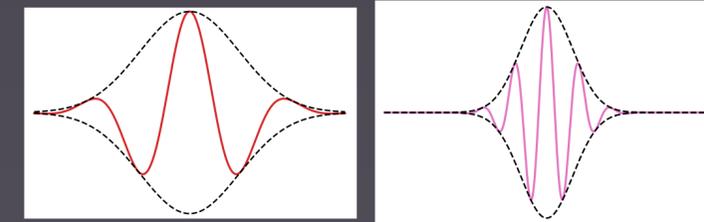
Ultrashort pulses

High brilliance

Small wavelength

Coherent laser light

Center wavelength
Ultrashort pulse

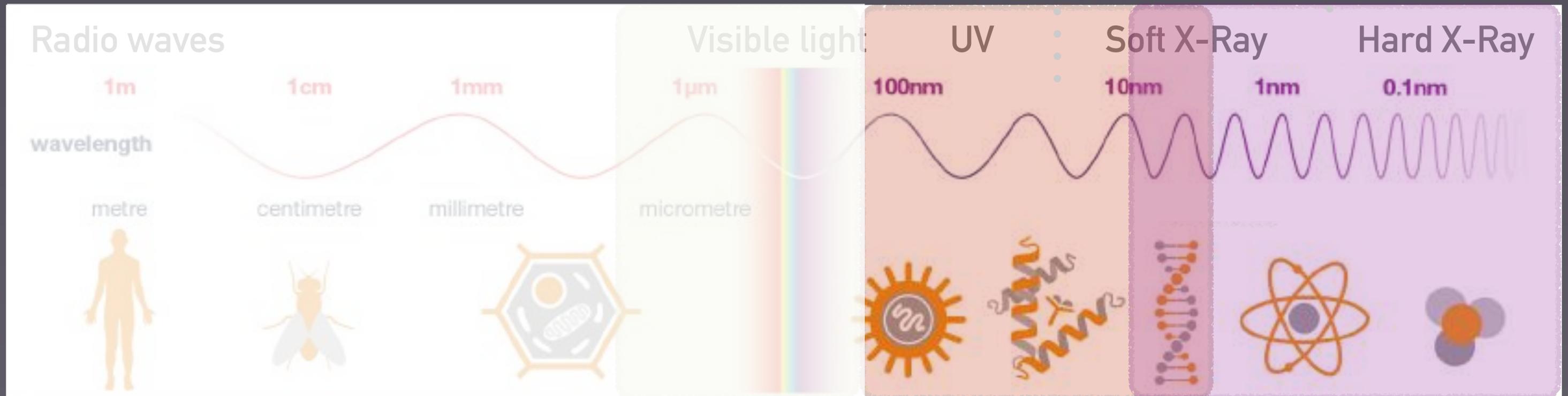


Diffraction limit

Conventional laser

Laboratory

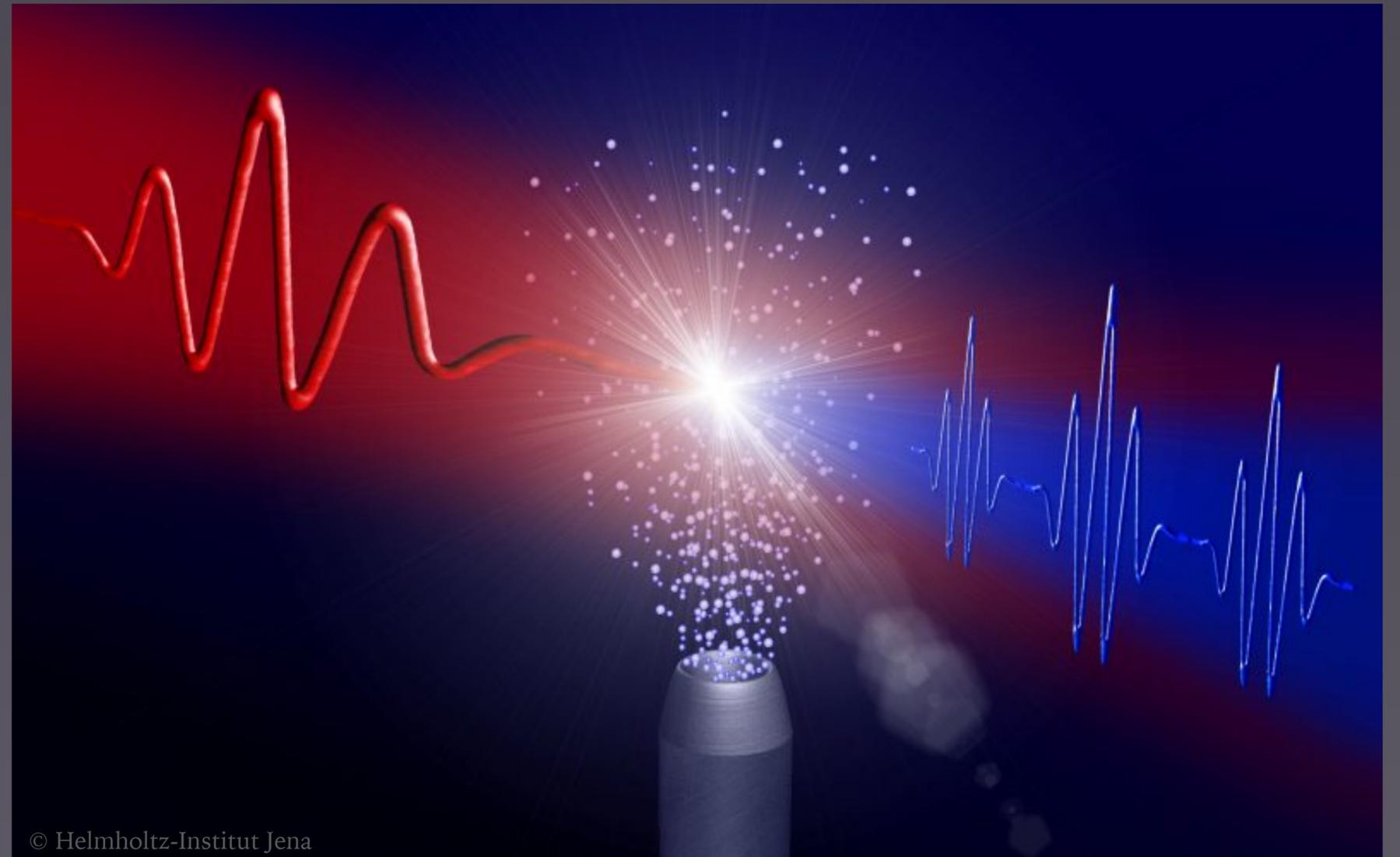
Free-Electron Laser



Ultrashort Laser Pulses in the Laboratory

High-harmonic generation (HHG)

- ▶ High-intensity red laser pulse
- ▶ Focused in gas cell
- ▶ Generates new frequencies of light



