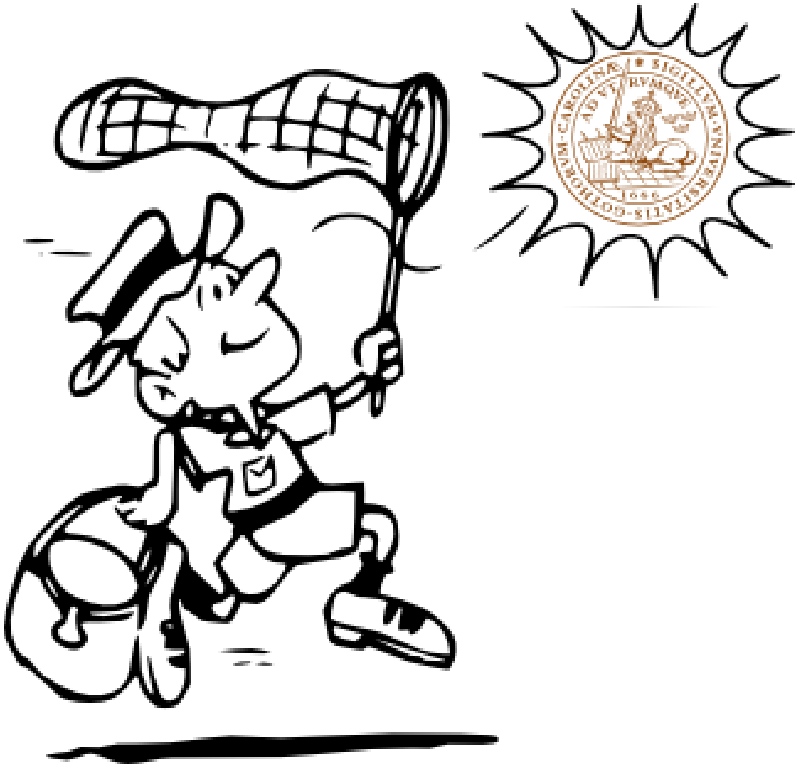
Science for and with laser based x-ray sources

Jens Uhlig et al.
Chemical Physics Lund University

Complementary Spectroscopic Approaches: Numbers



NanoLund
AT THE FOREFRONT OF NANOSCIENCE

LINXS

Lund Laser Centre

NIST
National Institute of Standards and Technology Quantum Devices Group

DTU

MAGYAR TUDOMÁNYOS AKADÉMIA 1825

LCLS

SLAC NATIONAL ACCELERATOR LABORATORY

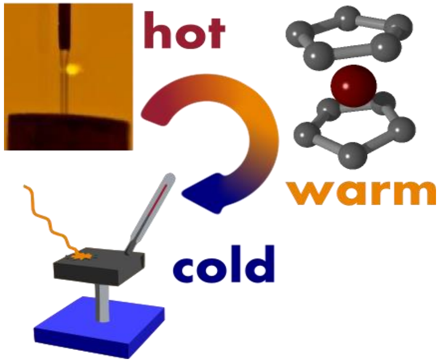
Argonne NATIONAL LABORATORY

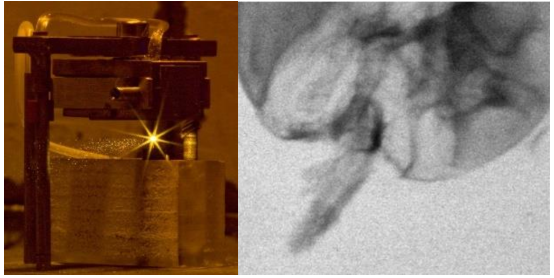
MAX IV

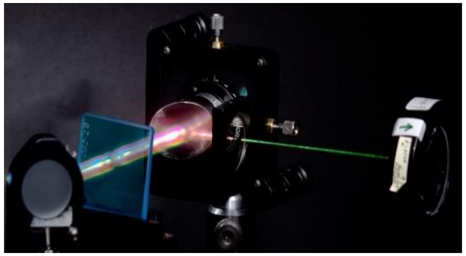
ESRF

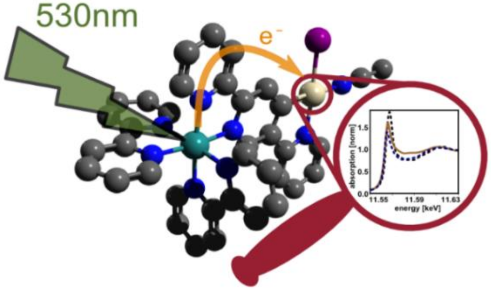
eli | beamlines

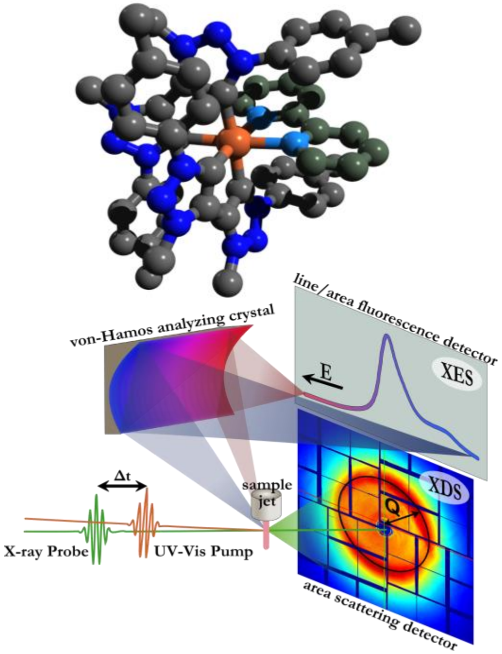
About me

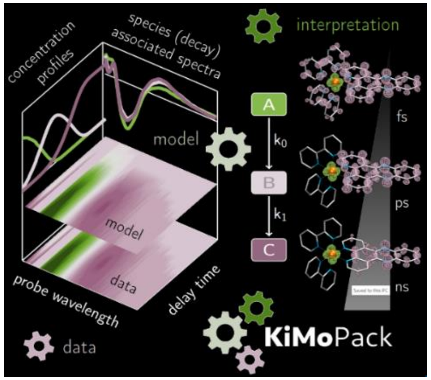

hot
warm
cold






530nm
e⁻
absorption [norm]
energy [keV]
11.55
11.59
11.63


von-Hamos analyzing crystal
line/area fluorescence detector
E
XES
Δt
sample jet
XDS
Q
X-ray Probe
UV-Vis Pump
area scattering detector


concentration profiles
species (decay) associated spectra
interpretation
model
model
data
probe wavelength
delay time
A
k0
B
k1
C
fs
ps
ns
data
KiMoPack

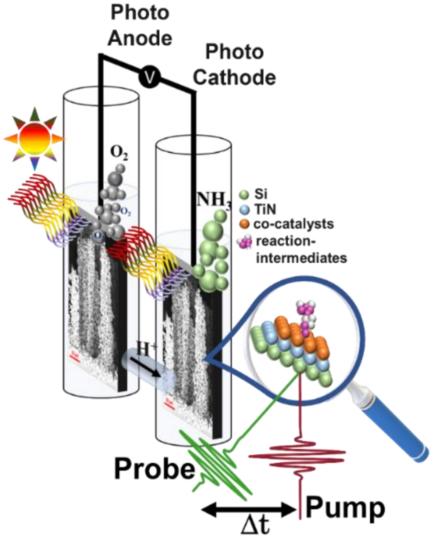
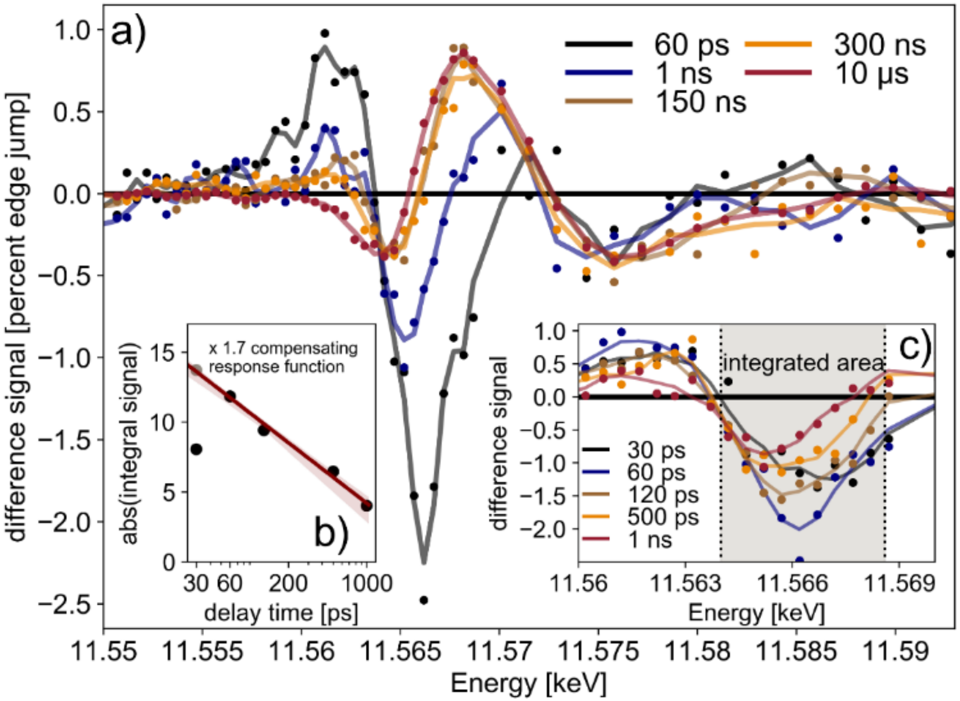
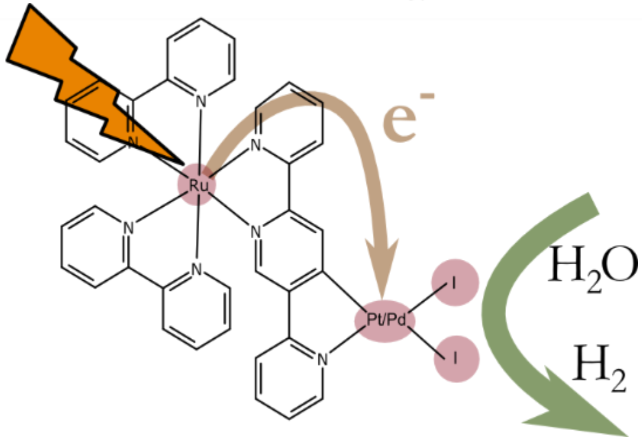
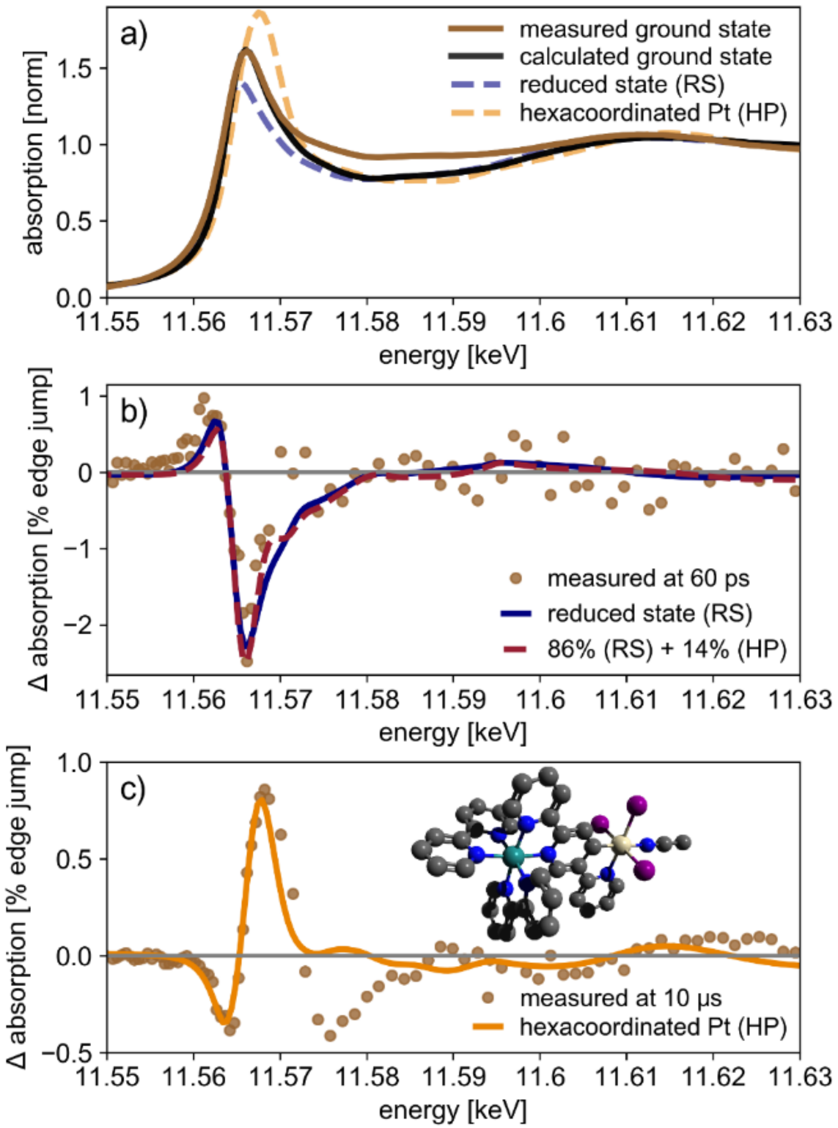


Photo Anode
Photo Cathode
V
O2
NH3
Si
TiN
co-catalysts
reaction-intermediates
H+
Probe
Pump
Δt

The advantage of synchrotrons

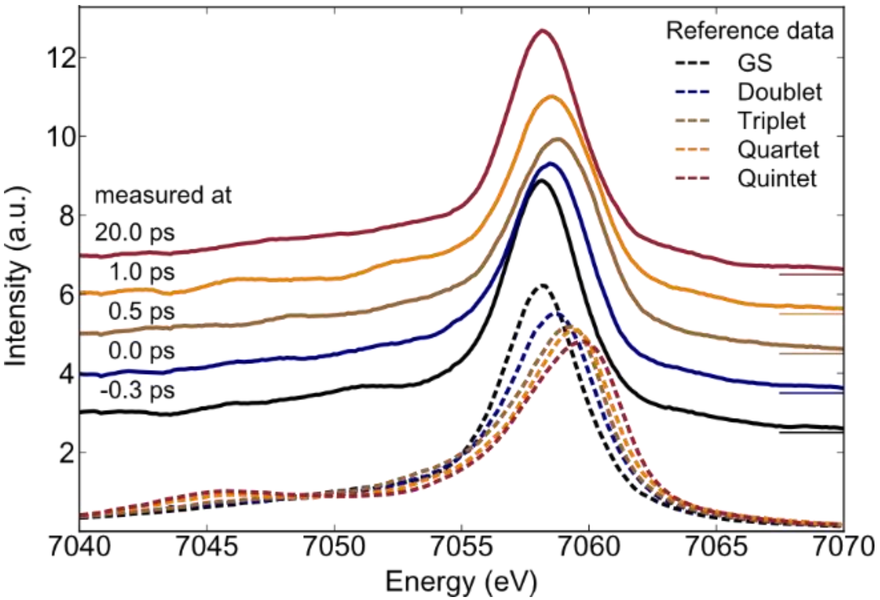


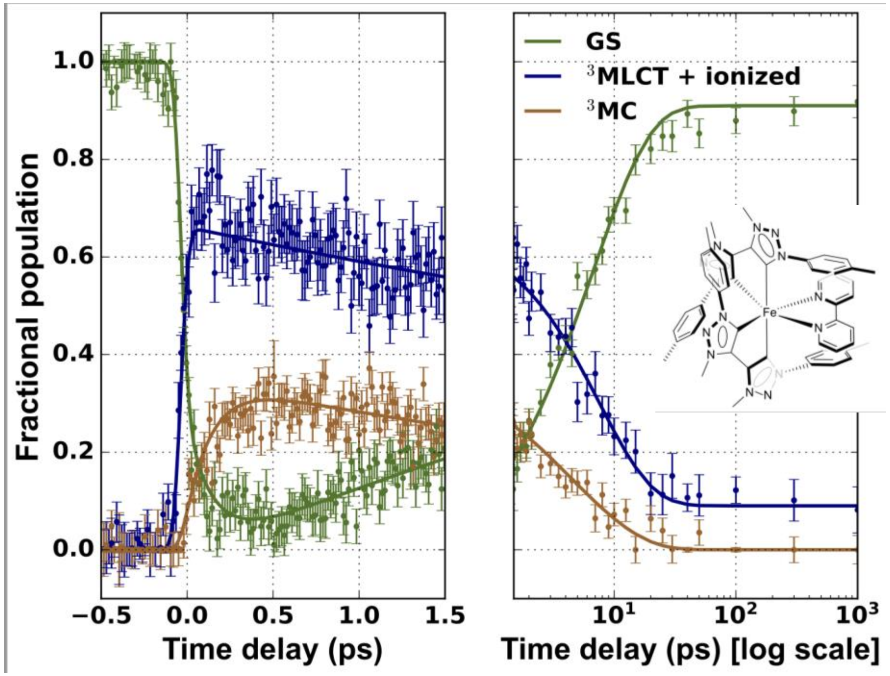


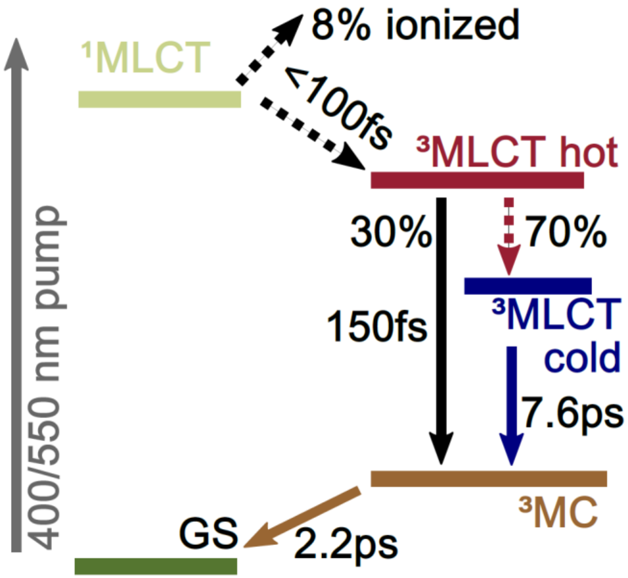


J. Phys. Chem. A 2018, 122, 31, 6396−6406 DOI:10.1021/acs.jpca.8b00916

Hot dynamics







8% ionized

^{1}MLCT

<100fs

^{3}MLCT hot

30%

70%

150fs

^{3}MLCT cold

7.6ps

^{3}MC

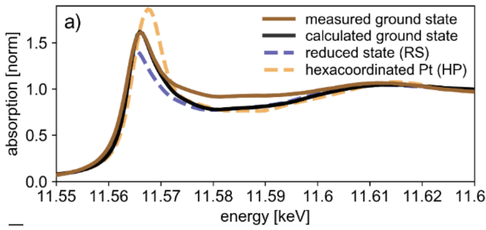
GS

2.2ps

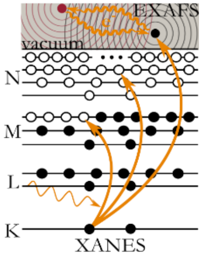
400/550 nm pump

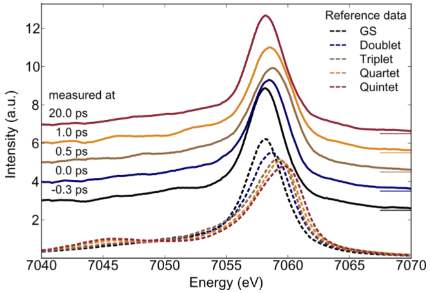
Angewandte Chemistry
https://doi.org/10.1002/anie.201908065

Four very different type of experiments

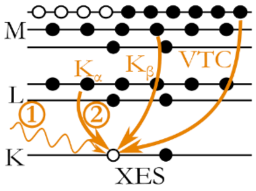


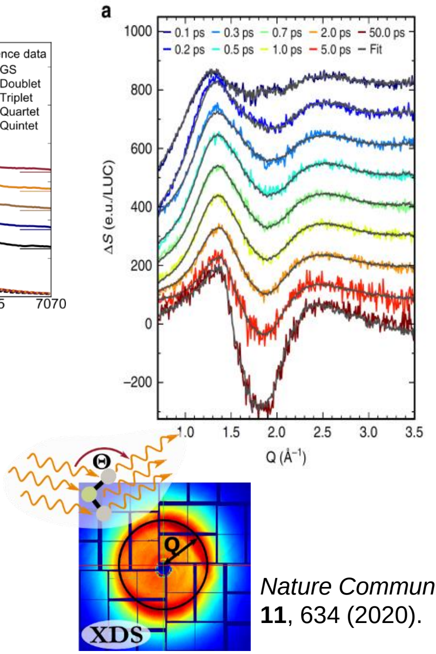
J. Phys. Chem. A 2018, 122, 31, 6396−6406
DOI:10.1021/acs.jpca.8b00916



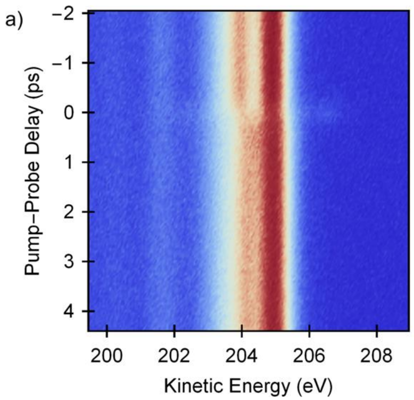


Angewandte Chemistry,
2019, *132*, 372-380

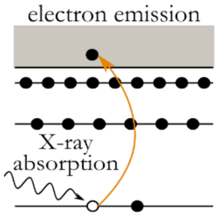


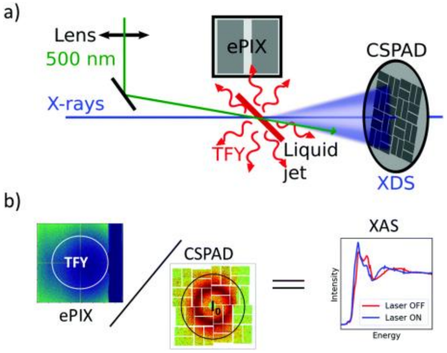


Nature Communications
11, 634 (2020).