© Helmholtz-Institut Jena



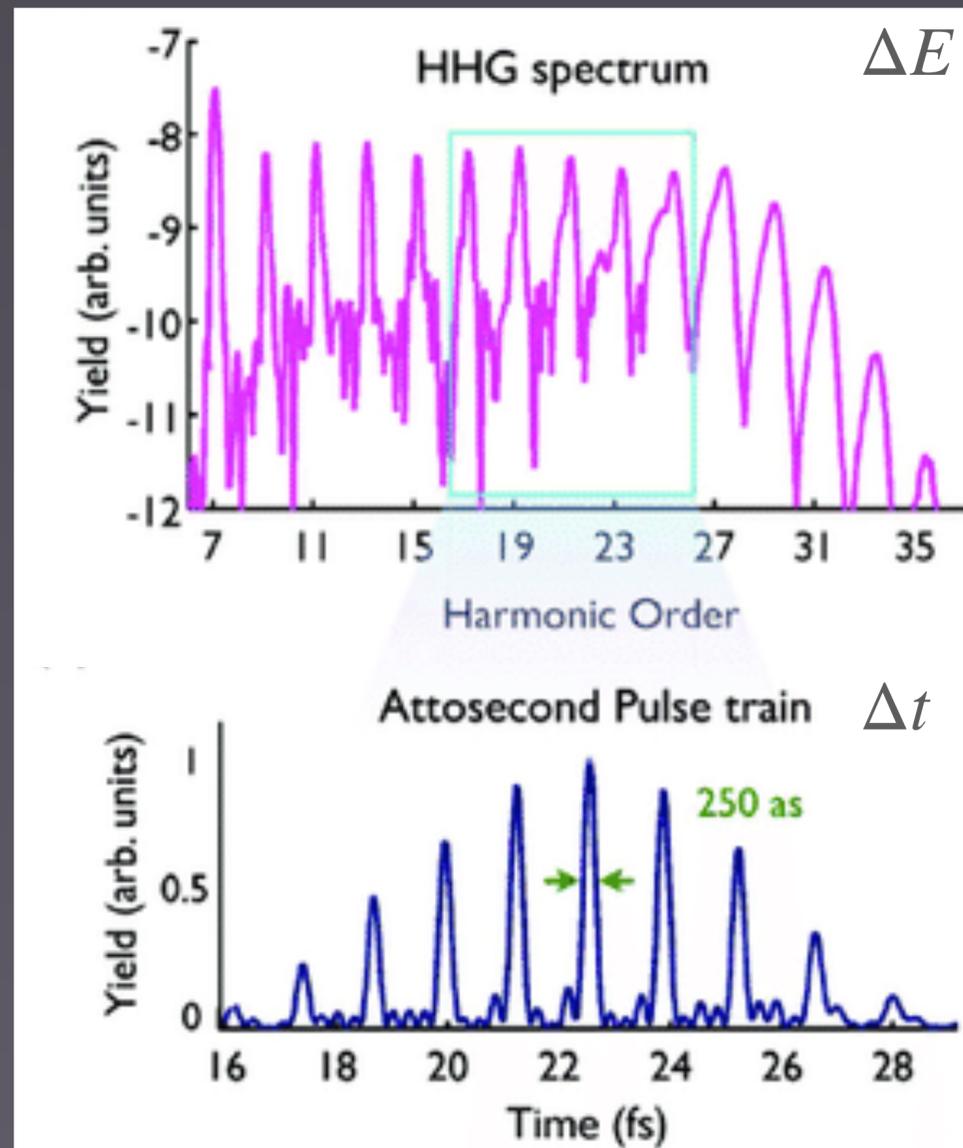
Donna Strickland

©Nobel Media, Alexander Mahmoud

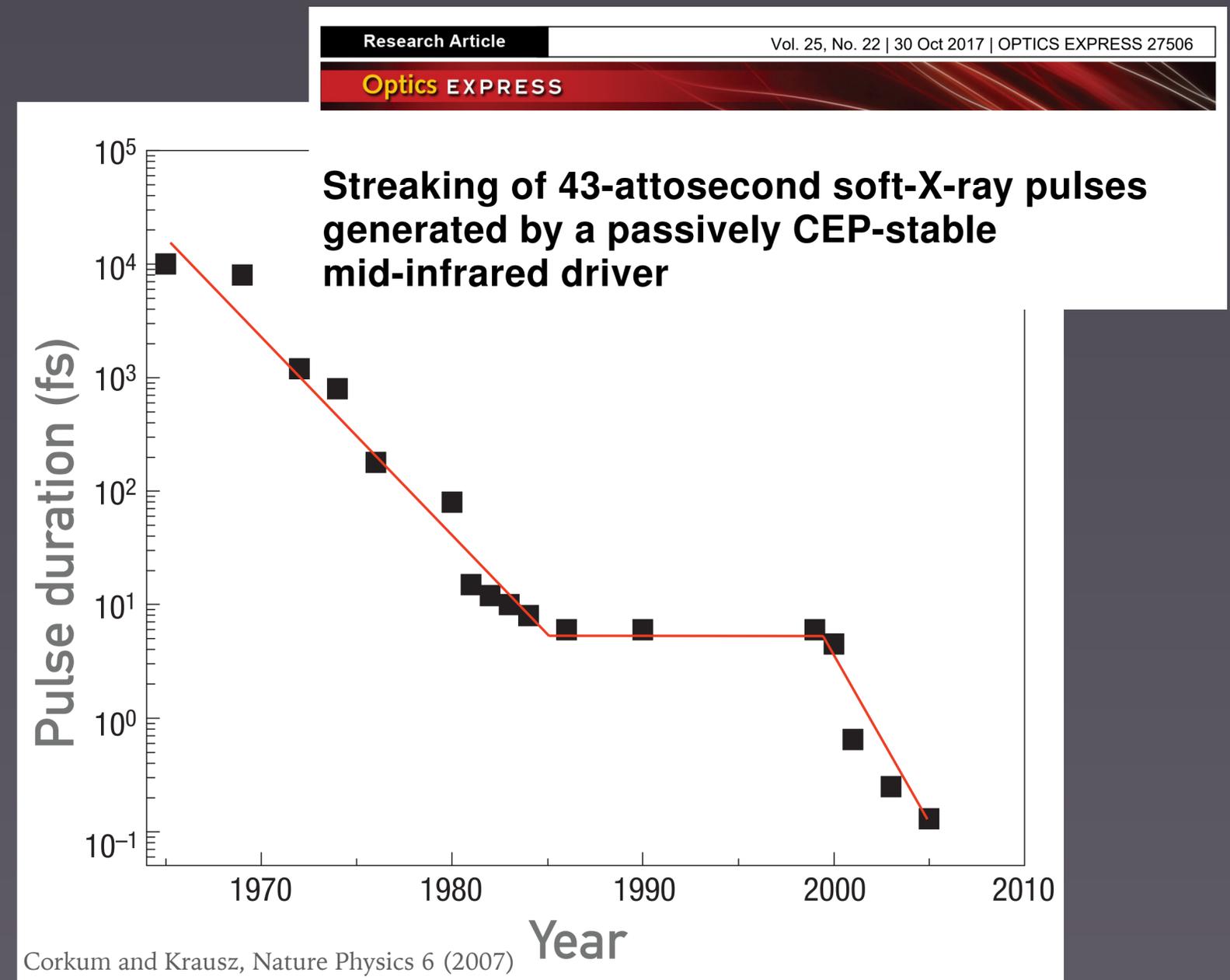
Gérard Mourou

Ultrashort Laser Pulses in the Laboratory

Fourier limit $\Delta E \cdot \Delta t = 0.442$



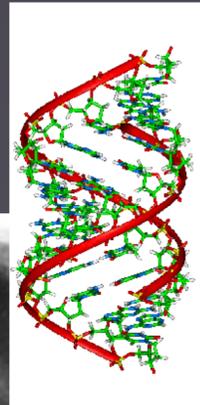
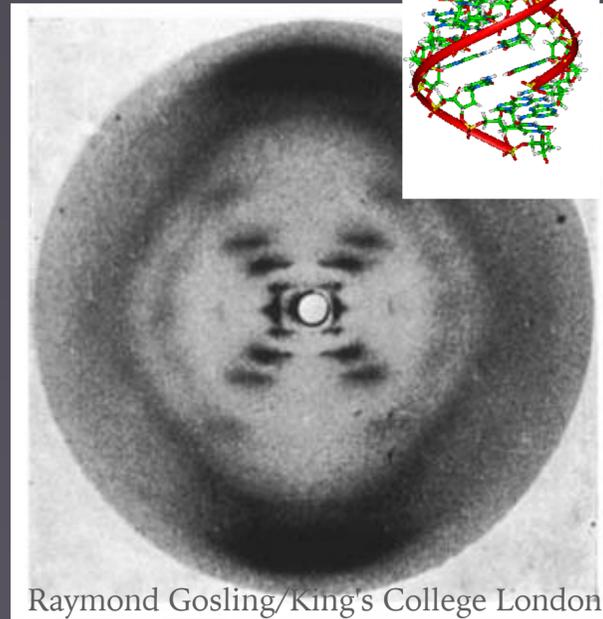
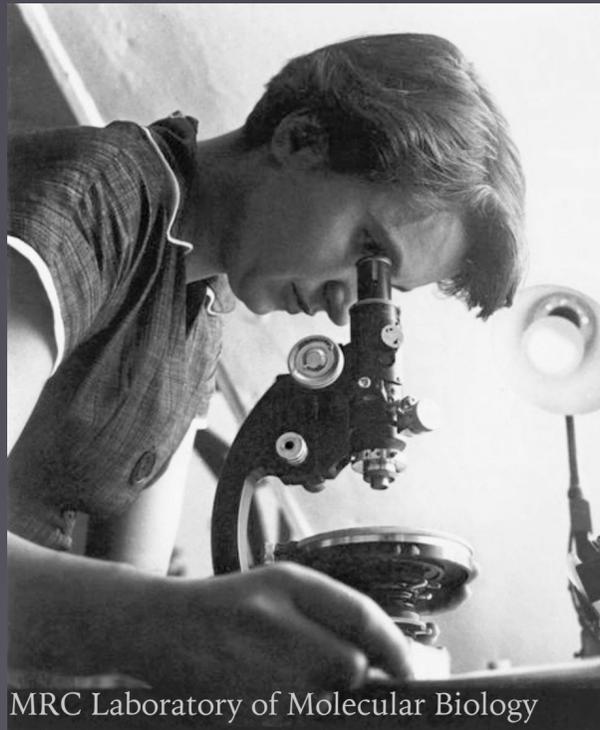
Ultrashort Extreme Ultraviolet Vortices. 10.5772/64908