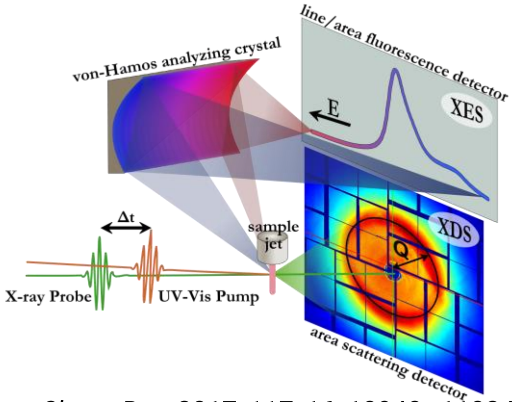


Nat Commun **12**, 1196 (2021).
DOI: 10.1038/s41467-021-21454-3





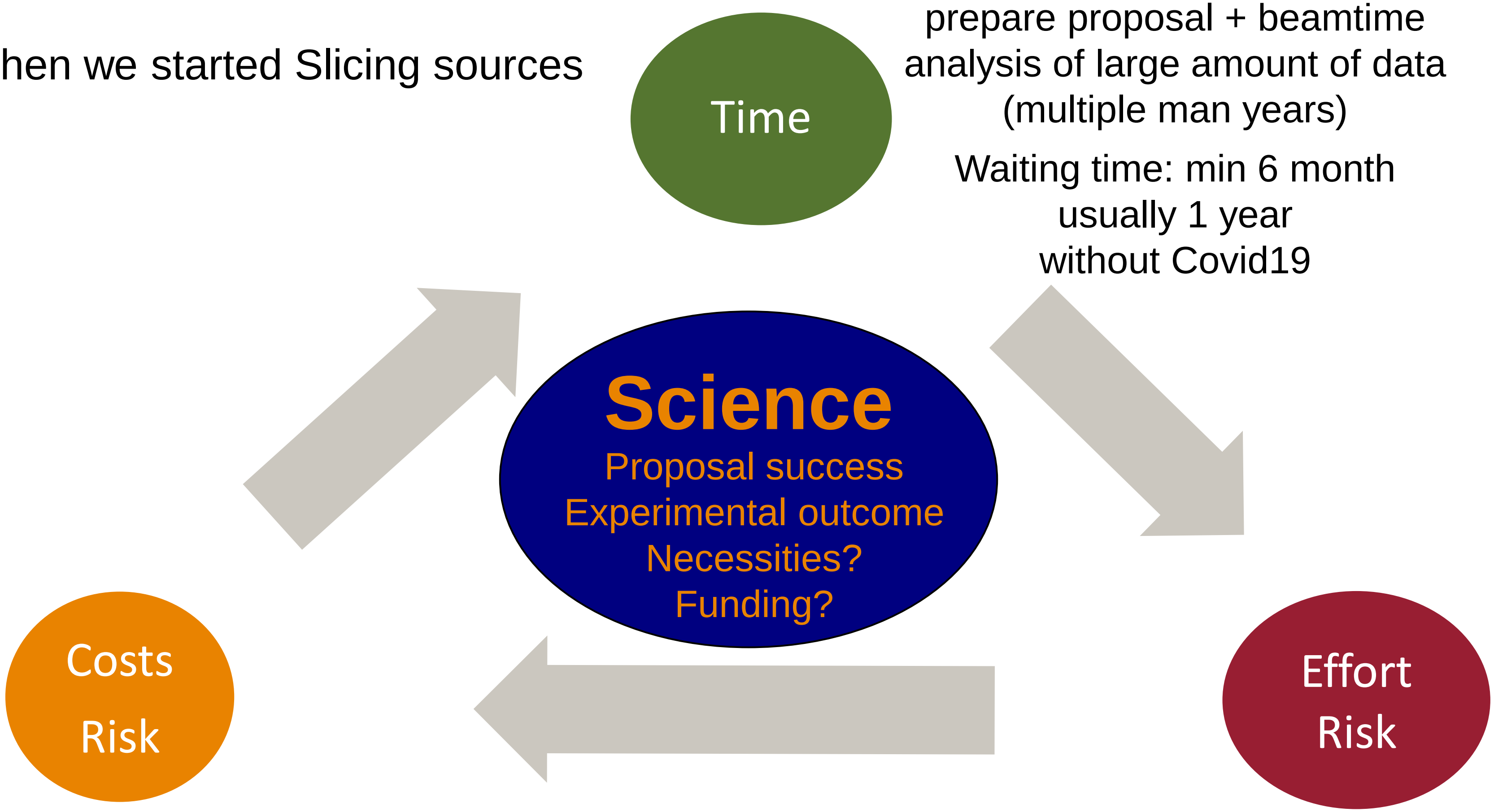
DOI: 10.1039/C9CP03483H
Phys. Chem. Chem. Phys., 2020, **22**, 2660-2666



Chem. Rev. 2017, 117, 16, 10940−11024
DOI: 10.1021/acs.chemrev.6b00807

Always start with WHY?

When we started Slicing sources



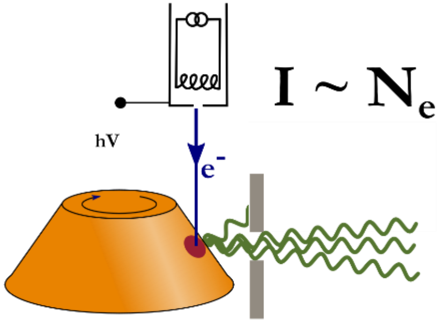
prepare proposal + beamtime
analysis of large amount of data
(multiple man years)

Waiting time: min 6 month
usually 1 year
without Covid19

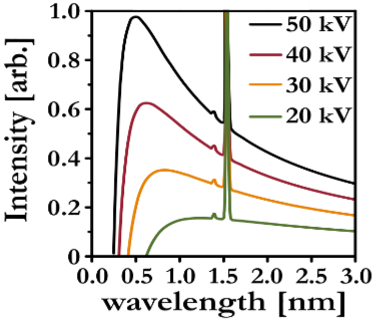
Time-resolved experiments XFEL:
10 persons 1 week USA = 40 kEuro
Facility 2-3 MEuro / beamtime

Proposal (>one week) + Preparation
Time-resolved experiments for multiple days:
1g sample at 20mg/week and person?
Damage? Success?
No guarantee of proposal success (Grants?)

Lab – based (steady state) - sources



$$I \sim N_e$$

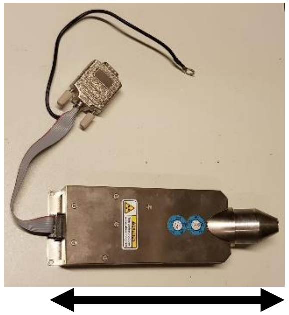


$$I(\lambda)d\lambda = \frac{K\,i\,Z}{\lambda^2}\left(\frac{\lambda}{\lambda_{max}} - 1\right)$$

Limit? Melting of copper!
20kV * 40mA on 100µm x 100µm close to 50% of melting power
Lamp 200V, 4A = 800W lamp

Brilliance photons /s / mm^2 /mrad^2

Micro-focus sealed tube	2 x 10^9	(70x70 micron)	
Micro-focus rotating anode	12 x 10^9	(80x80 micron)	undulator
Liquid metal jet	26 x 10^9	(20x20 micron)	1 x 10^{14}



15 cm

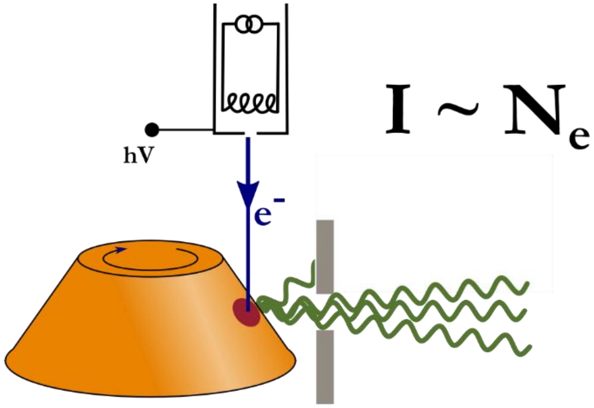
In characteristic emission lines or full spectrum (assume a few keV bandwidth)
corresponding:1 x 10^5 ph /s / mm^2 /mrad^2 /eV

undulator
1 x 10^{13}

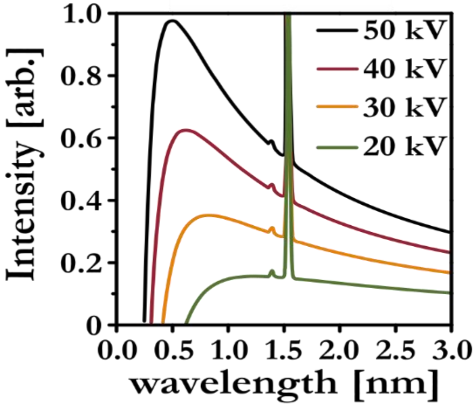
How to use:
efficient detection, experiment design

T. Skarzynski, “Collecting data in the home laboratory: evolution of X-ray sources, detectors and working practices,” Acta Crystallographica Section D Biological Crystallography, vol. 69, no. 7, Art. no. 7, Jun. 2013, doi: 10.1107/s0907444913013619

X-ray tube bremsstrahlung



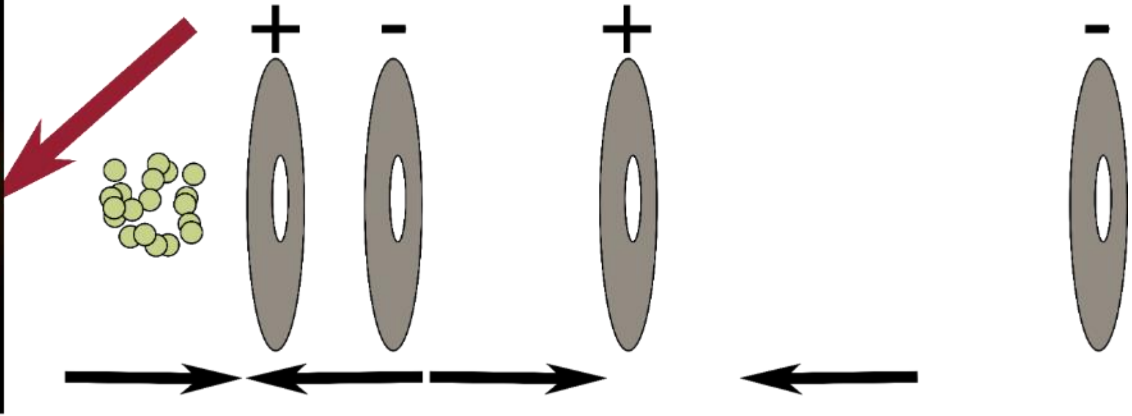
$I \sim N_e$

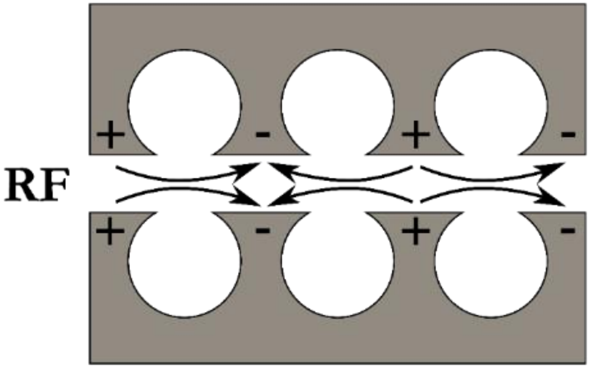


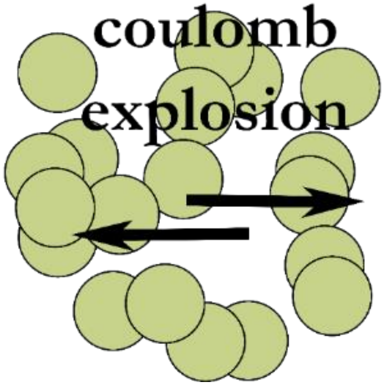
microsource
rotating anode
liquid metal jet
plasma sources

un-polarized

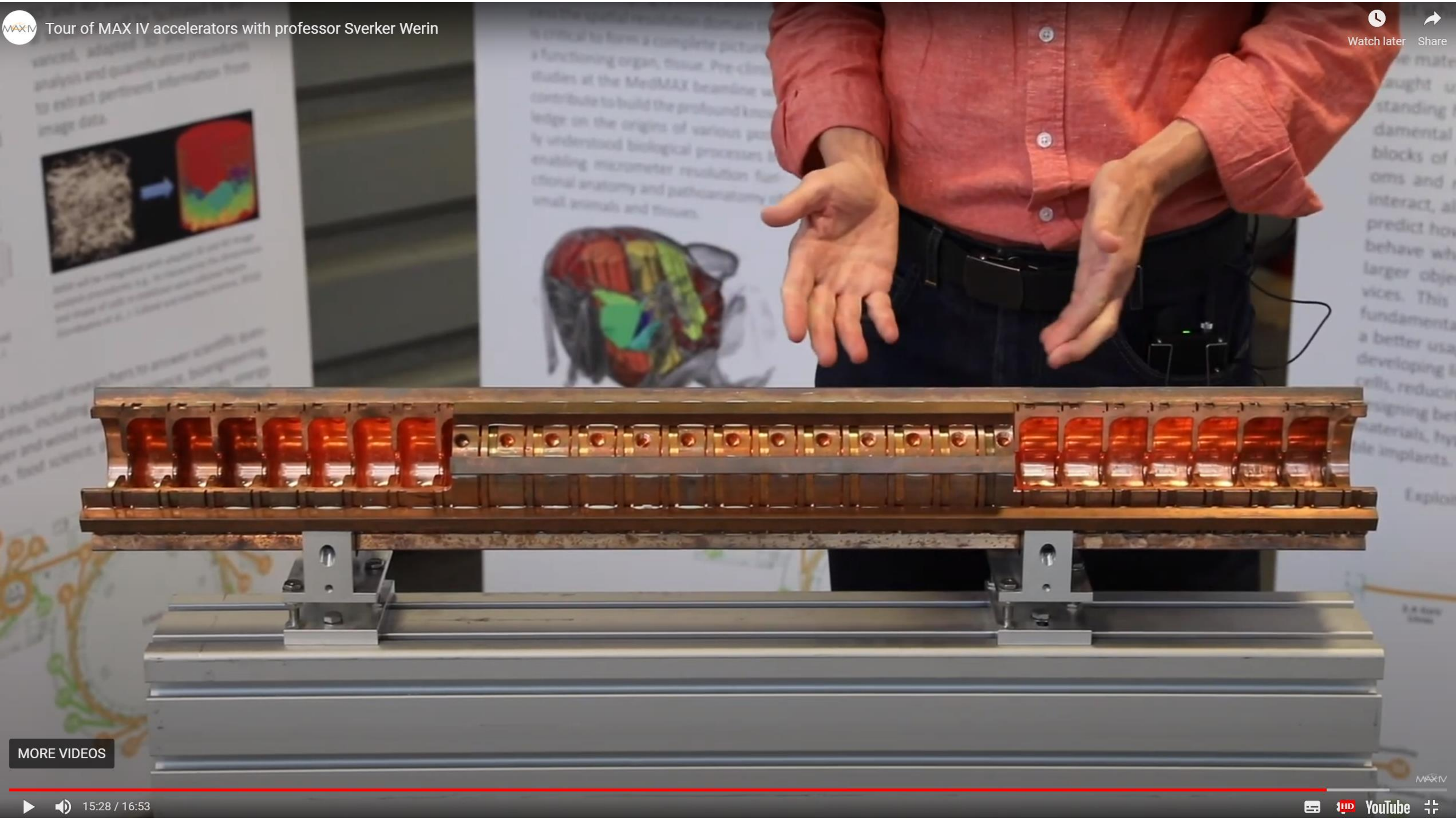
accelerating electrons



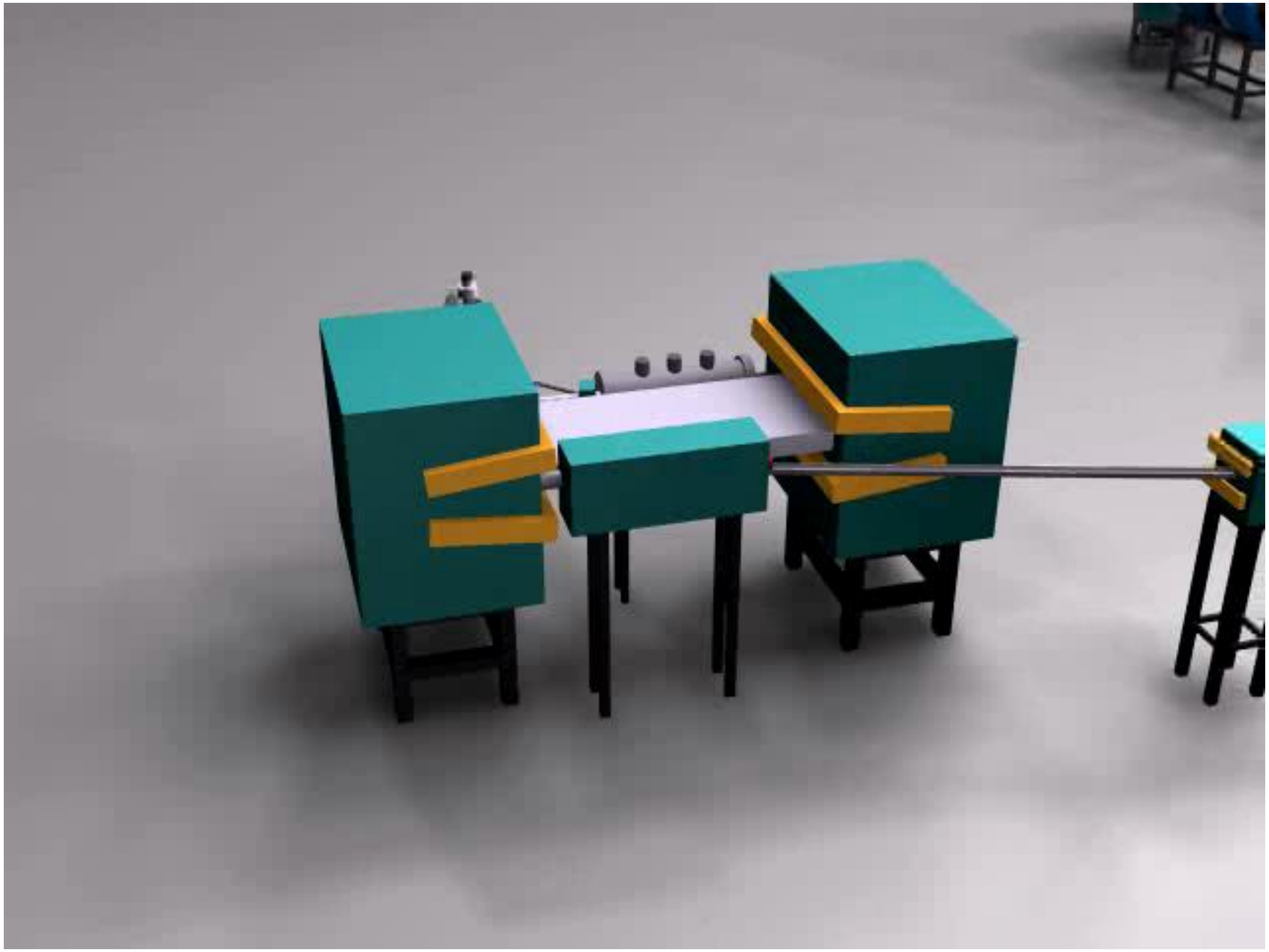




clystron


Tour of MAX IV accelerators with professor Sverker Werin
Watch later
Share
MORE VIDEOS
15:28 / 16:53
HD
YouTube
MAX IV