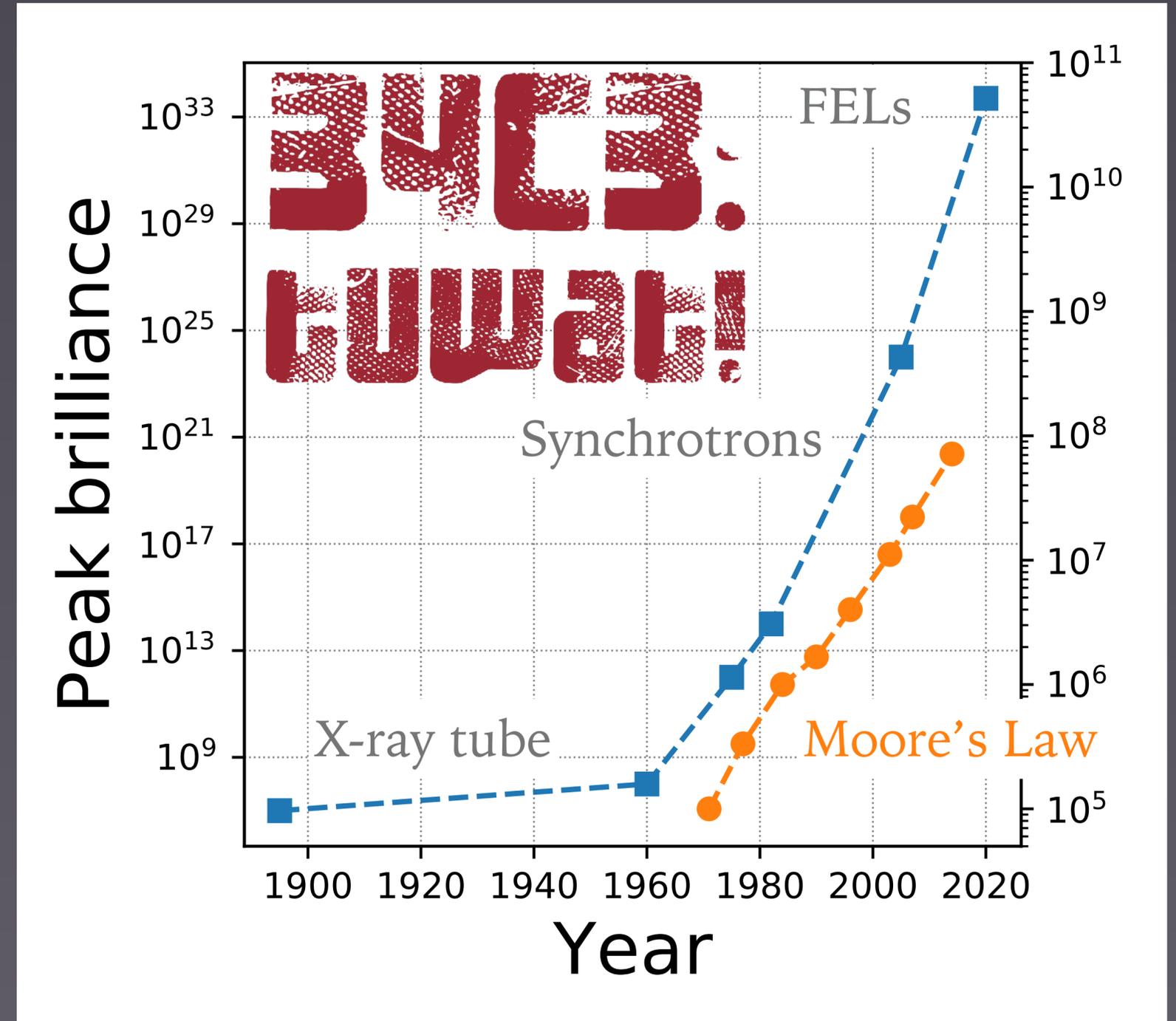
Cannot go to hard x-rays

Light Source for X-Ray Diffraction

Rosalind Franklin (1920 - 1958)

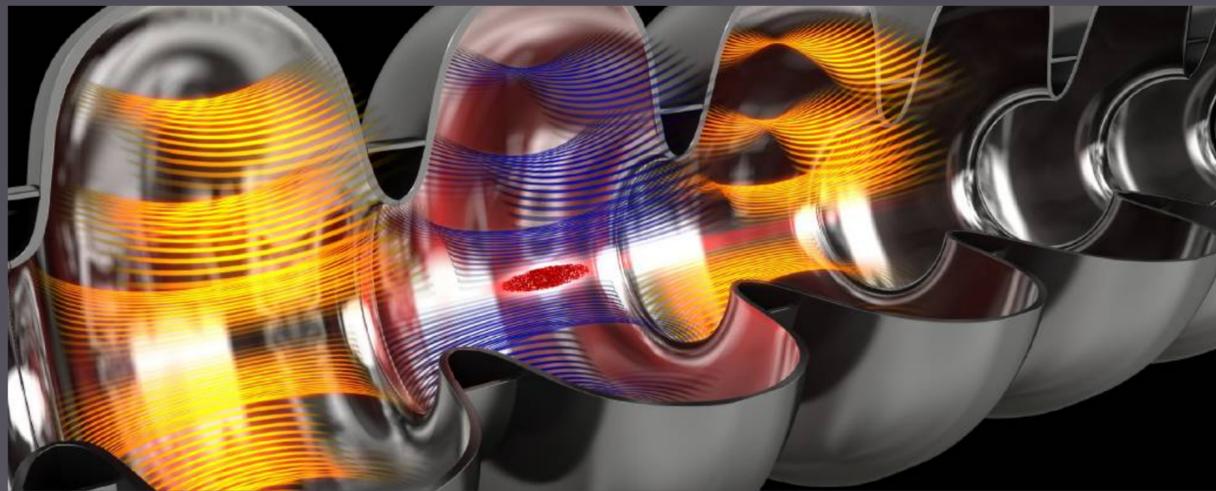


- X-ray diffraction revealed DNA structure
- Shorter pulses require high brilliance
(Photons in the beam per second)



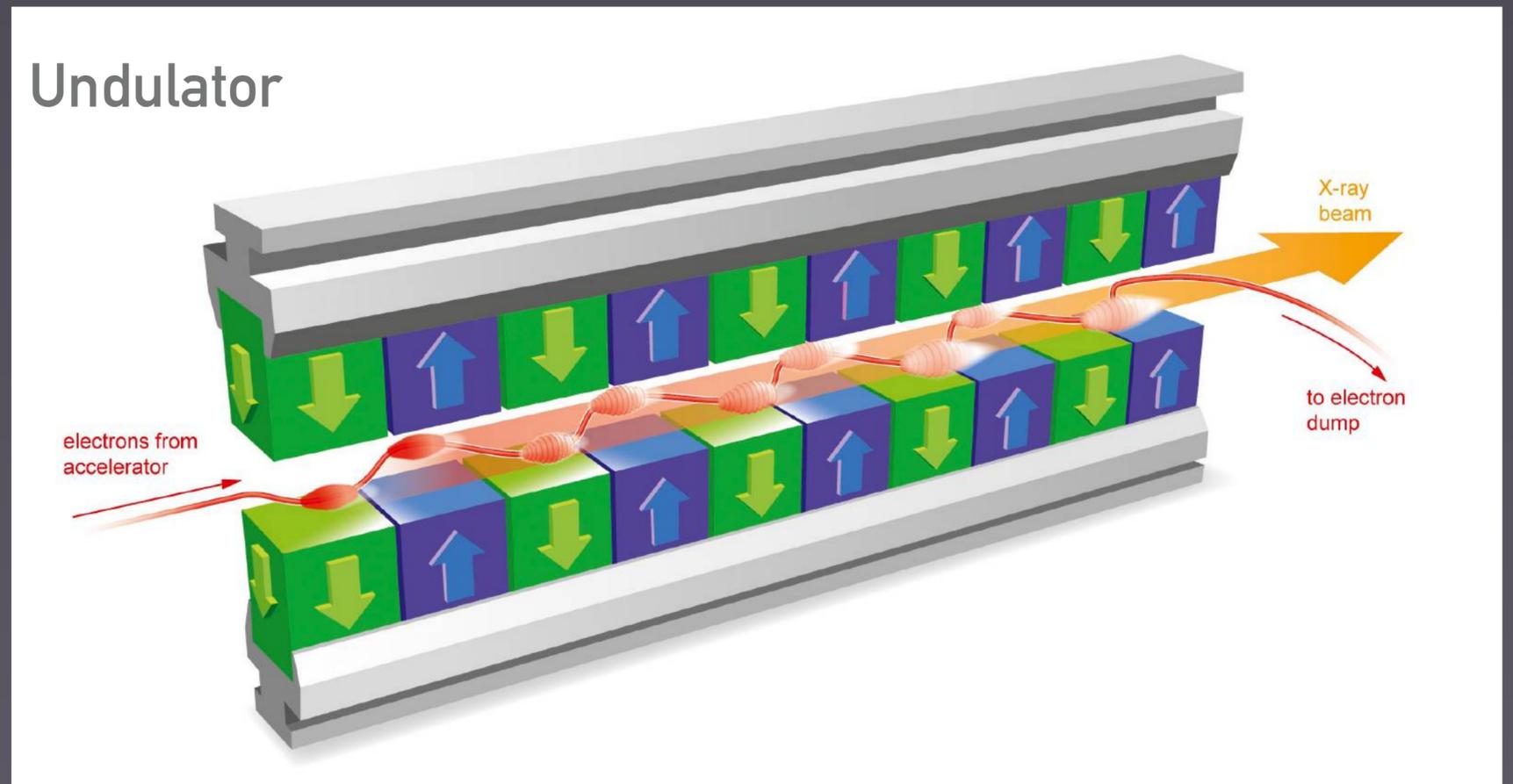
Free-Electron Laser

Accelerate electrons to relativistic speed



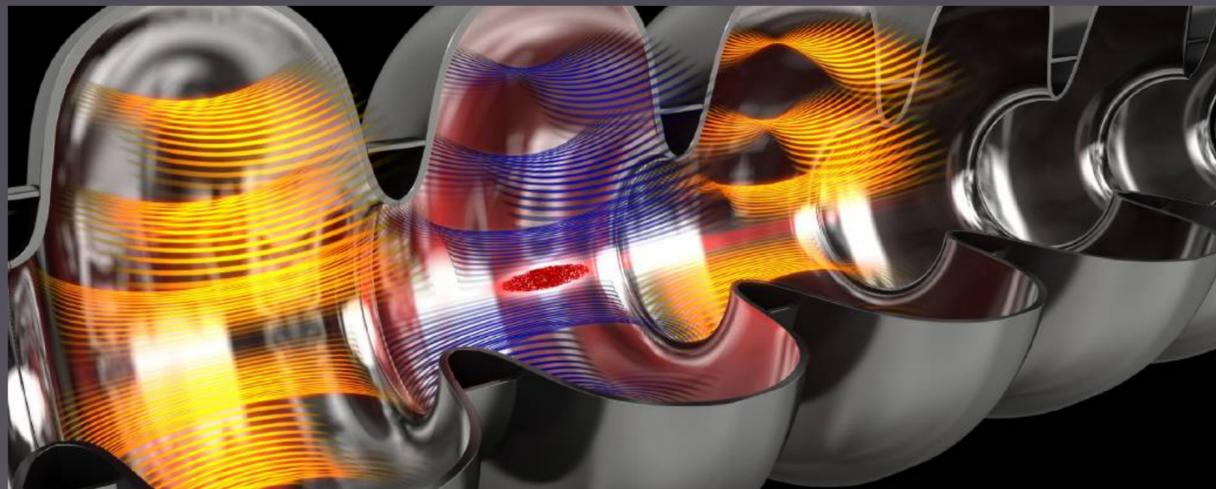
Electrons enter undulator

- Magnets force wiggling motion
- X-ray radiation emitted



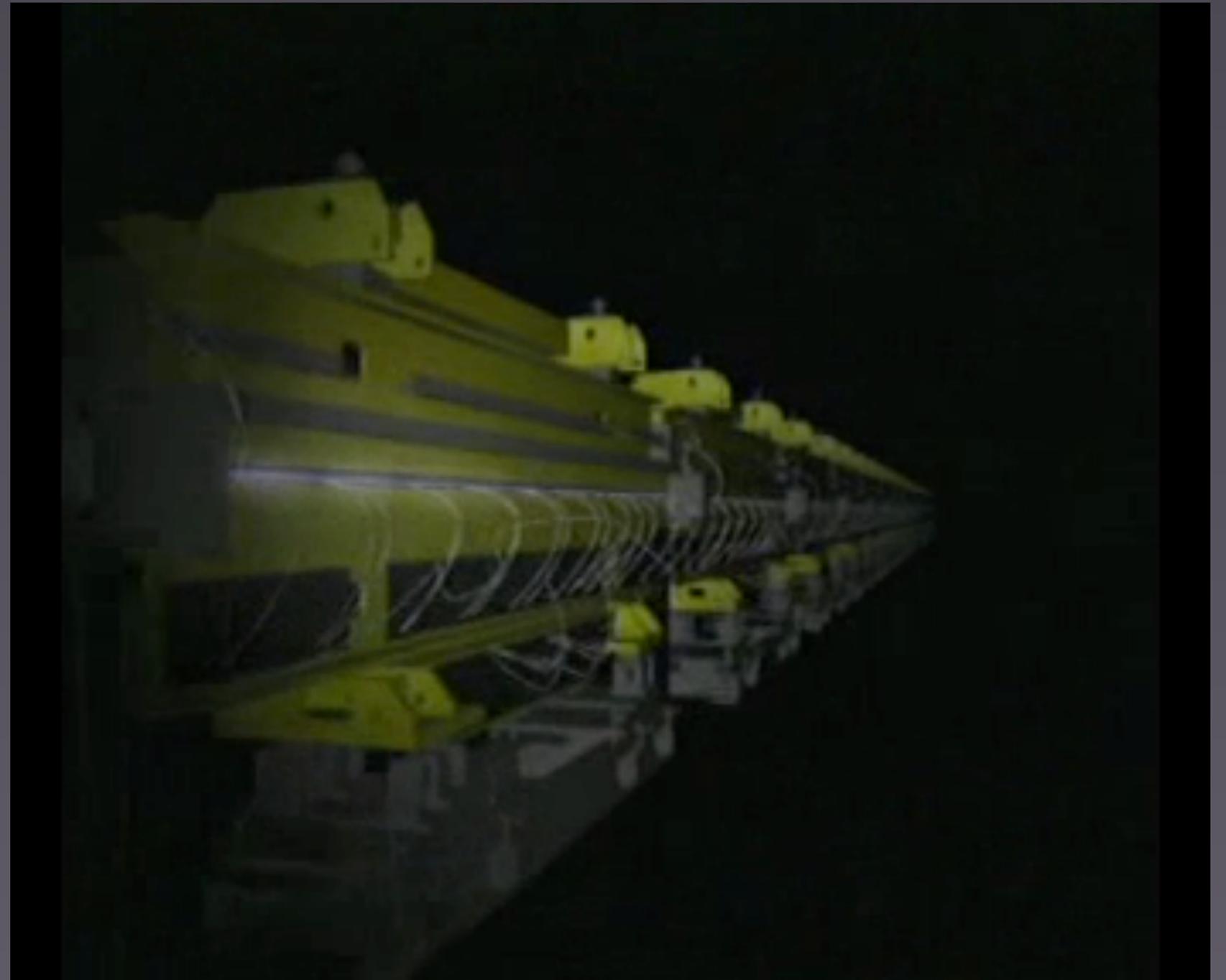
Free-Electron Laser

Accelerate electrons to relativistic speed

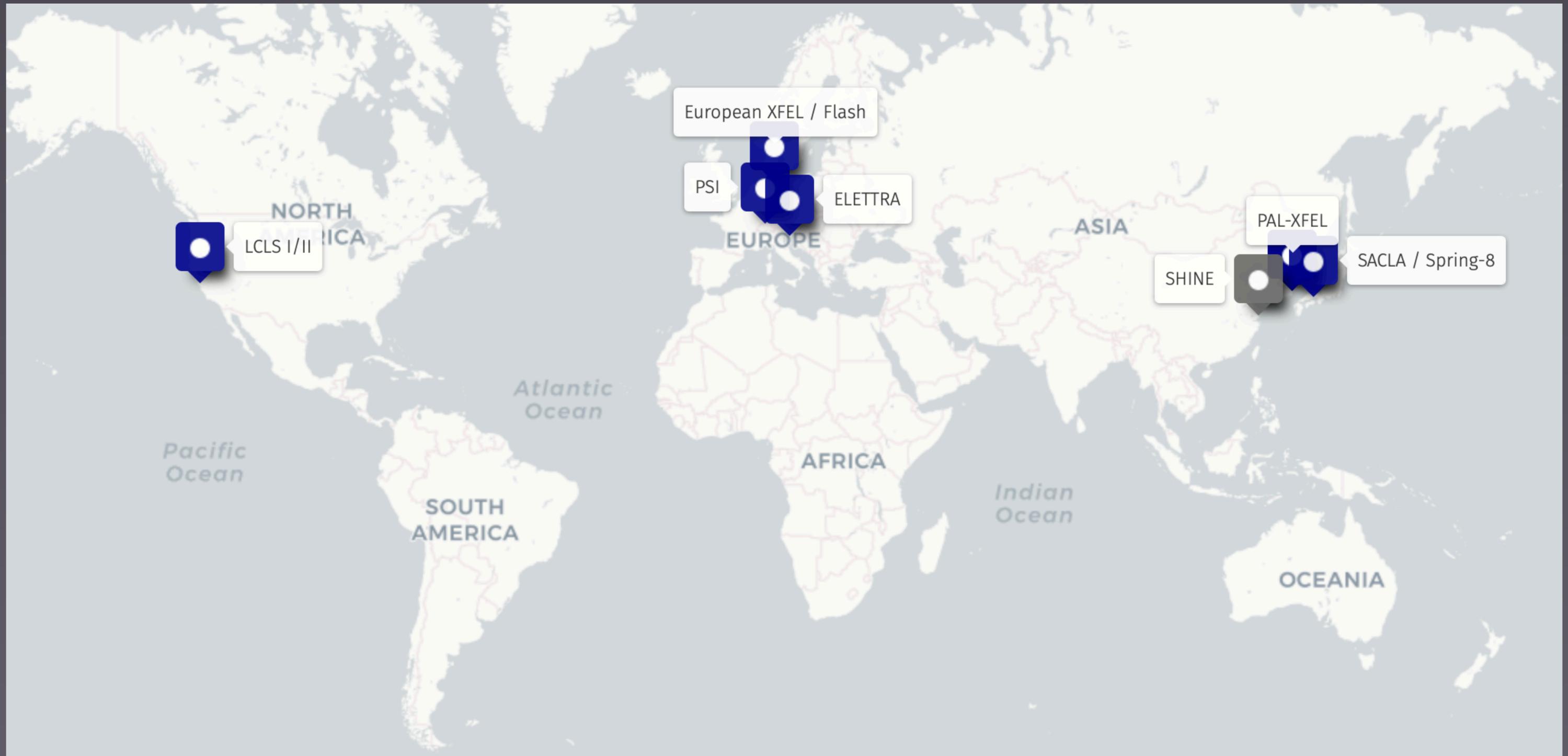


Electrons enter undulator