Video Circulating electrons

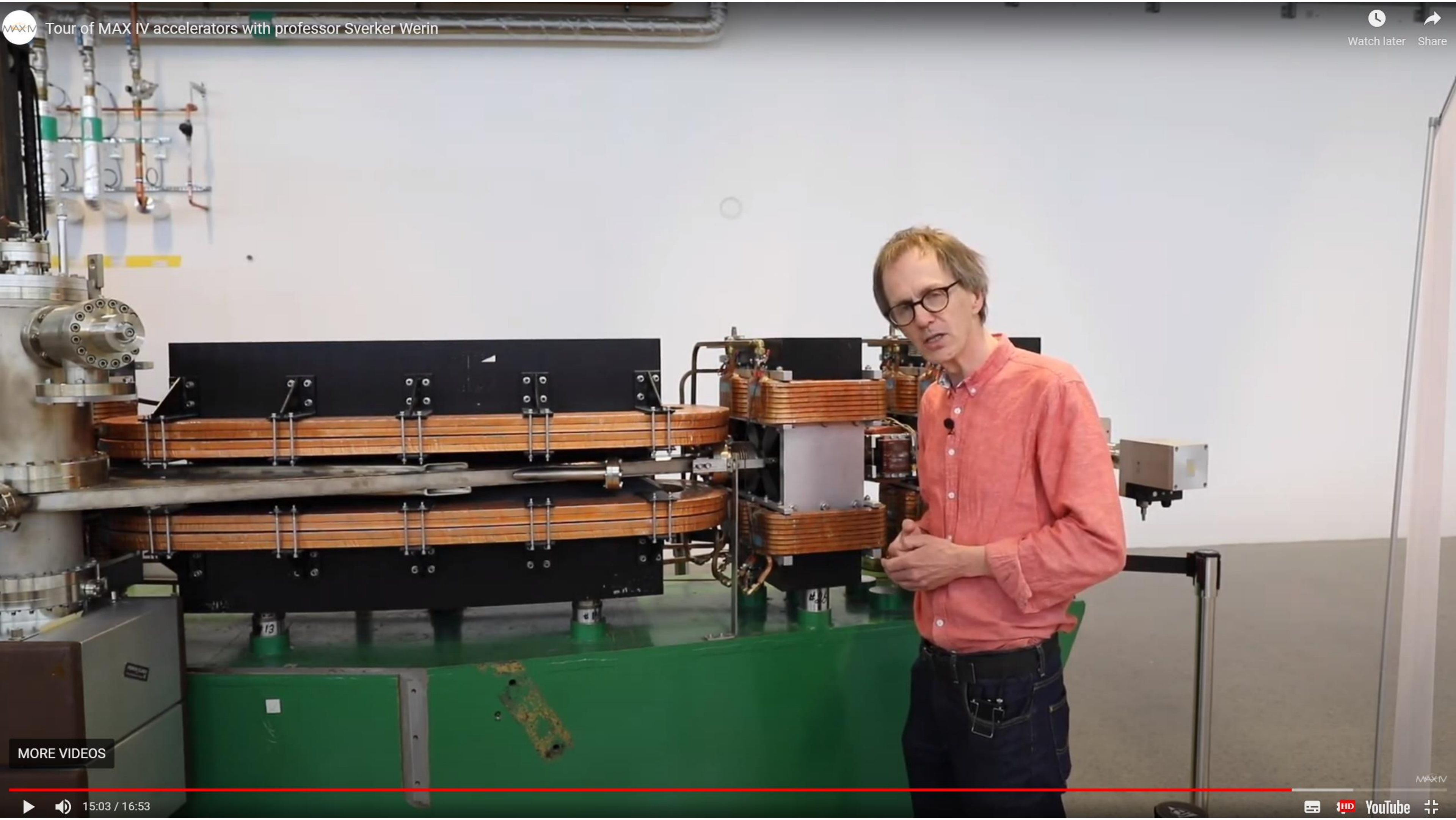


ISA, Centre for Storage Ring Facilities, Aarhus
https://www.isa.au.dk/animations/animations.asp
Accessed May 11 2020

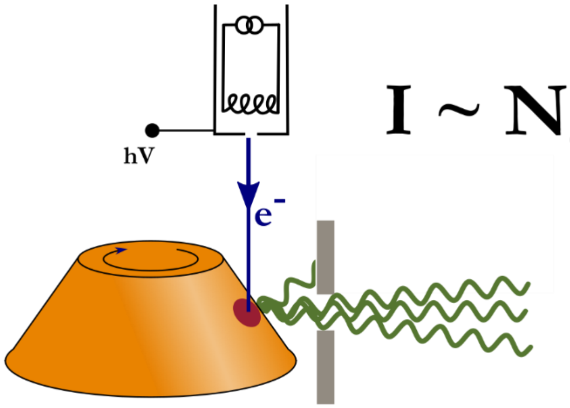
Video Insertion devices tangential beam

ISA, Centre for Storage Ring Facilities, Aarhus
https://www.isa.au.dk/animations/animations.asp
Accessed May 11 2020

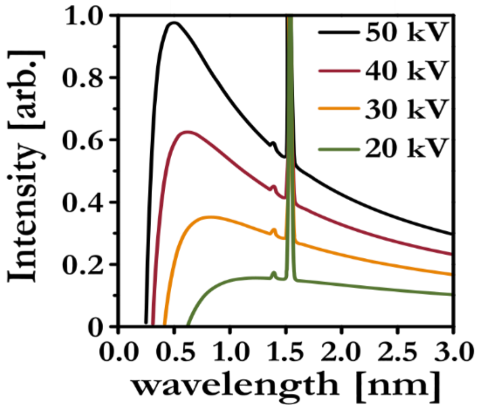
Old Bending magnet



Bending magnet

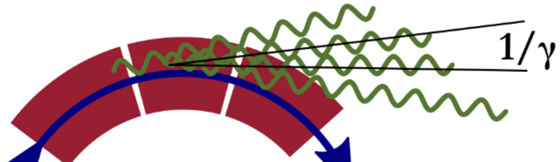


$I \sim N_e$



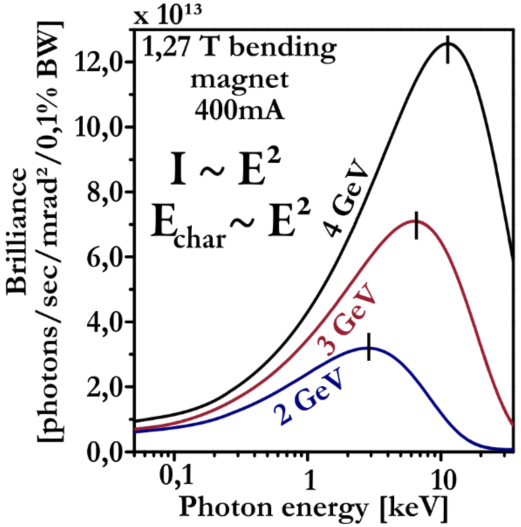
microsource
rotating anode
liquid metal jet
plasma sources

un-polarized



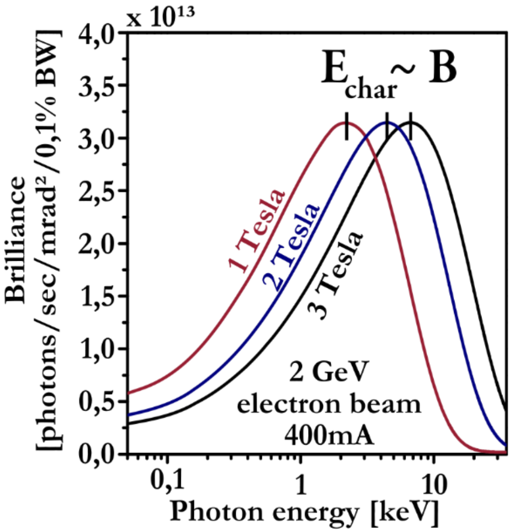
$I \sim N_e$

Polarized



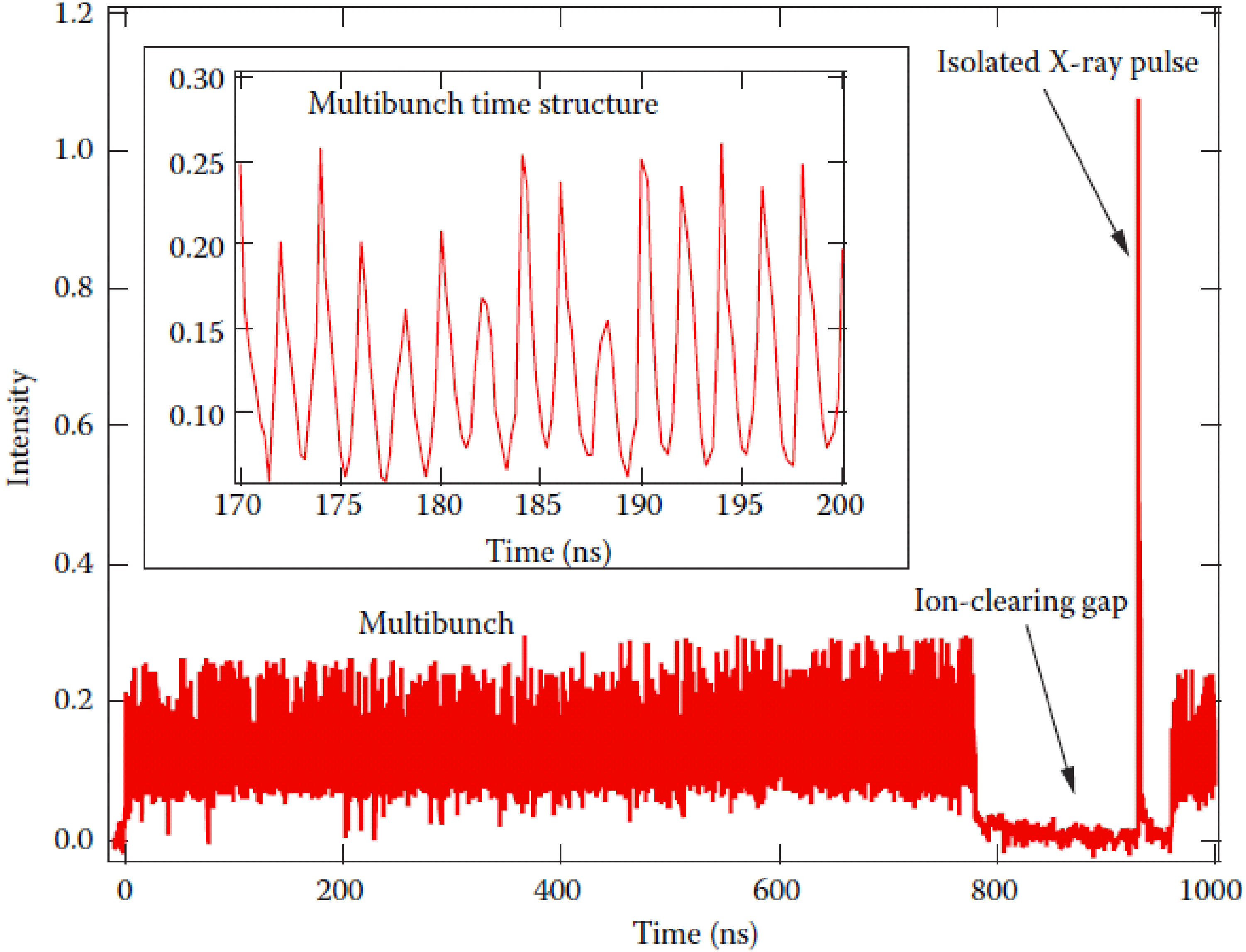
$I \sim E^2$

$E_{char} \sim E^2$

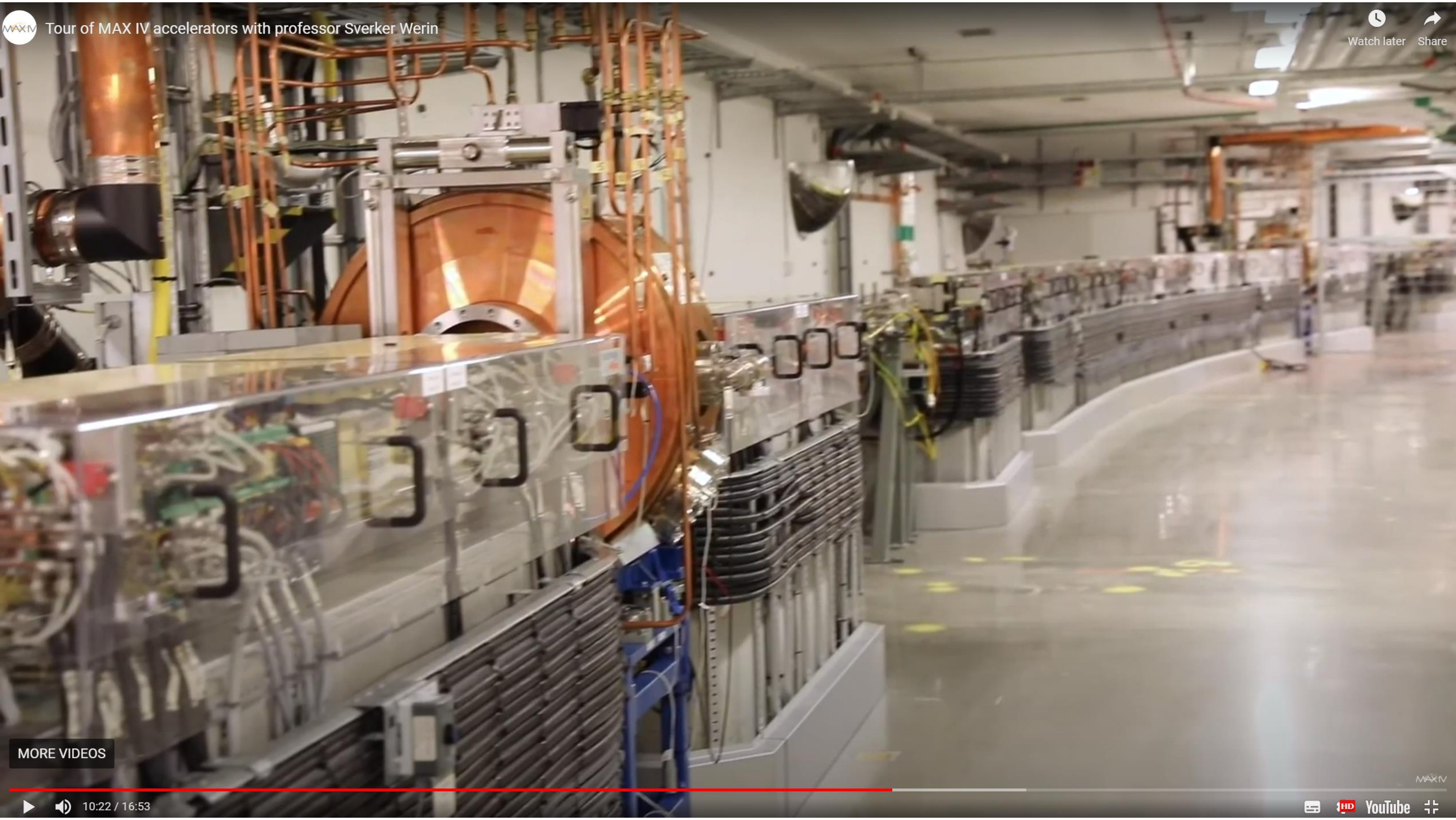


$E_{char} \sim B$

Bunch modes


Multibunch time structure
0.30
0.25
0.20
0.15
0.10
170
175
180
185
190
195
200
Time (ns)
Isolated X-ray pulse
Multibunch
Ion-clearing gap
Intensity
1.2
1.0
0.8
0.6
0.4
0.2
0.0
0
200
400
600
800
1000
Time (ns)

Look into ring

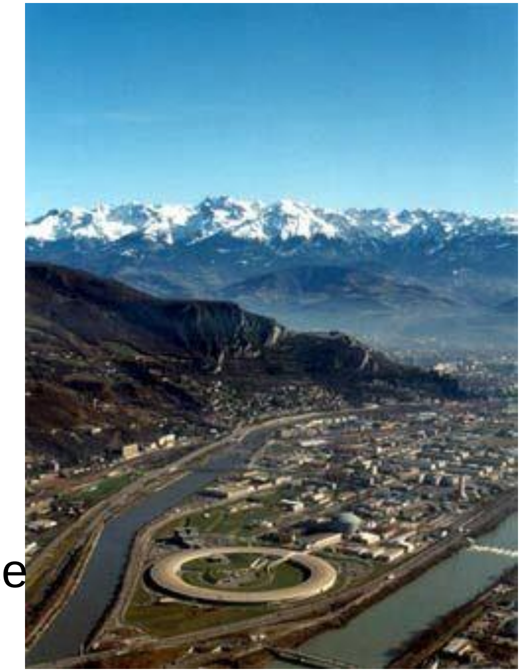


Synchrotrons

MaxIV, Lund 3 & 1.5 GeV ring

APS Chicago 6 GeV ring
100ps 10^13ph/s

8 GeV ring
SACLA 1ps 10^12ph/s
Osaka/Japan



ESRF Grenoble /France
6 GeV ring
100ps 10^14ph/s

Video Insertion devices tangential beam

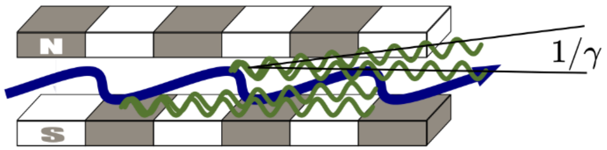
ISA, Centre for Storage Ring Facilities, Aarhus
https://www.isa.au.dk/animations/animations.asp
Accessed May 11 2020

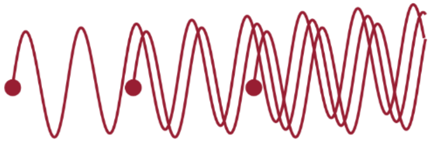
Photo Soleil, beamlines

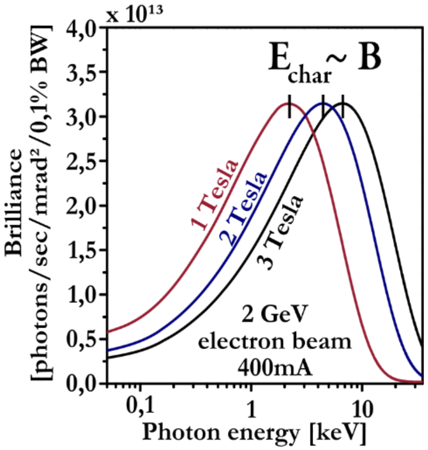
Look onto Undulator

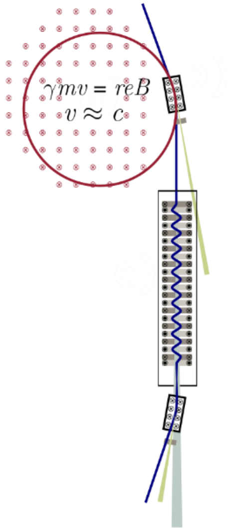
Wiggler and Undulator

$$I \sim N_{magnets} * N_e$$



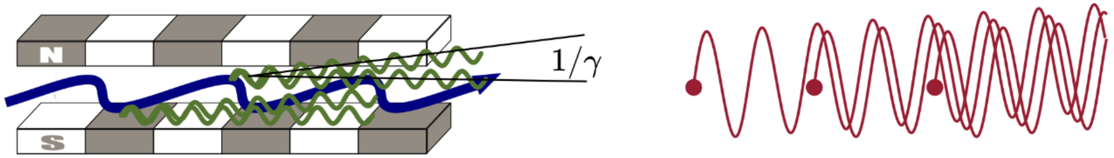




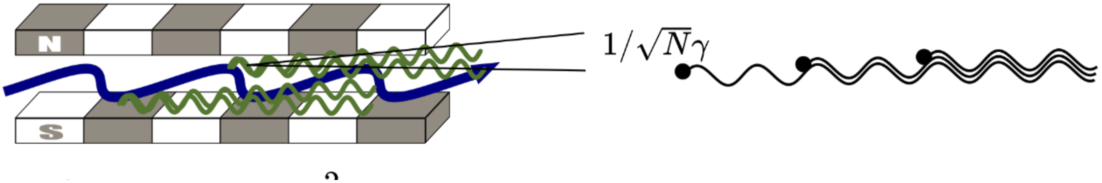


Wiggler and Undulator

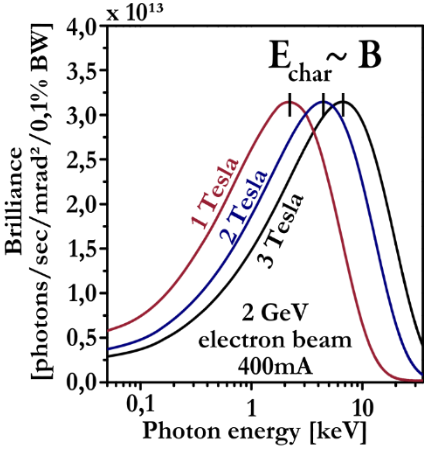
$$I \sim N_{magnets} * N_e$$

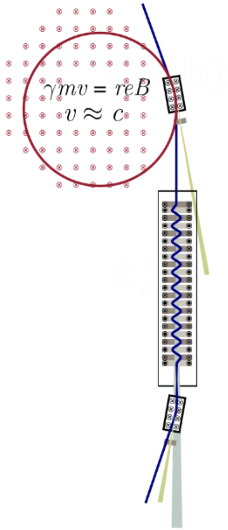


$$I \sim N^2_{magnets} * N_e$$



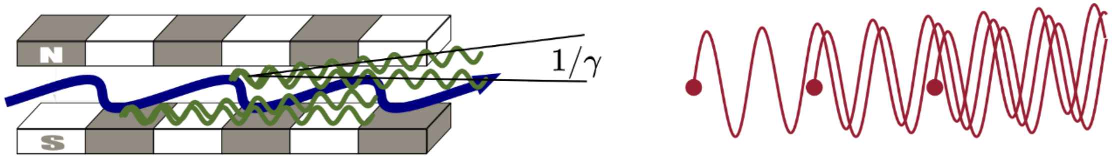
6 GeV
$\gamma=10^4$
magnets 1.5cm
Λ = 0.8Å
deflection = 0.2 μm

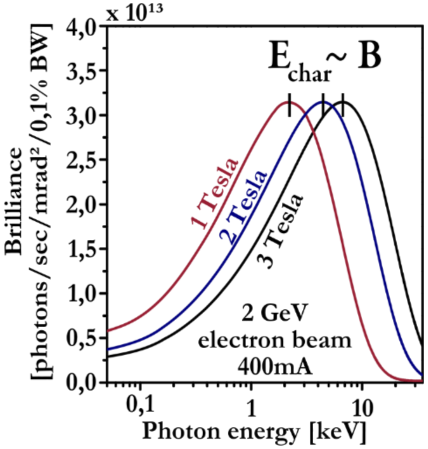




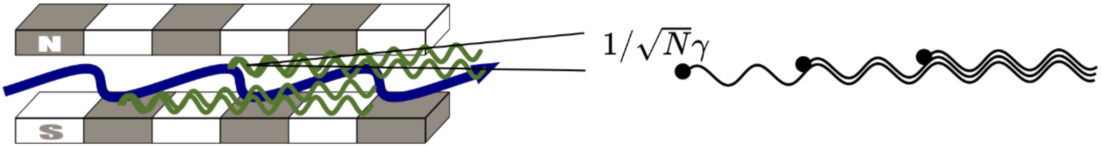
Wiggler and Undulator

$$\mathbf{I} \sim \mathbf{N}_{\mathbf{magnets}} * \mathbf{N}_{\mathbf{e}}$$





$$\mathbf{I} \sim \mathbf{N}^2_{\mathbf{magnets}} * \mathbf{N}_{\mathbf{e}}$$



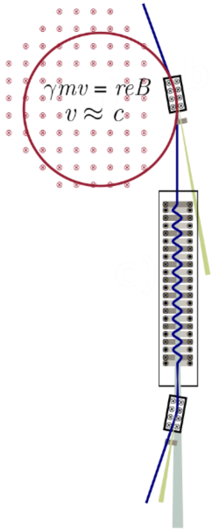
6 GeV

γ=10^4

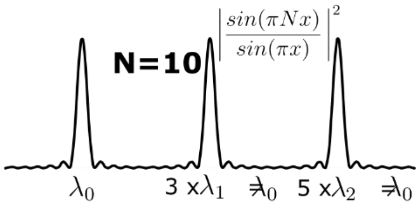
magnets 1.5cm

Λ = 0.8Å

deflection = 0.2 µm



$$\lambda_{resonance} \sim B_0^2$$



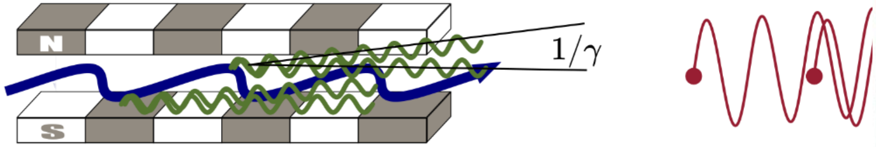
$$E_{single\ electron} = \sum_{i=1}^{N_{magnets}} \cos(\omega t + \phi_i)$$

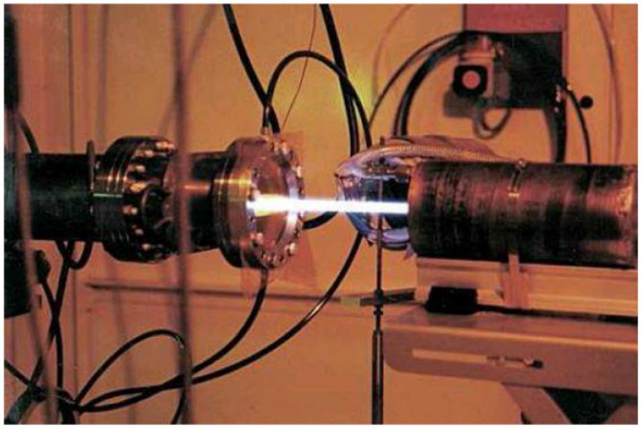
$$I_{single\ electron} = E^2 = \sum_{i=1}^{N_{magnets}} \cos(\omega t + \phi_i)^2 + \sum_{i \neq j}^{N_{magnets}} \cos(\omega t + \phi_i)\cos(\omega t + \phi_j)$$

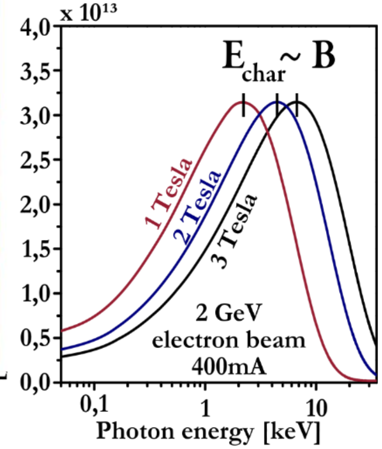
$$K = \frac{e\lambda_u B_0}{2\pi c m_e}$$

Wiggler and Undulator

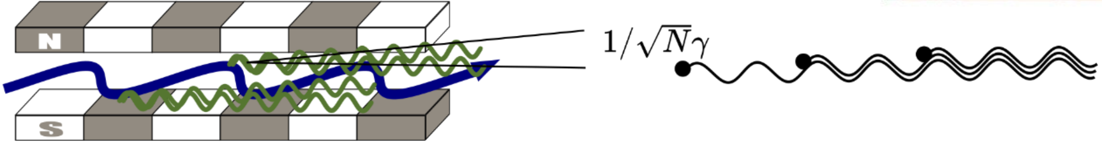
$$I \sim N_{magnets} * N_e$$





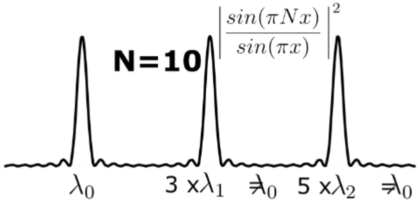


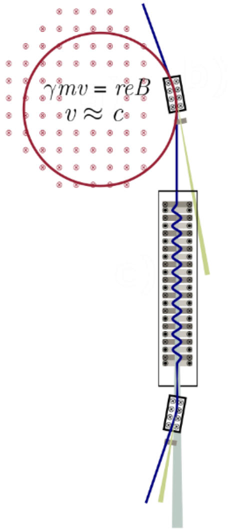
$$I \sim N^2_{magnets} * N$$



magnets 1.5cm
Λ = 0.8Å
deflection = 0.2 μm

$$\lambda_{resonance} \sim B_0^2$$

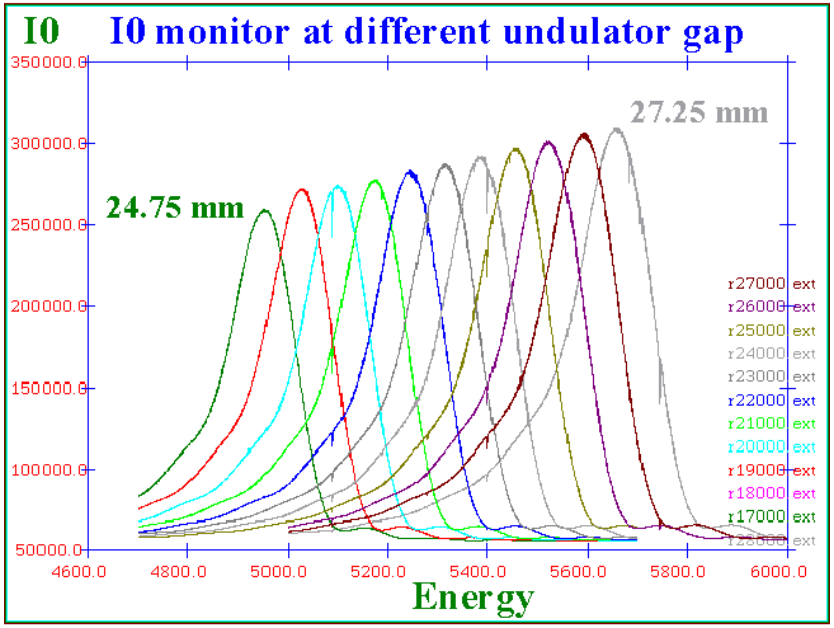




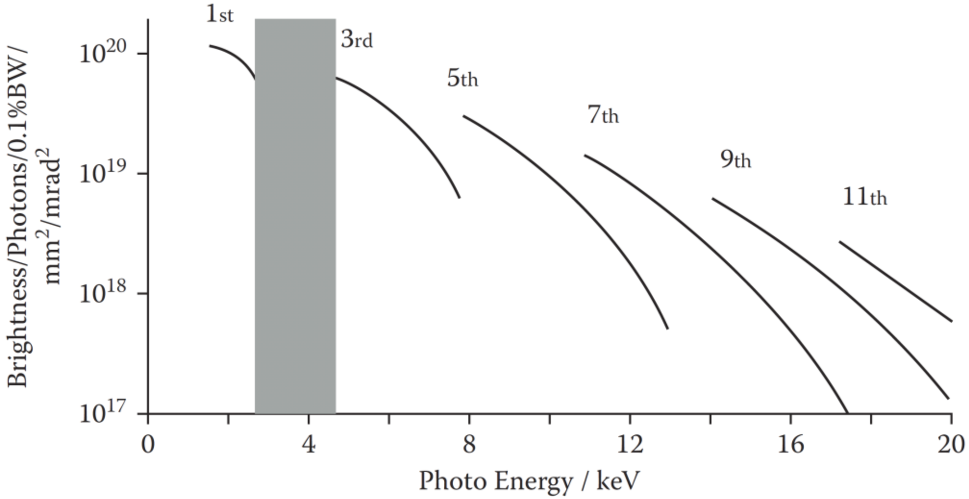
$$E_{single\ electron} = \sum_{i=1}^{N_{magnets}} \cos(\omega t + \phi_i)$$

$$I_{single\ electron} = E^2 = \sum_{i=1}^{N_{magnets}} \cos(\omega t + \phi_i)^2 + \sum_{i \neq j}^{N_{magnets}} \cos(\omega t + \phi_i)\cos(\omega t + \phi_j)$$

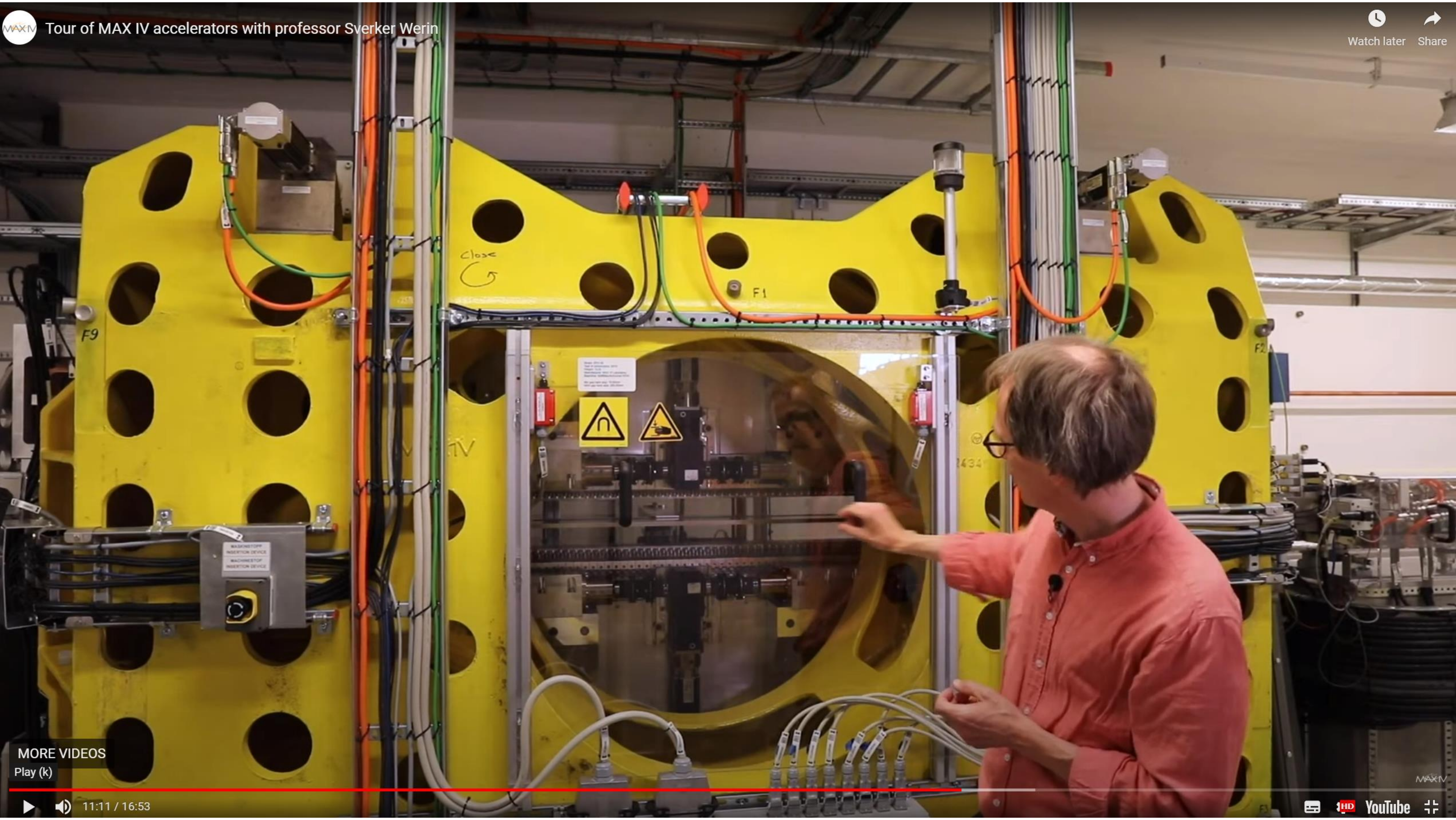
Undulator spectrum



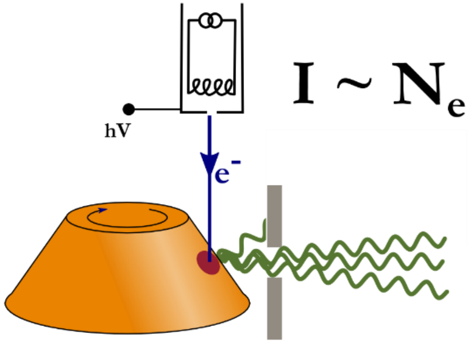
ESRF ID26
https://www.esrf.eu/home/UsersAndScience/Experiments/EMD/ID26/Characteristics/ScanningModes.html