- Magnets force wiggling motion
- X-ray radiation emitted

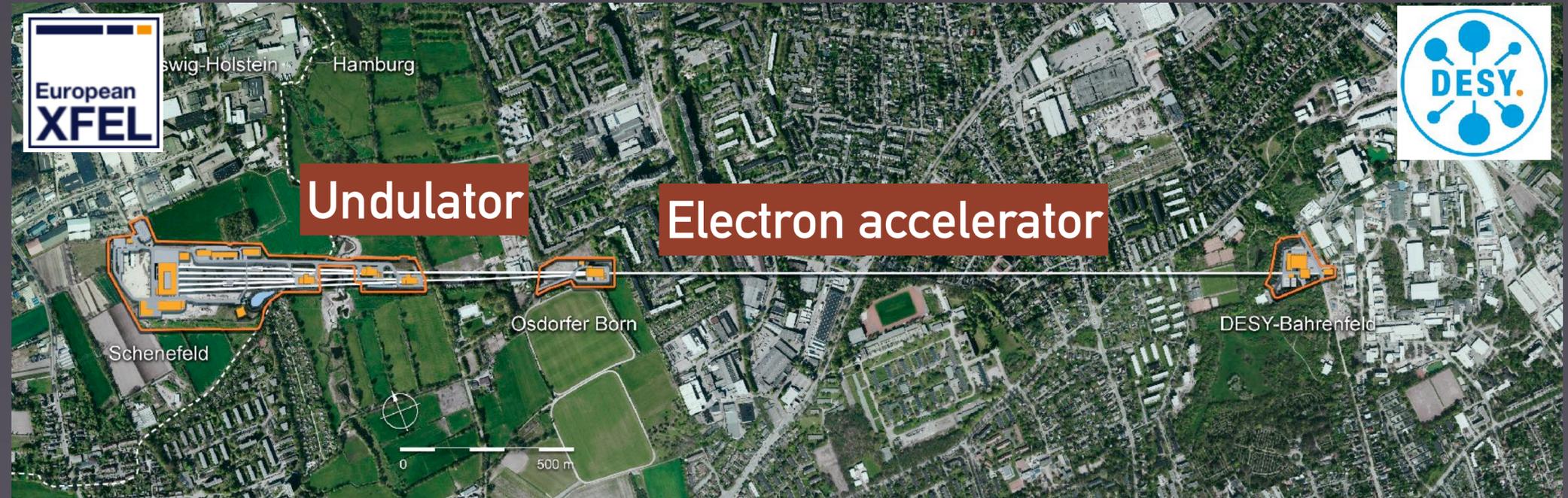
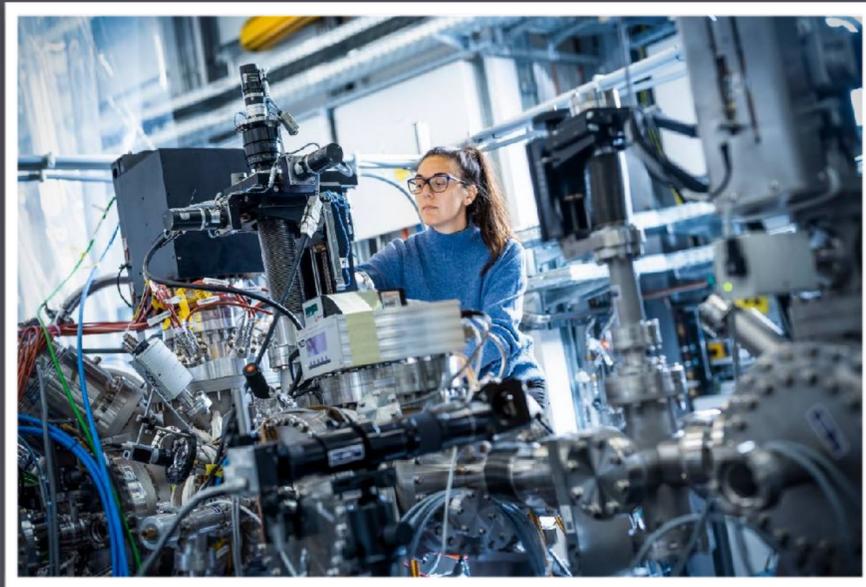


Free-Electron Lasers in the World



European X-Ray Free-Electron Laser

3.4 km long

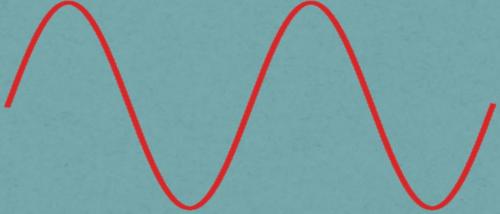
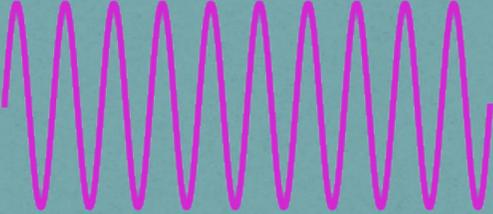


- Six experimental end stations
- Material science, biomolecule imaging, chemistry, ...
- 27,000 flashes / second