Look onto undulator from outside

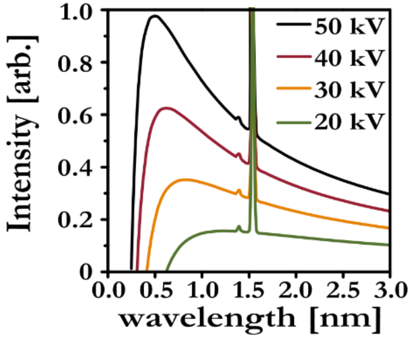


Sources 1st-3rd generation

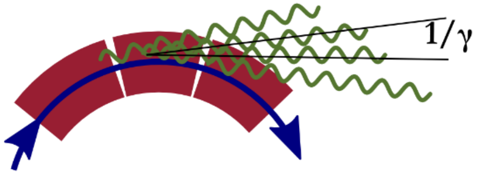


$I \sim N_e$

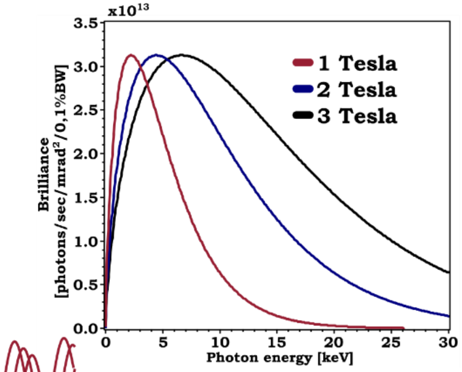
un-polarized



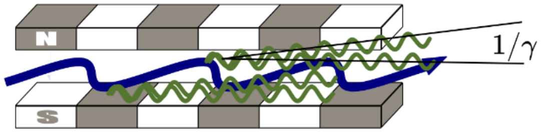
$I \sim N_e$

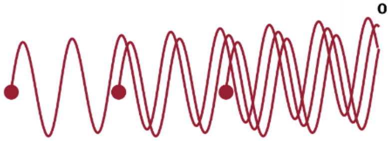


Polarized



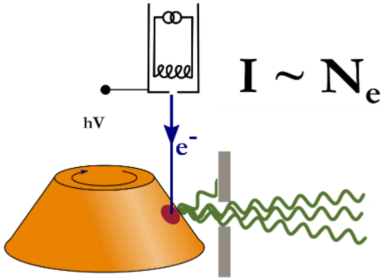
$I \sim N_{magnets} * N_e$

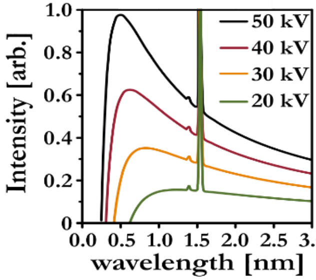




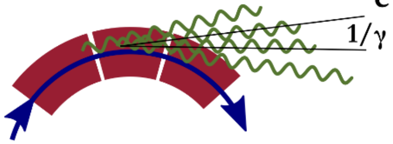
Synchrotron sources

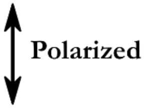
$$I \sim N_e$$

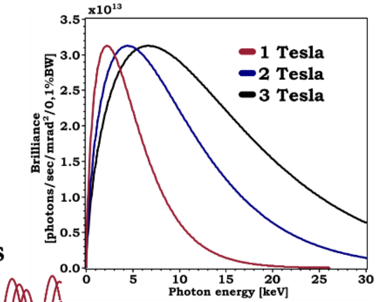




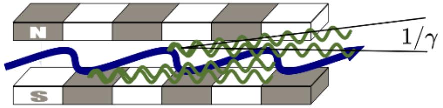
$$I \sim N_e$$

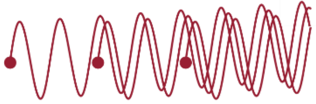




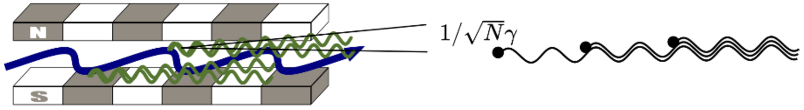


$$I \sim N_{magnets} * N_{electrons}$$

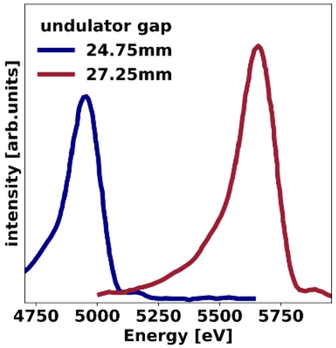




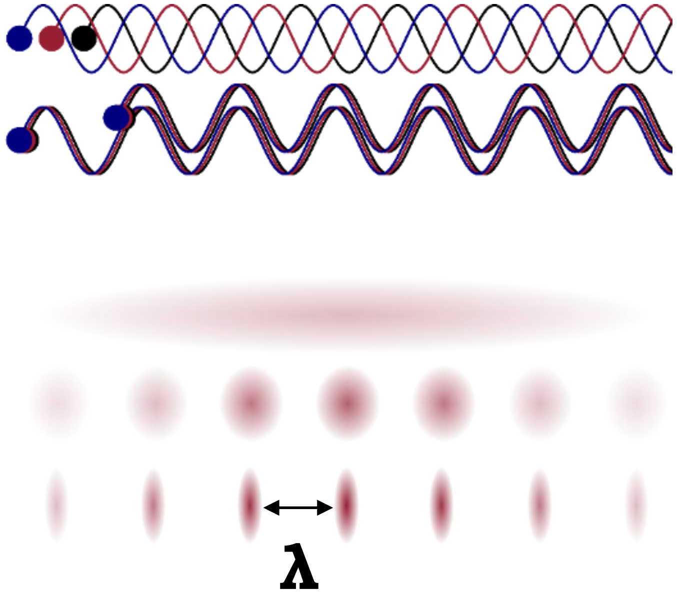
$$I \sim N^2_{magnets} * N_{electrons}$$

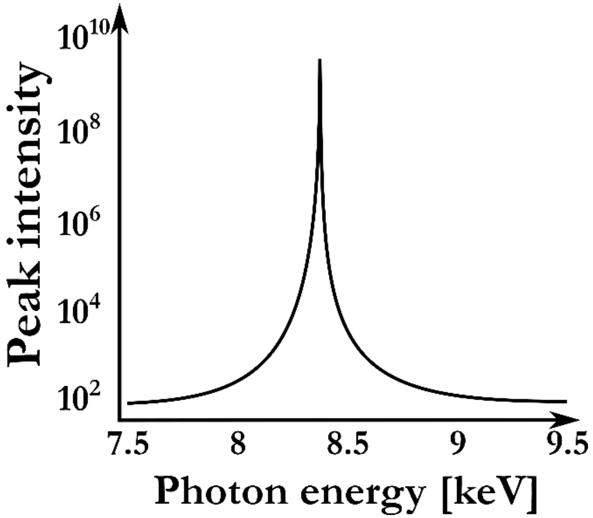


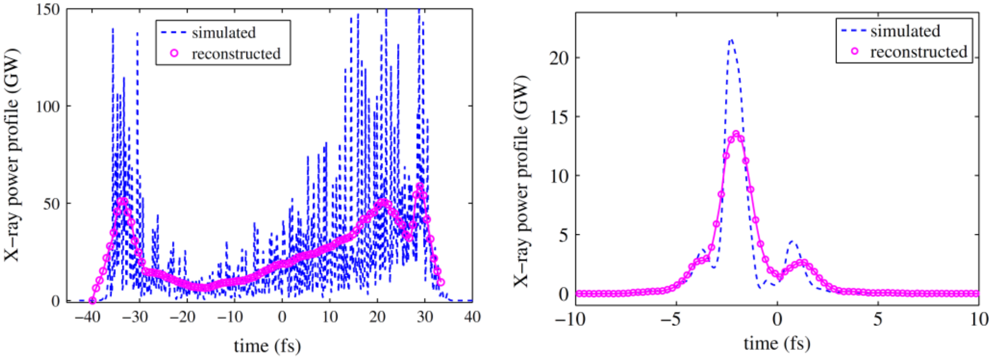
$$\lambda_{resonance} \sim B_0^2$$



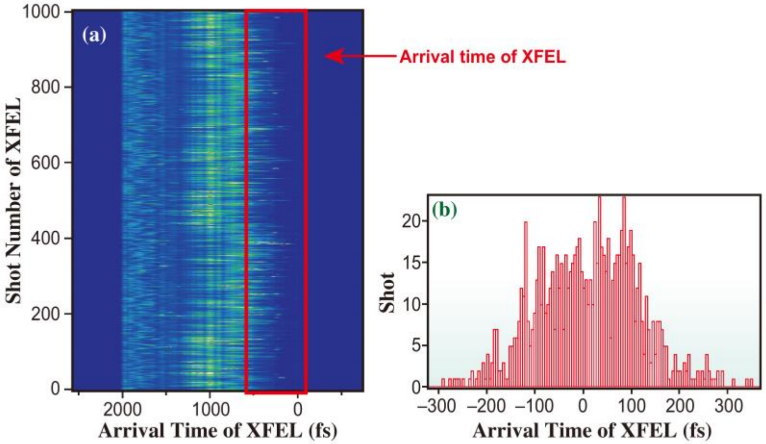
We create structure in the electron bunches







Y. Ding, C. Behrens, P. Emma, J. Frisch, Z. Huang, H. Loos, P. Krejcik, M.-H. Wang, *Physical Review Special Topics - Accelerators and Beams*. 2011, *14* (12). DOI: 10.1103/physrevstab.14.120701.

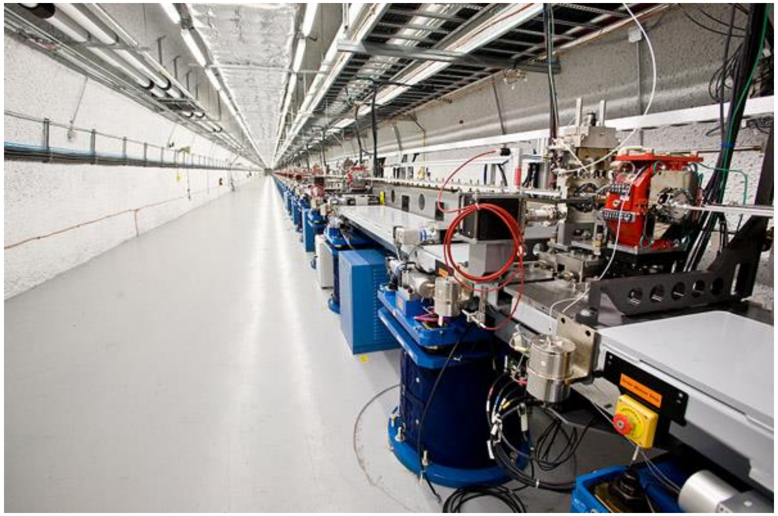


SACLA Research Frontiers 2015 , Sato, T. and Yabashi, M.
http://www.spring8.or.jp/pdf/en/res_fro/15/128_129.pdf

$$I_{undulator + modulated\ electron\ bunch} \sim N_{magnets}^{2} * N_{electrons}^{1<x<2}$$

Short pulse sources

Swizzfel 50fs 10^12ph/s
Villigen, close to Zurich/Switzerland



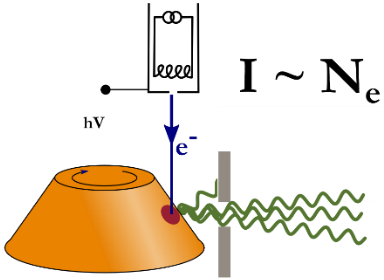
3km linac 100fs 10^12ph/s
LCLS Stanford /California

European XFEL 50fs 10^17ph/s
Hamburg

FEMTOMAX at MAXIV 100fs 10^7ph/s
scanable, Lund

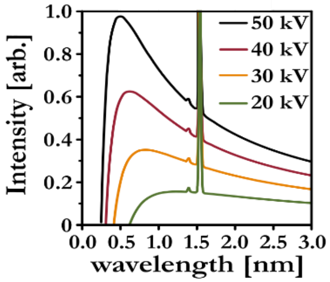
Sources

ph/s (in 1eV)



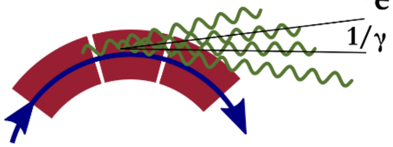
$I \sim N_e$

un-polarized

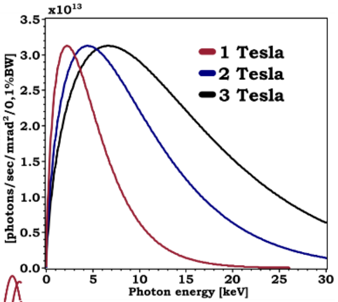


10^9
Excillium
10^{11}

$I \sim N_e$

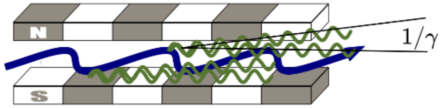


Polarized



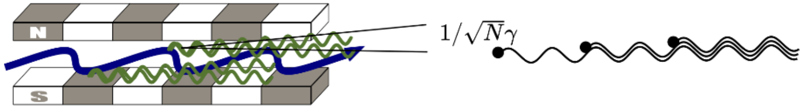
10^{11}

$I \sim N_{magnets} * N_{electrons}$

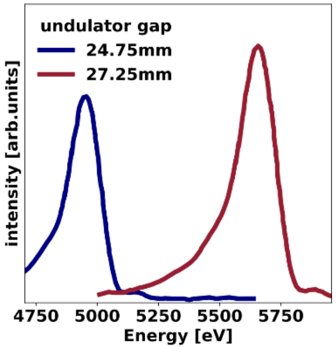


10^{13}

$I \sim N^2_{magnets} * N_{electrons}$

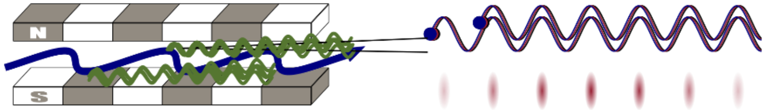


$\lambda_{resonance} \sim B_0^2$



10^{14}

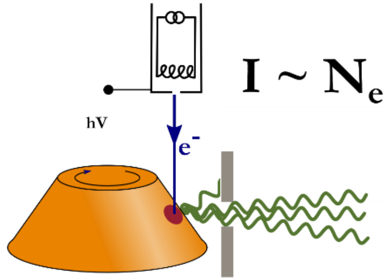
$I \sim N^2_{magnets} * N^{1\text{-}2}_{electrons}$



$\lambda_{resonance} \sim B_0^2$

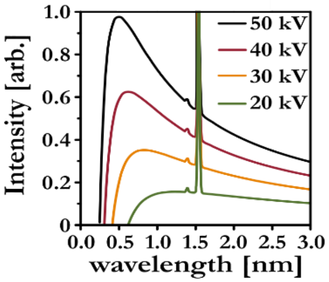
10^{13}

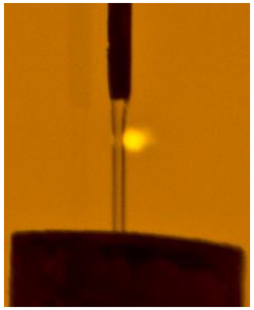
Accellerator based sources



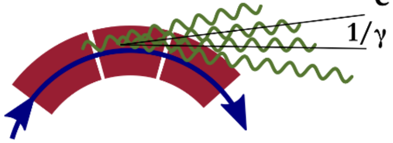
$I \sim N_e$


un-polarized

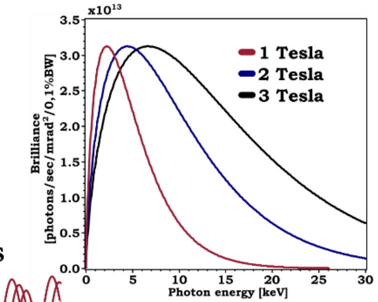




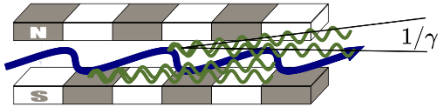
$I \sim N_e$

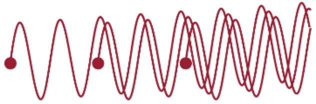


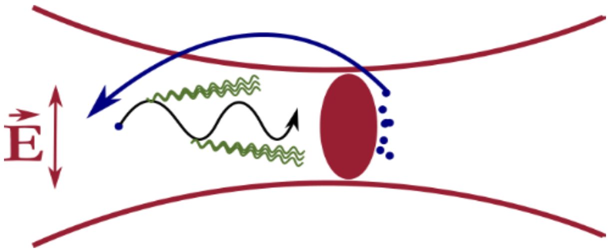

Polarized



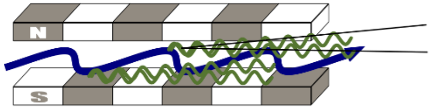
$I \sim N_{magnets} * N_{electrons}$



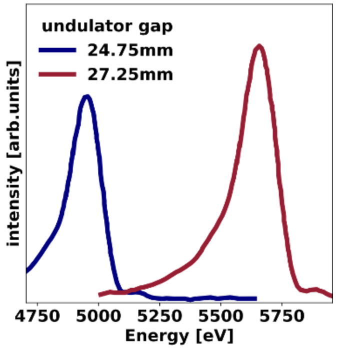


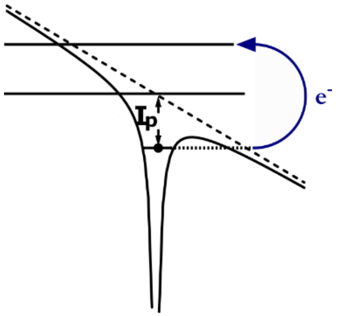


$I \sim N^2_{magnets} * N_{electrons}$

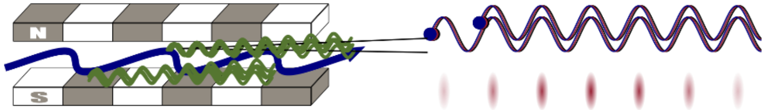


$\lambda_{resonance} \sim B_0^2$





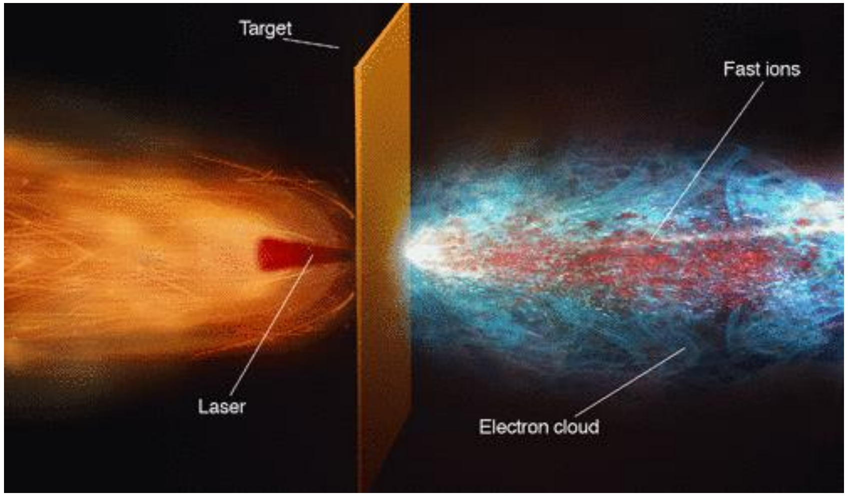
$I \sim N^2_{magnets} * N^{1\text{-}2}_{electrons}$

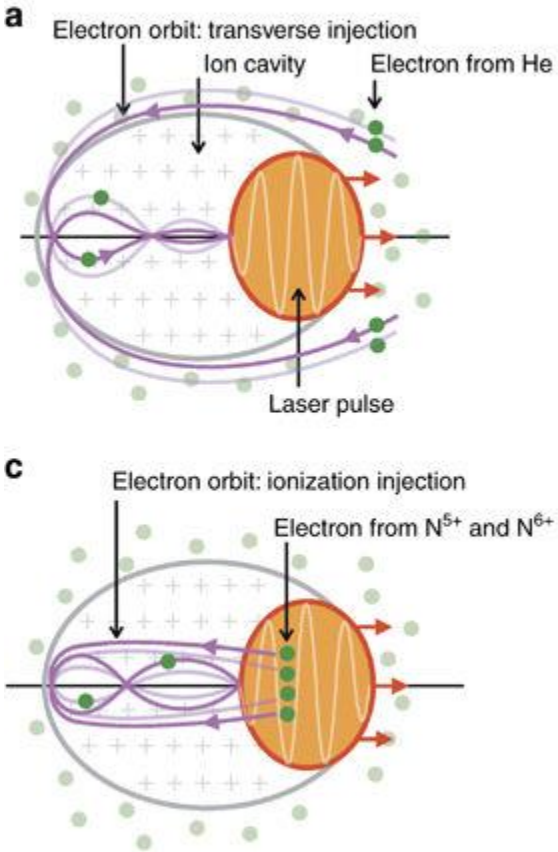


$\lambda_{resonance} \sim B_0^2$

Betatron 1

2-3 J in 50 fs

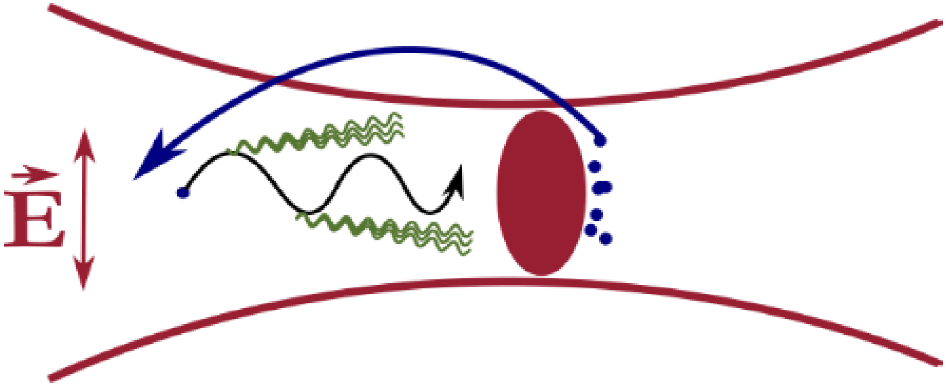


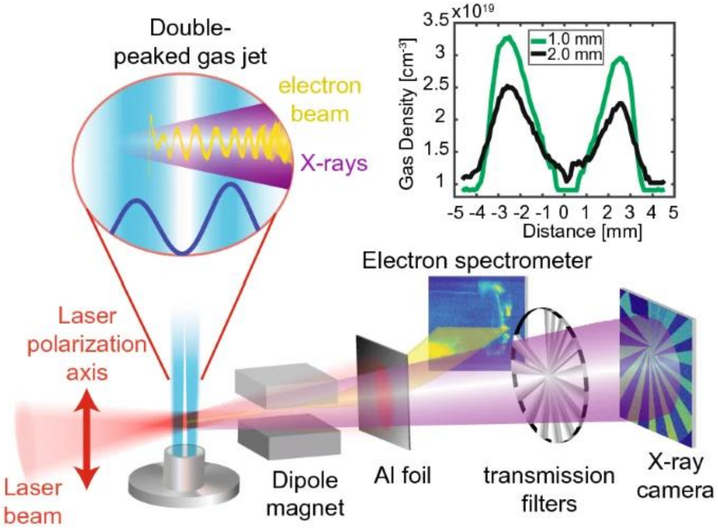


A. Macchi, M. Borghesi and M. Passoni, Ion acceleration by superintense laser-plasma interaction, *Reviews of Modern Physics, 2013,* ***85, 751–793.***

A. Döpp, B. Mahieu, A. Lifschitz, C. Thaury, A. Doche, E. Guillaume, G. Grittani, O. Lundh, M. Hansson, J. Gautier, M. Kozlova, J. P. Goddet, P. Rousseau, A. Tafzi, V. Malka, A. Rousse, S. Corde and K. T. Phuoc, Stable femtosecond X-rays with tunable polarization from a laser-driven accelerator, *Light: Science & Applications, 2017,* ***6, e17086.***

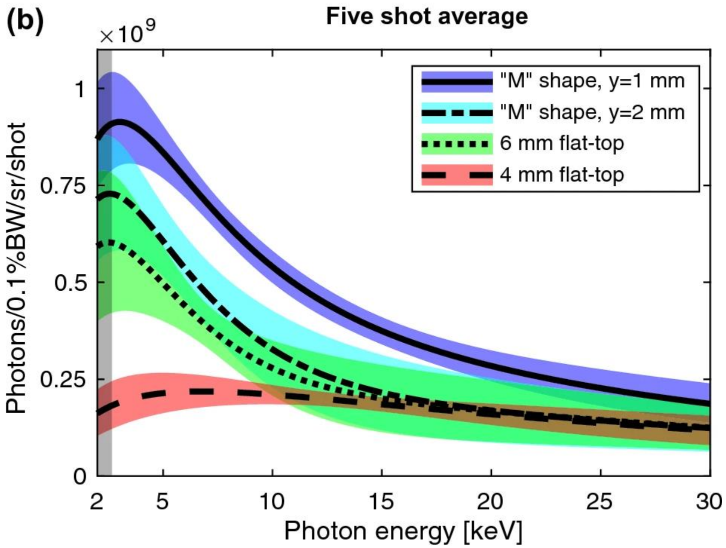
Betatron 2





High power laser facilities (dream beam)
Betatron oscillations
very short pulses

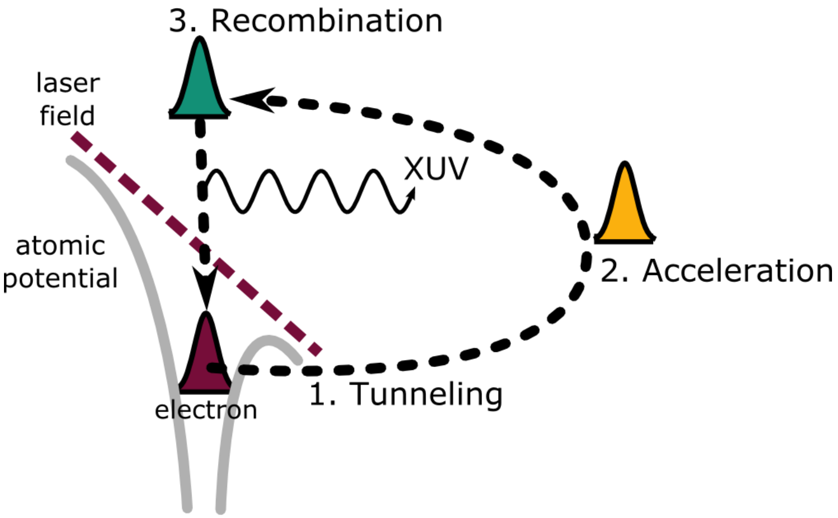
Inverse Compton sources
very high energy photons

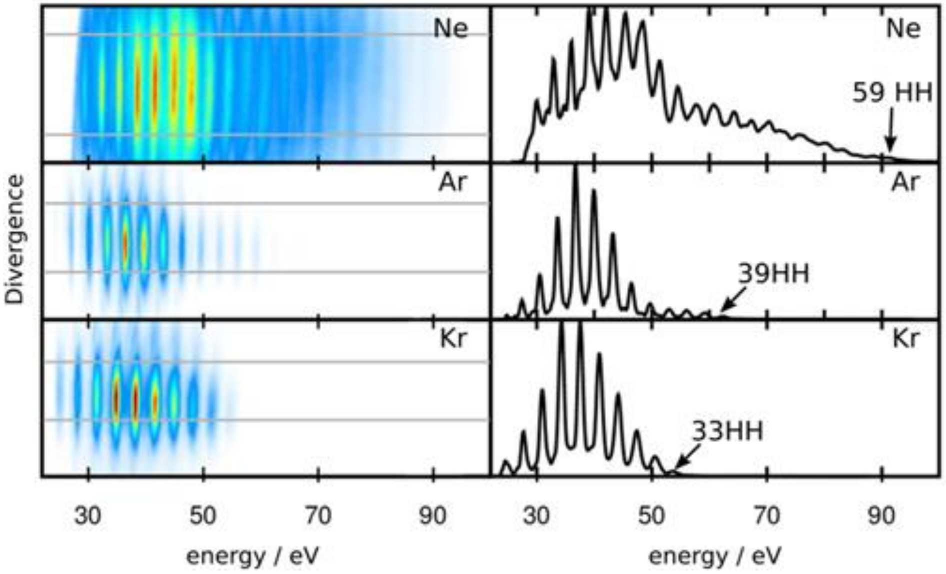


@7keV = 7eV so
1e8 photons/shot/str
200 shots = 1e10 photons/h/str/eV
5Hz = 2e12 photons/h/str/eV
1KHz (plan) = 2e13 photons/h/str/eV

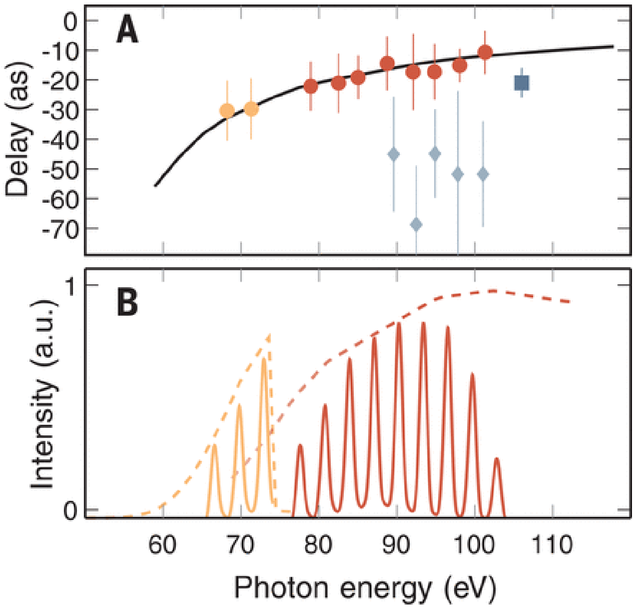
mages: Sci Rep **12**, 10855 (2022). https://doi.org/10.1038/s41598-022-14748-z
Lund: Sci Rep . 2020 Oct 8;10(1):16807. doi: 10.1038/s41598-020-73805-7.

HHG 1





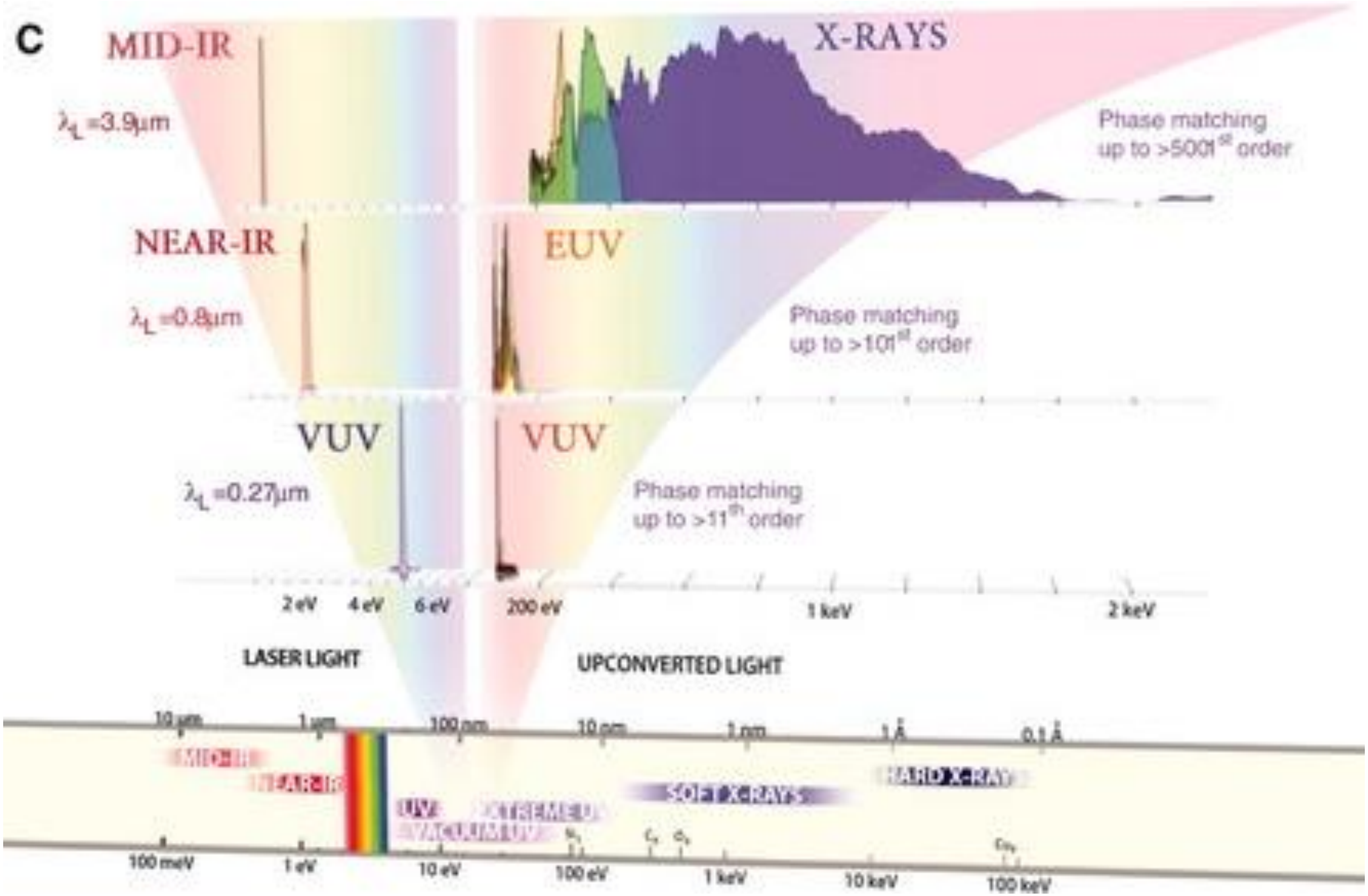
A. Harth, C. Guo, Y.-C. Cheng, A. Losquin, M. Miranda, S. Mikaelsson, C. M. Heyl, O. Prochnow, J. Ahrens, U. Morgner, A. L’Huillier and C. L. Arnold, Compact 200 kHz HHG source driven by a few-cycle OPCPA, *Journal of Optics, 2018,* ***20, 14007.***



Time delay differences [$\tau_A(2s) - \tau_A(2p)$] in neon

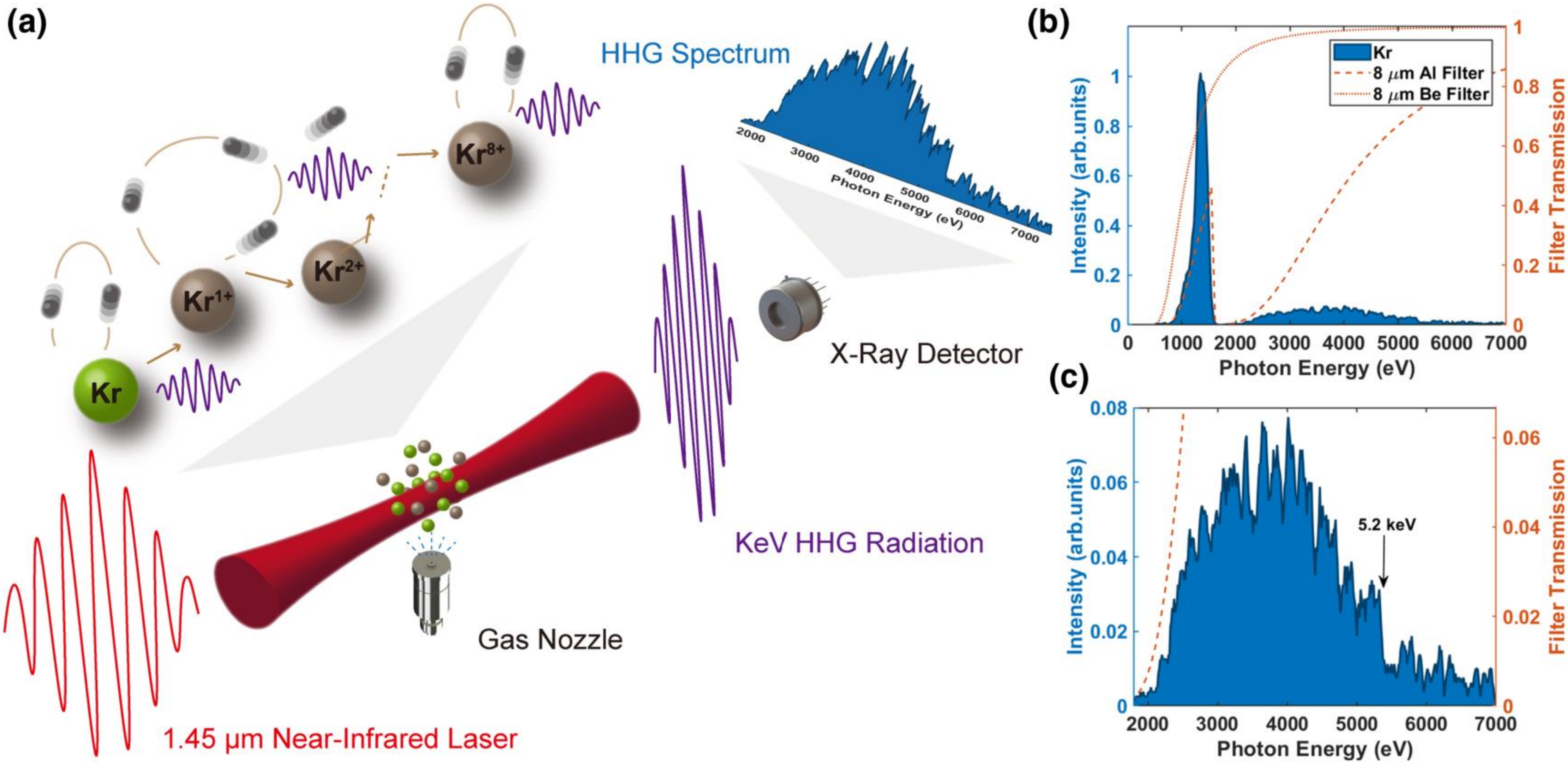
M. Isinger, R. J. Squibb, D. Busto, S. Zhong, A. Harth, D. Kroon, S. Nandi, C. L. Arnold, M. Miranda, J. M. Dahlström, E. Lindroth, R. Feifel, M. Gisselbrecht and A. L’Huillier, Photoionization in the time and frequency domain, *Science, 2017,* ***358, 893–896.***

HHG 2



Science, Kapteyn Volume: 336, Issue: 6086,
Pages: 1287-1291, DOI: (10.1126/science.1218497)

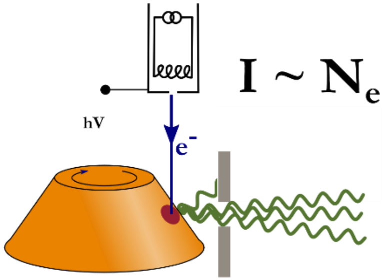
HHG 3



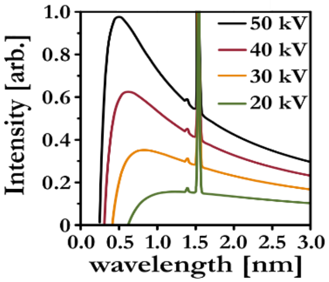
3e5 ph/str/h @4keV,
But 1e12/str/h @400eV (heard)

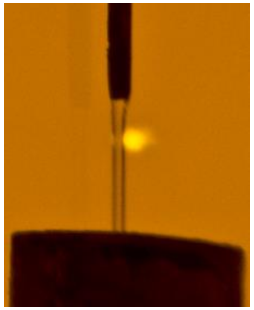
Optica, Vol. 9, No. 9 / September 2022 / 1003, https://doi.org/10.1364/OPTICA.456481