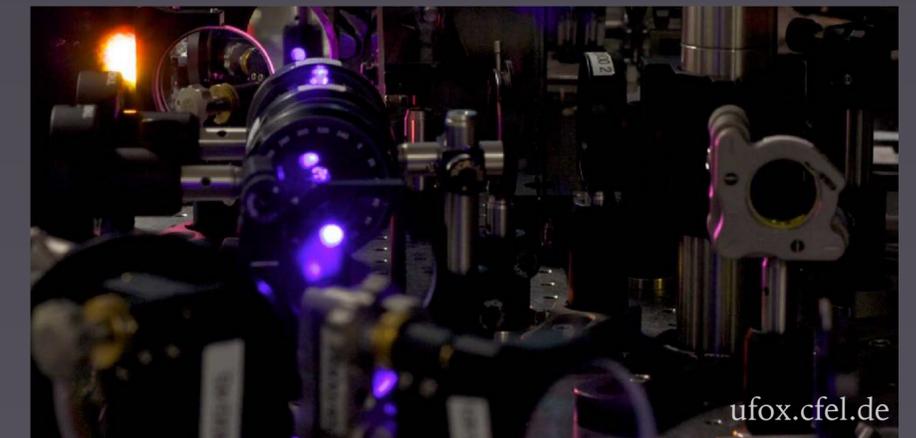
- Total construction costs
8 years / 1.25 billion €
- Elbphilharmonie
9 yrs / 870 million €



Comparison of Light Sources

	Laboratory	XFEL
Min pulse duration	< 100 attoseconds	10 femtoseconds
Peak brilliance		
Wavelength		
Control		
Cost and availability		