Source comparison

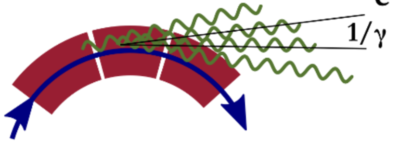


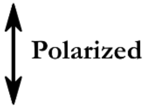
$I \sim N_e$

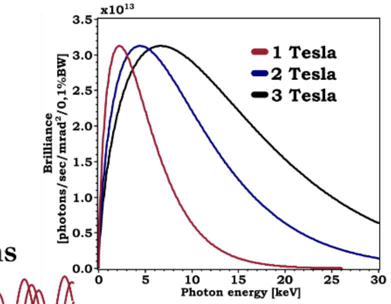




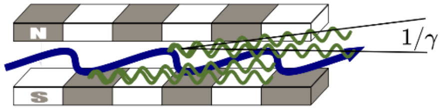
$I \sim N_e$



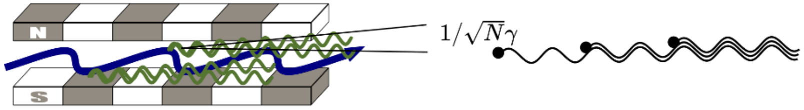




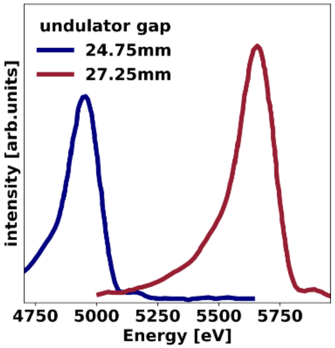
$I \sim N_{magnets} * N_{electrons}$



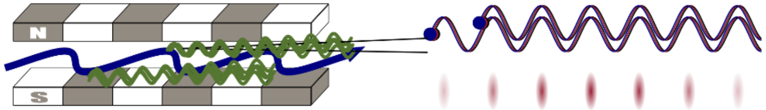
$I \sim N^2_{magnets} * N_{electrons}$



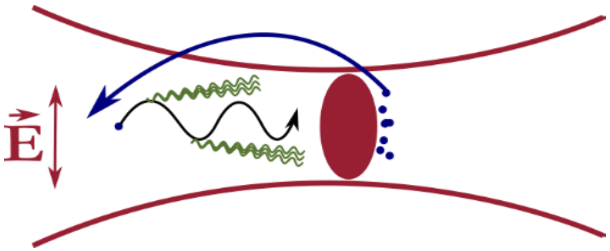
$\lambda_{resonance} \sim B_0^2$

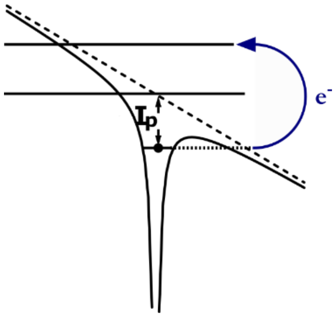


$I \sim N^2_{magnets} * N^{1\text{-}2}_{electrons}$

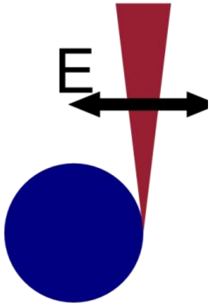


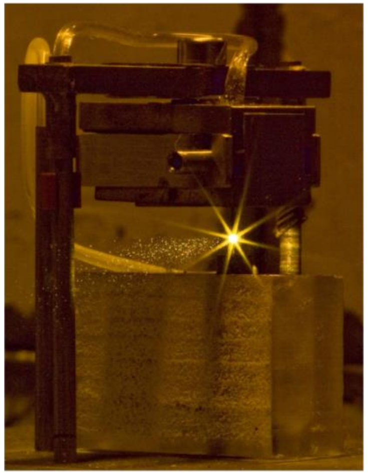
$\lambda_{resonance} \sim B_0^2$



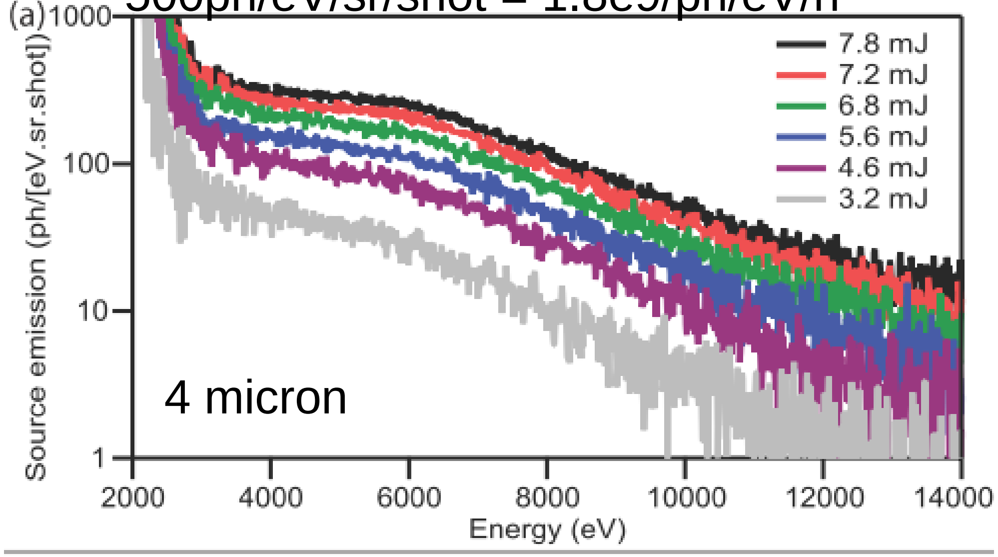


x-rays from water in general

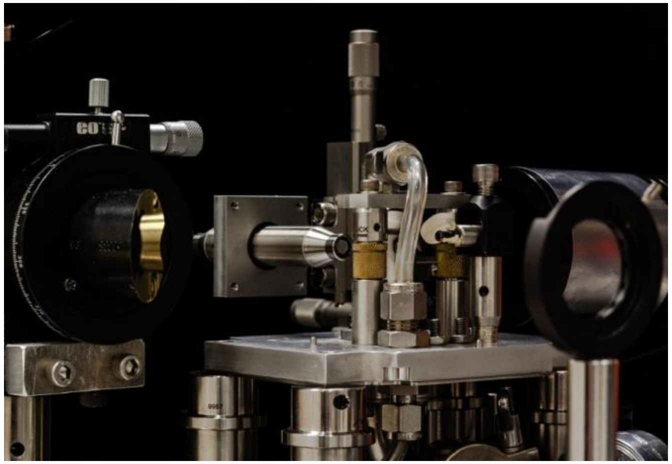


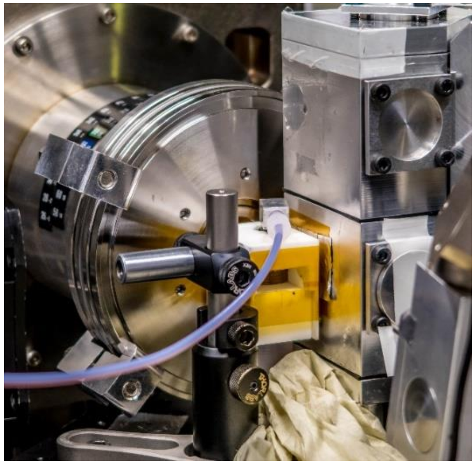


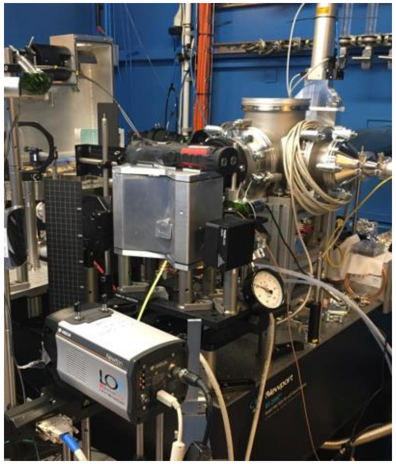
500ph/eV/sr/shot = 1.8e9/ph/eV/h



4 micron





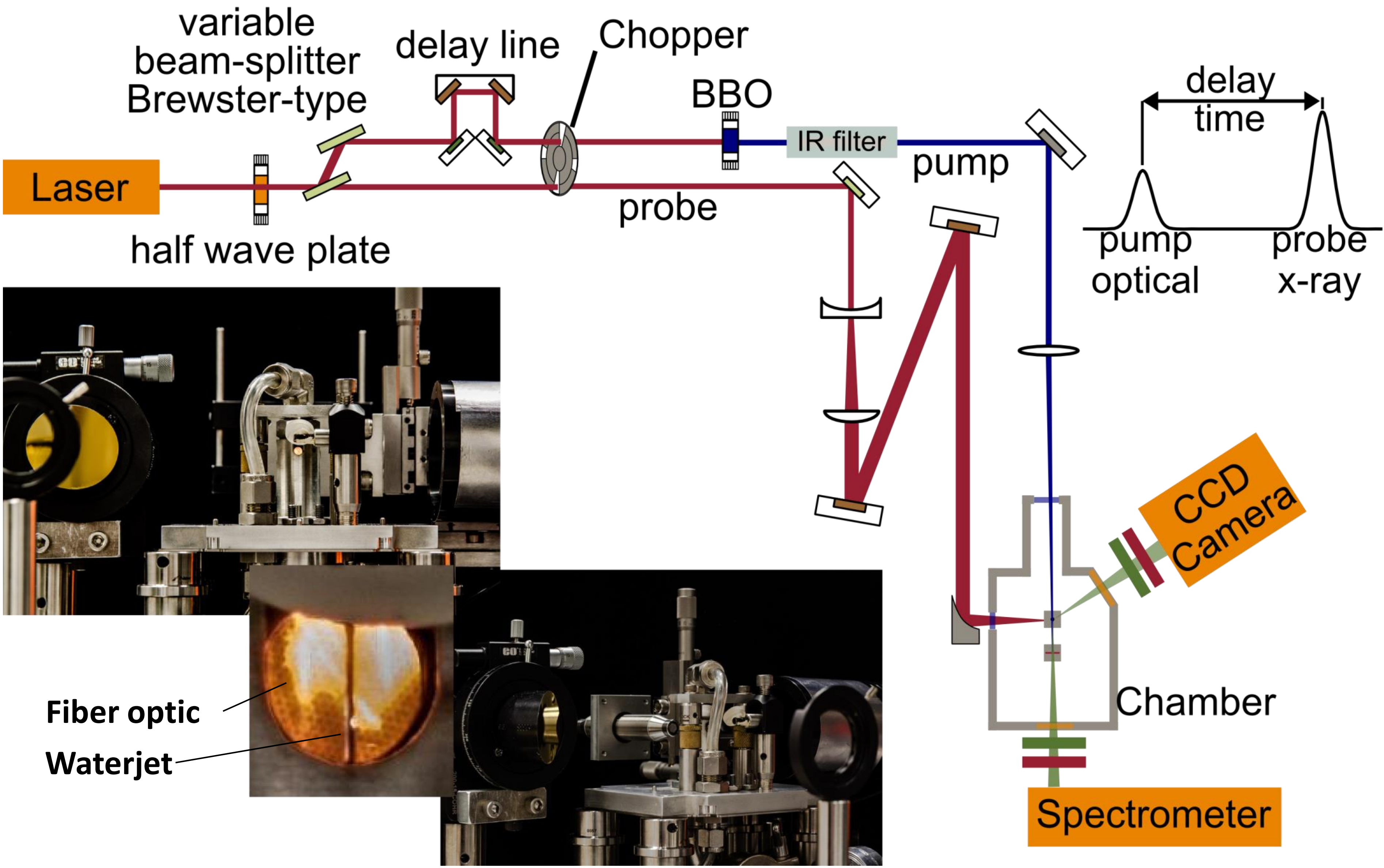


NIST
National Institute of
Standards and Technology
Quantum Devices Group

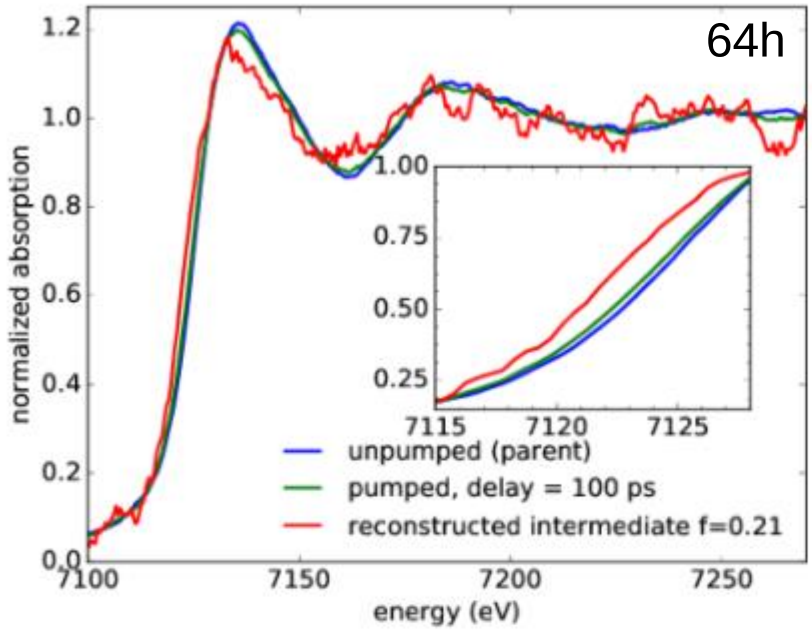
Lund
Laser Centre

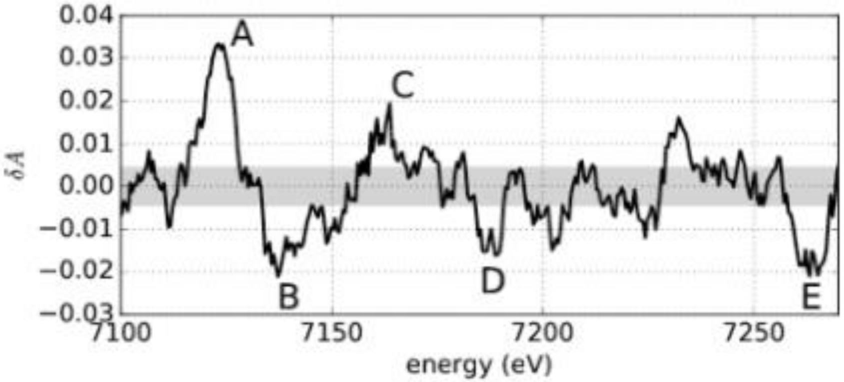
eli
beamlines

Laboratory pump – probe setup

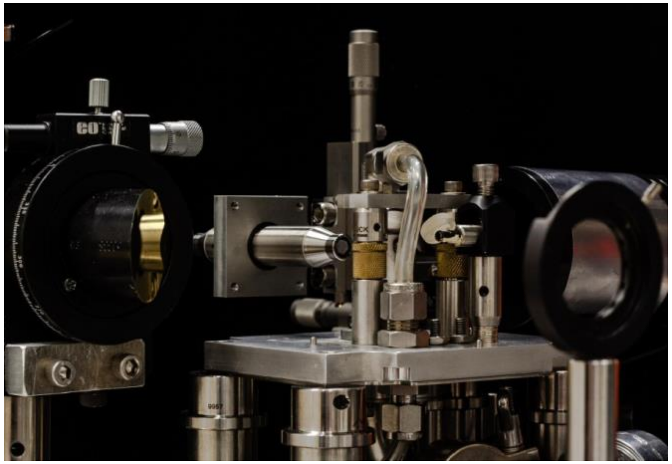


time-resolved XAFS and XES at low excitation yield





The Journal of Physical Chemistry Letters, vol. 8, no. 5, pp. 1099–1104, Feb. 2017, doi: 10.1021/acs.jpclett.7b00078.



NIST
National Institute of
Standards and Technology
Quantum Devices Group

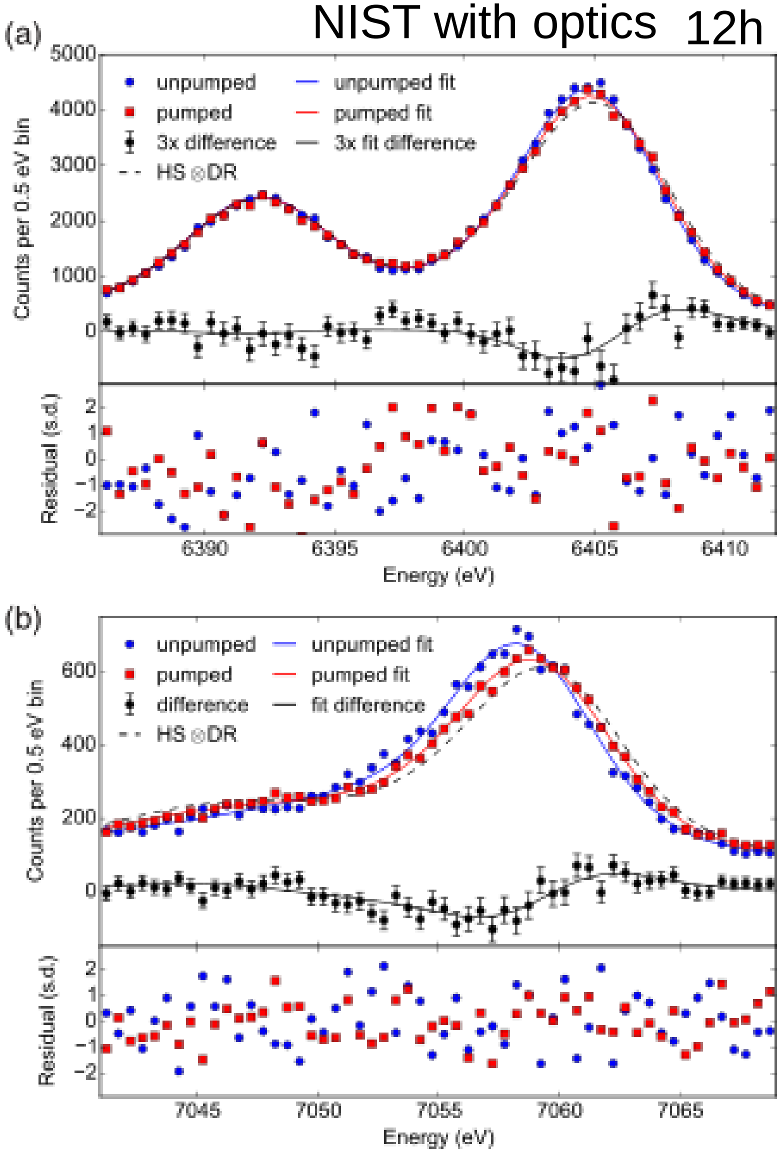
Wrong Experiment!!!

5e9ph/str/eV/h @4mJ

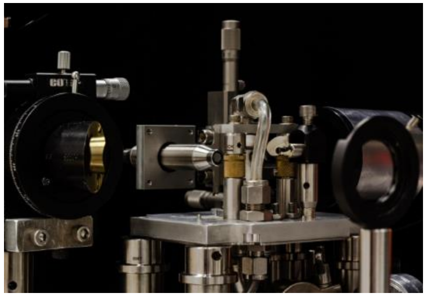
XES:
5e12ph/str/h (1000eV absorbed)
No detector saturation

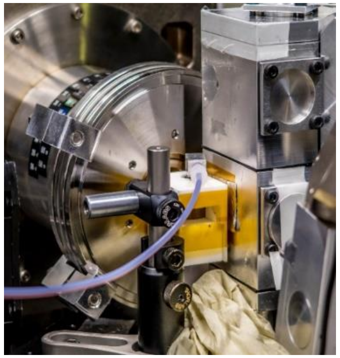
time-resolved XAFS and XES at low excitation yield

NIST with optics 12h

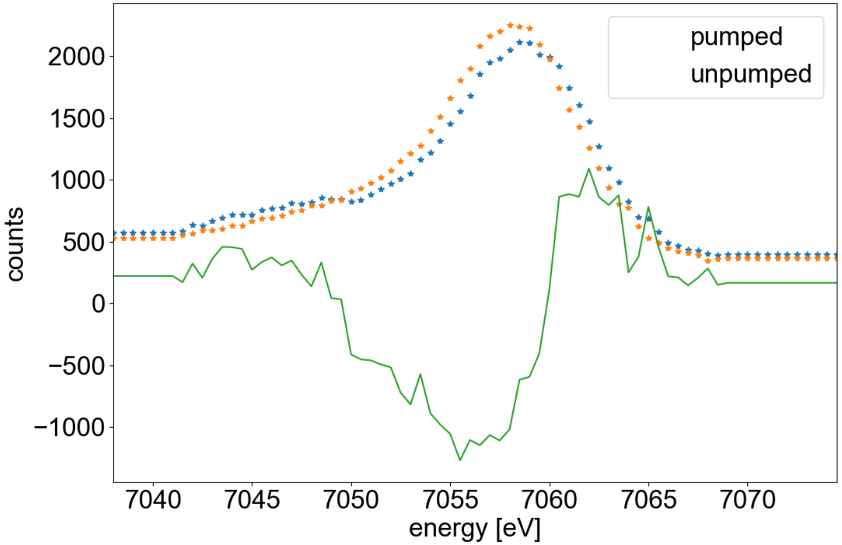


Physical Review X, vol. 6, no. 3, Sep. 2016,
doi: 10.1103/physrevx.6.031047.