Laboratory setup

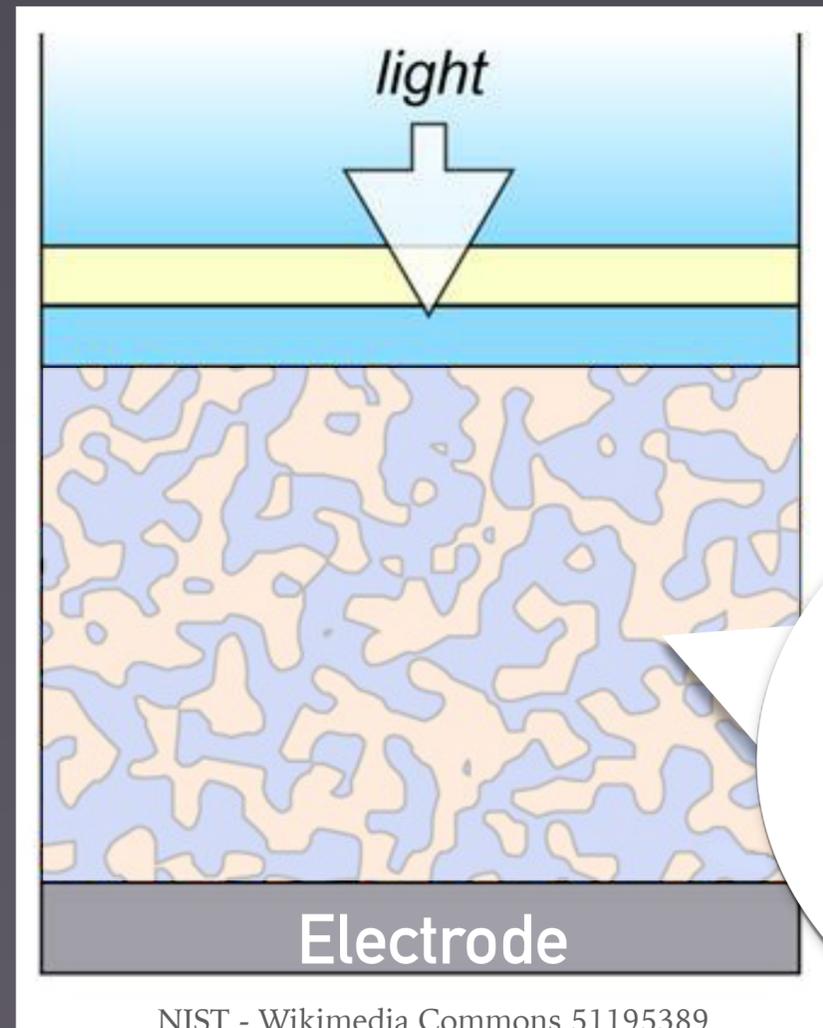


XFEL tunnel



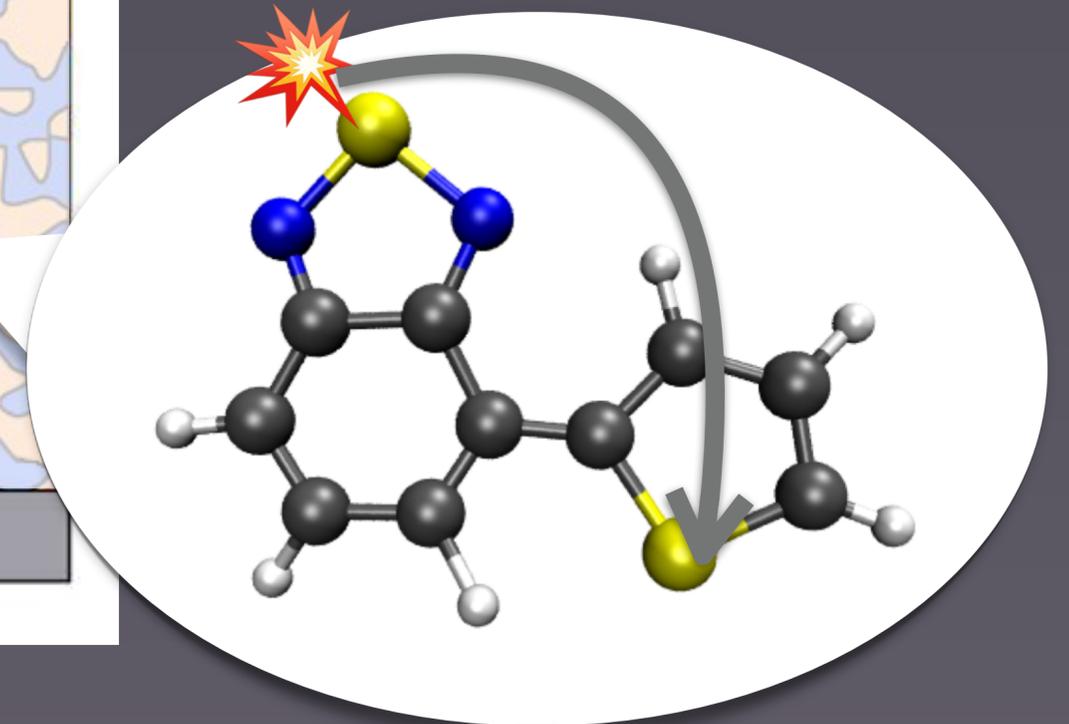
Organic Solar Cell

An example for insights from ultrafast science



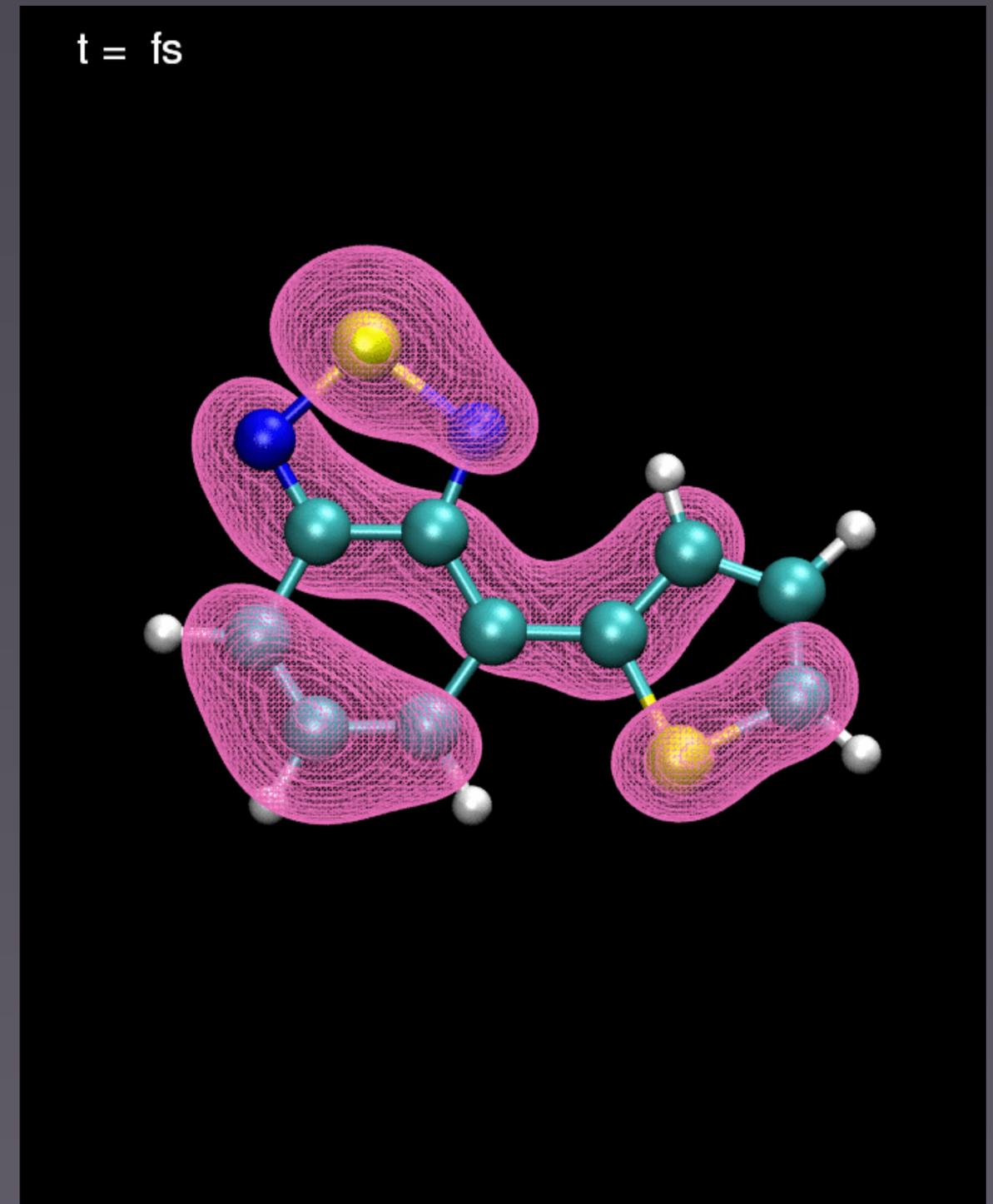
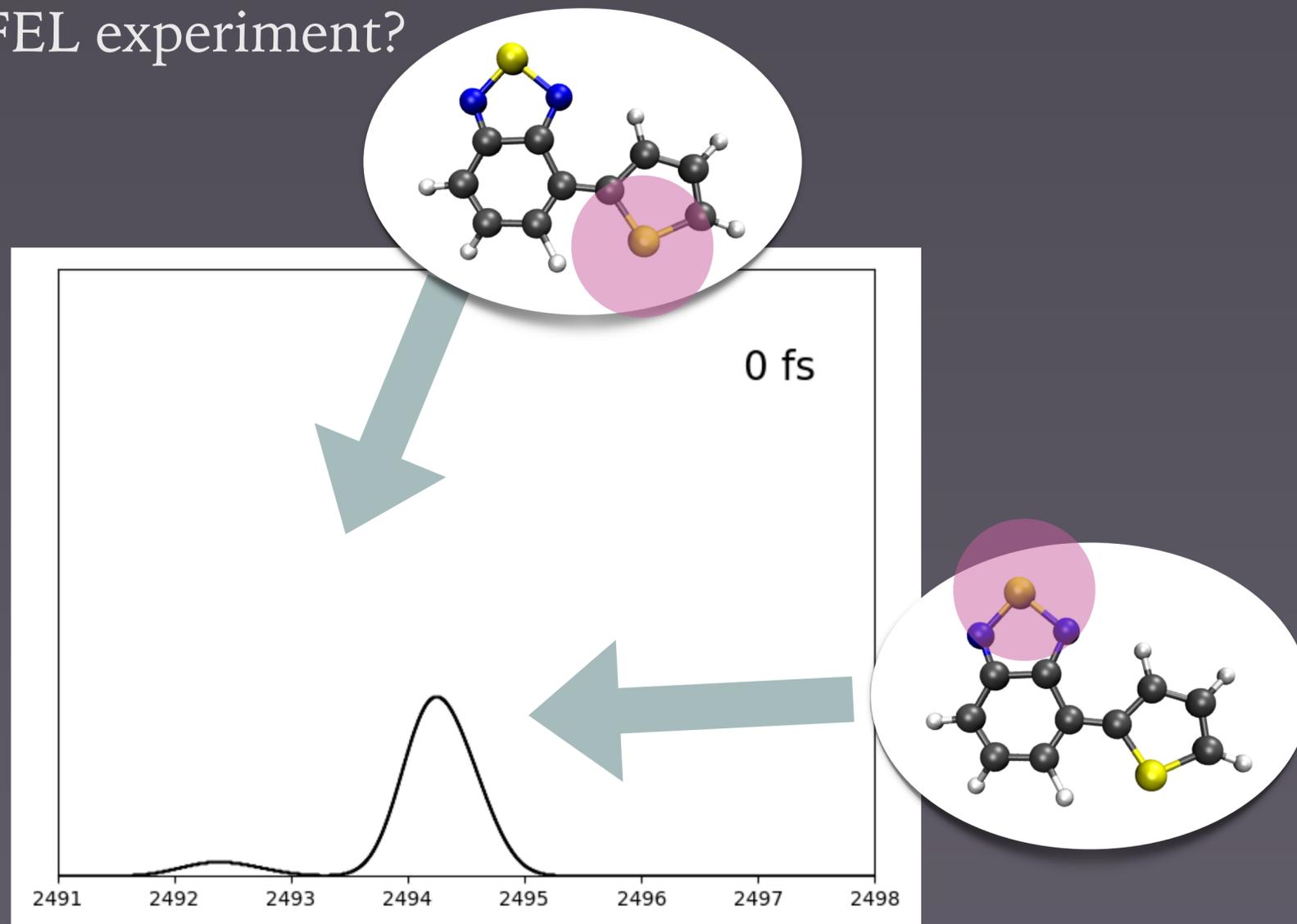
Need to transport the charges

Polymer BT-1T



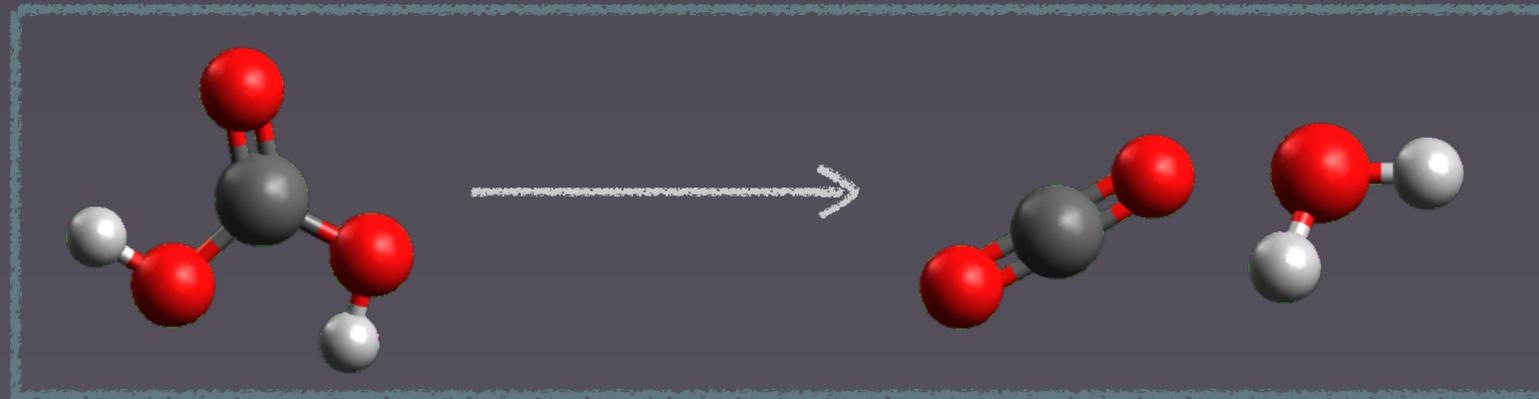
Observe Charge Migration

- Calculate the ultrafast charge migration in BT-1T
- Shows up in the x-ray absorption spectrum
- XFEL experiment?



Beyond the Molecular Movie

Chemical reactions involve molecular dynamics



Trigger

Control reaction with light

Optimisation routine

```
> ./optimise_pulses
```

Generate sequence

Ultrafast wish list

- Take the molecular movie
- Direct the molecular movie

An Ultrafast Developing Field

Google Scholar "Ultrafast"



naturep

Article | Published: 23 Sep 2017

**Femtosecond
fragmentation
following X-ray**

N. Berrah , A. Sanchez

nature reviews chemi

Perspective | Published: 29 May 2018

**The ultrafast X-ray
revolution in chem**

Peter M. Kraus , Michael Zürc, S
Stephen R. Leone 

OPEN ACCESS
IOP Publishing
J. Phys. B: At. Mol. Opt. Phys. 51 (2018) 032003 (45pp)

Roadmap
**Roadmap of ultrafast
molecular physics**

nd Dynamics with

es using



tion in
irs of adenine

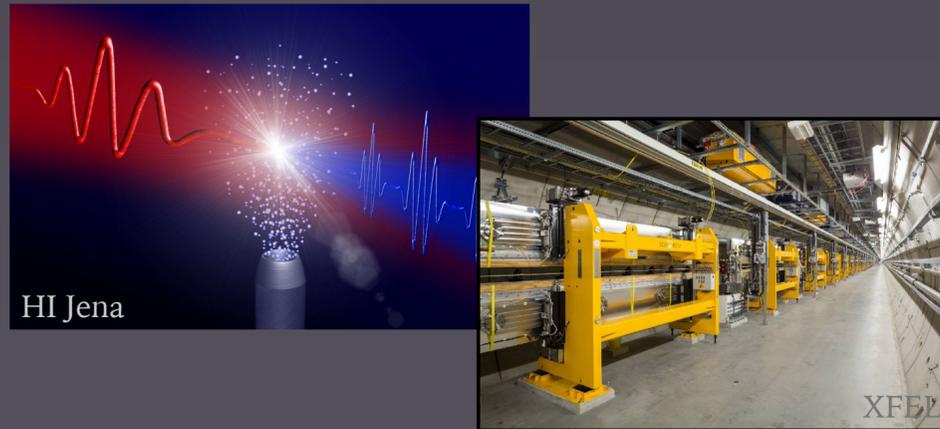
nylalanine initiated

by attosecond pulses

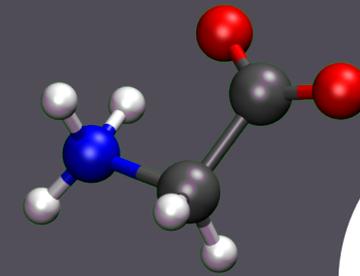
F. Calegari¹, D. Ayuso², A. Trabattoni³, L. Belshaw⁴, S. De Camillis⁴, S. Anumula³, F. Frassetto⁵, L. Poletto⁵, A. ...

What's Next?

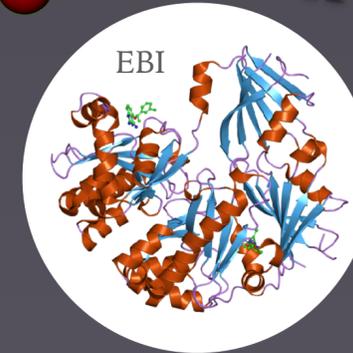
Develop light sources



Theoretical calculations

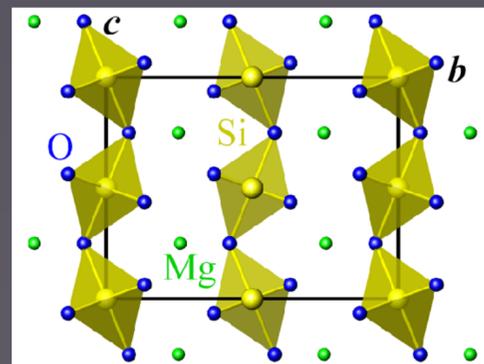


$$\hat{H} = \hat{H}_0 + \lambda \hat{H}_{\text{int}}$$

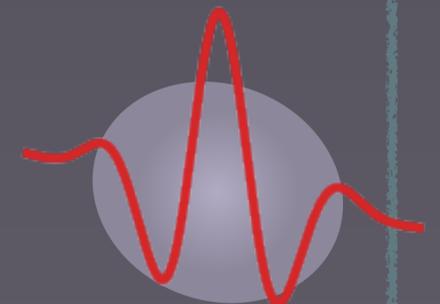


$$i \frac{\partial}{\partial t} \psi = \hat{H} \psi$$

Larger systems



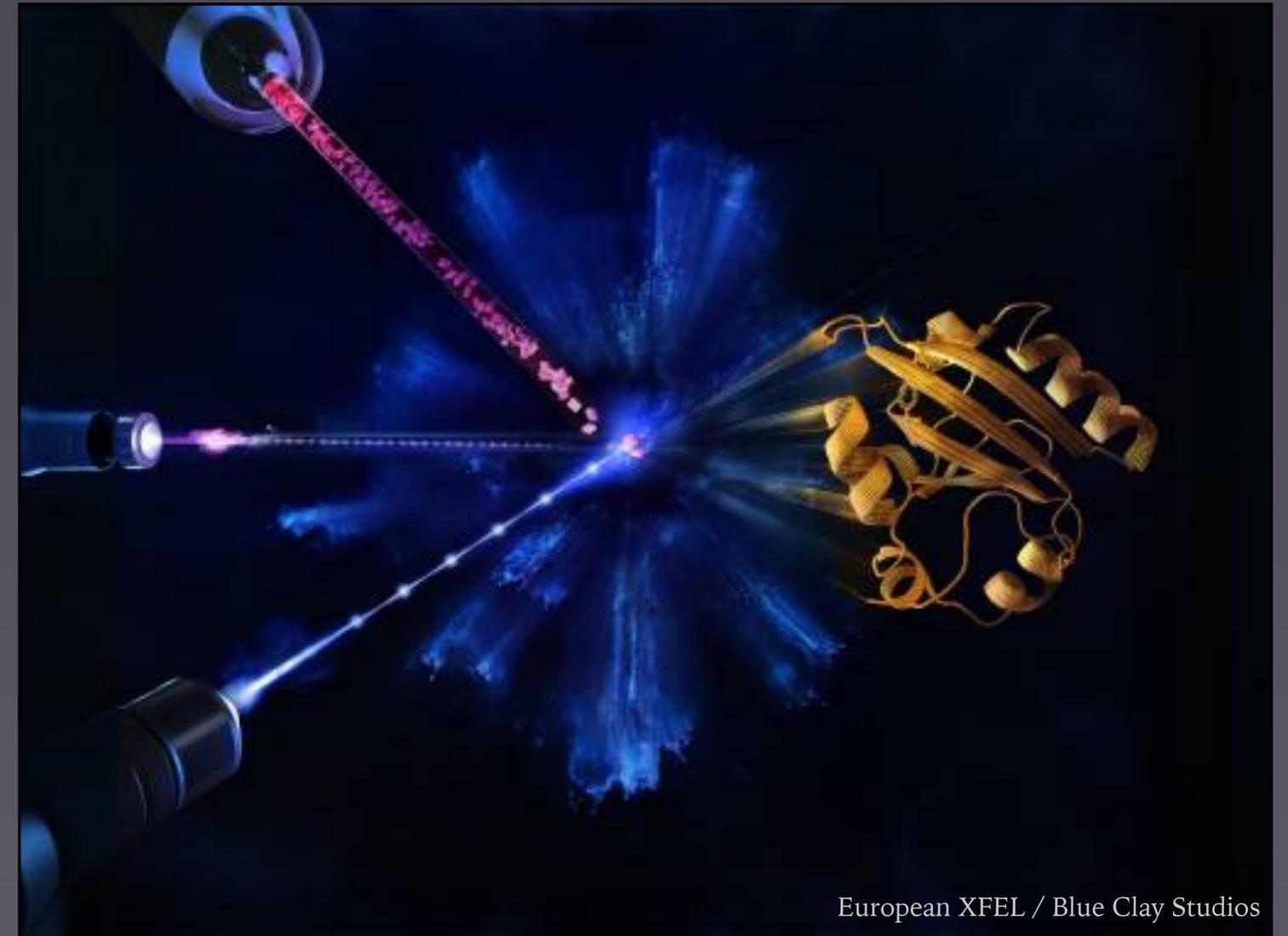
Control reactions with light



Thank You for Your Attention

- Femtosecond dynamics are fundamental for physics, chemistry, and biology
- Ultrashort laser pulses can take molecular movies
- Generated in the laboratory or at free-electron lasers
- Understanding a phenomenon gives us the possibility to control it!

Thanks to the supporting institutions



European XFEL / Blue Clay Studios