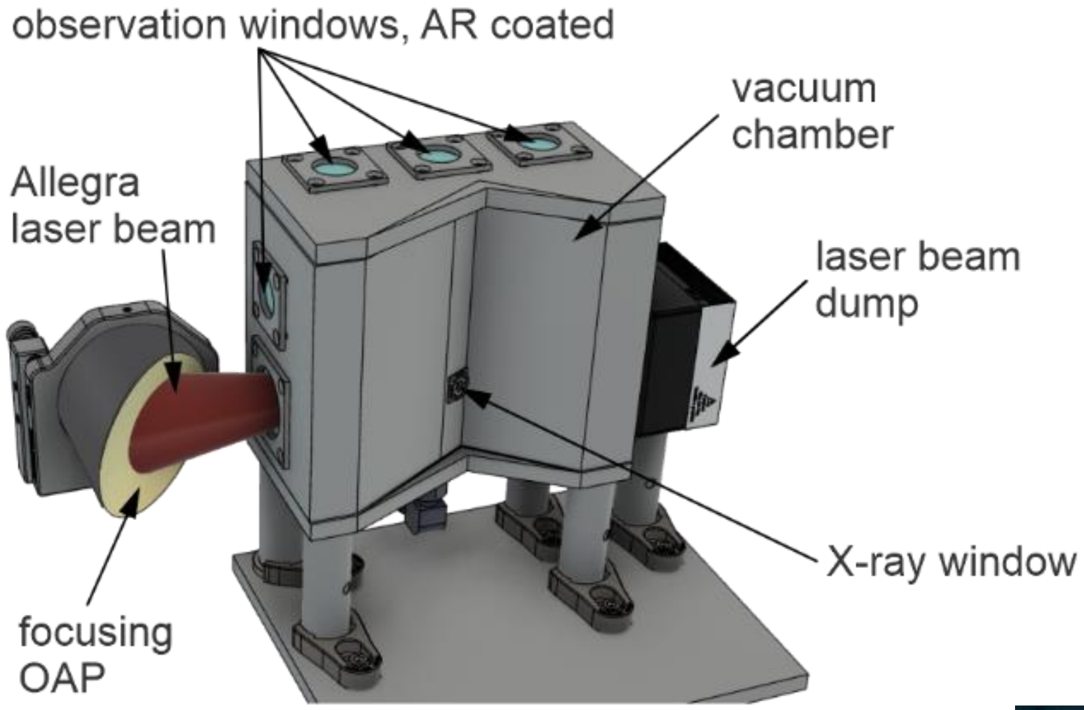


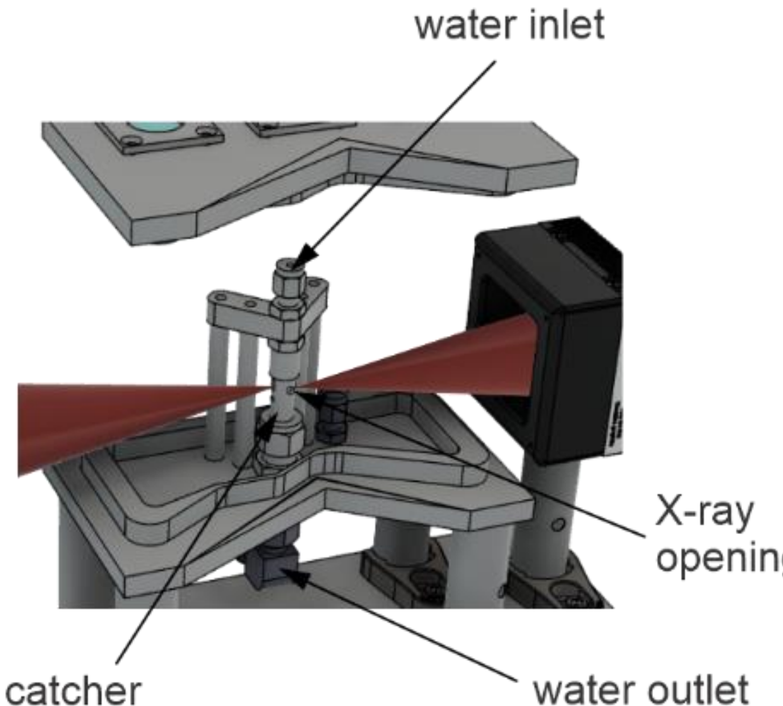


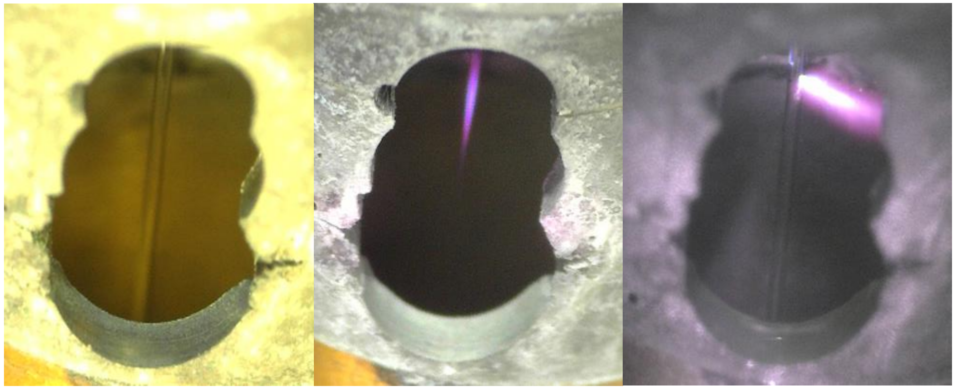
1 person setup and run,
4h accumulation,
3d printed chamber
NO optics
8ml at 6m/s, 10mMol, 15%



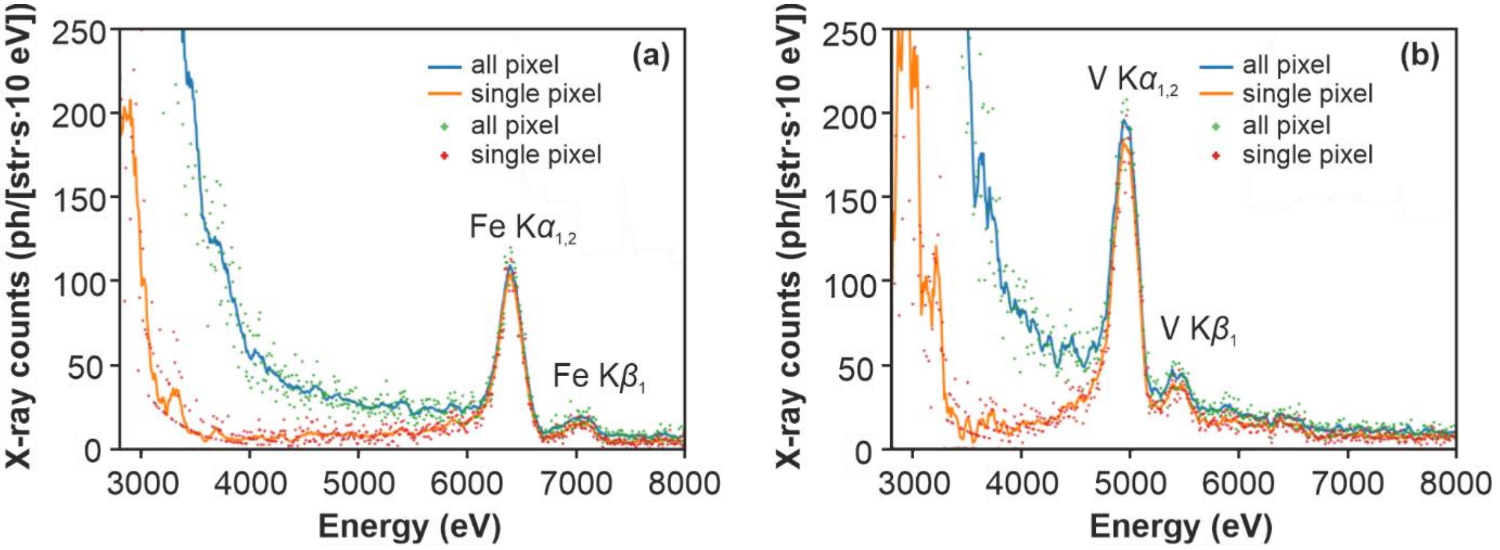
My confession, I also went for bigger sources

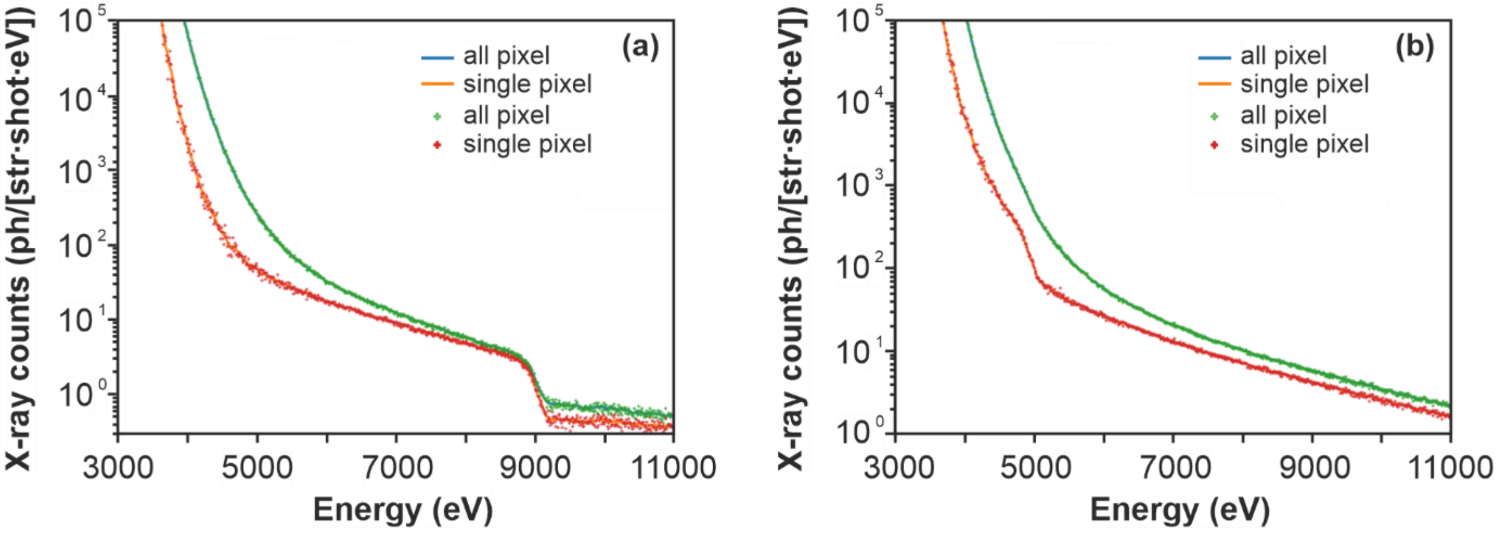






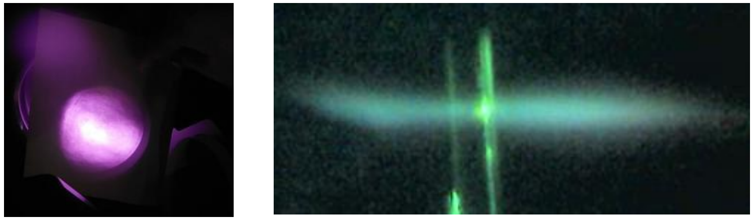
Spectrum at Eli

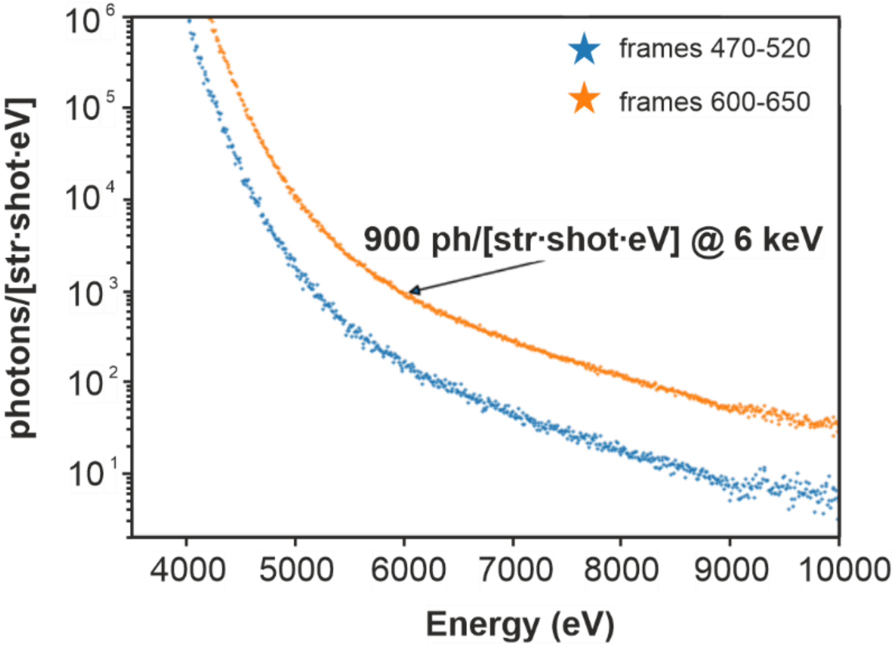




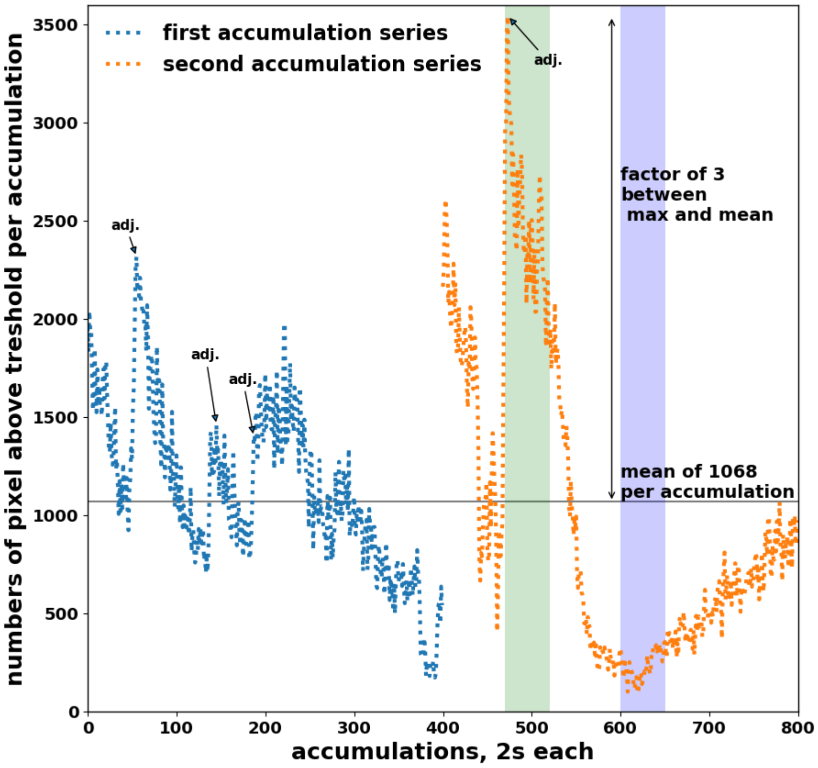
J. Synchrotron Radiat., vol. 28, no. 6, pp. 1778–1785, Nov. 2021, doi: 10.1107/s1600577521008729.

Flux and fluctuations last year





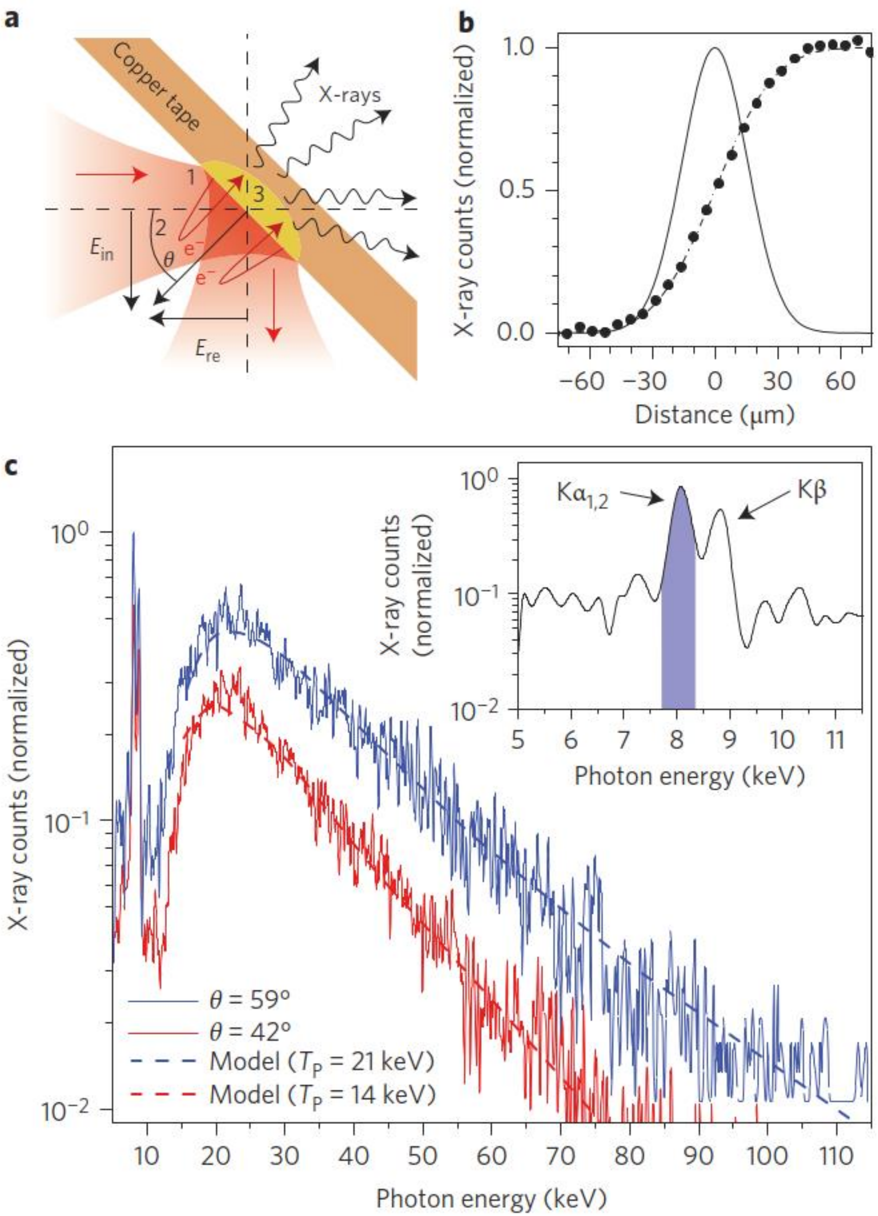
3e9ph/str/eV/h @30mJ (now is 100mJ)

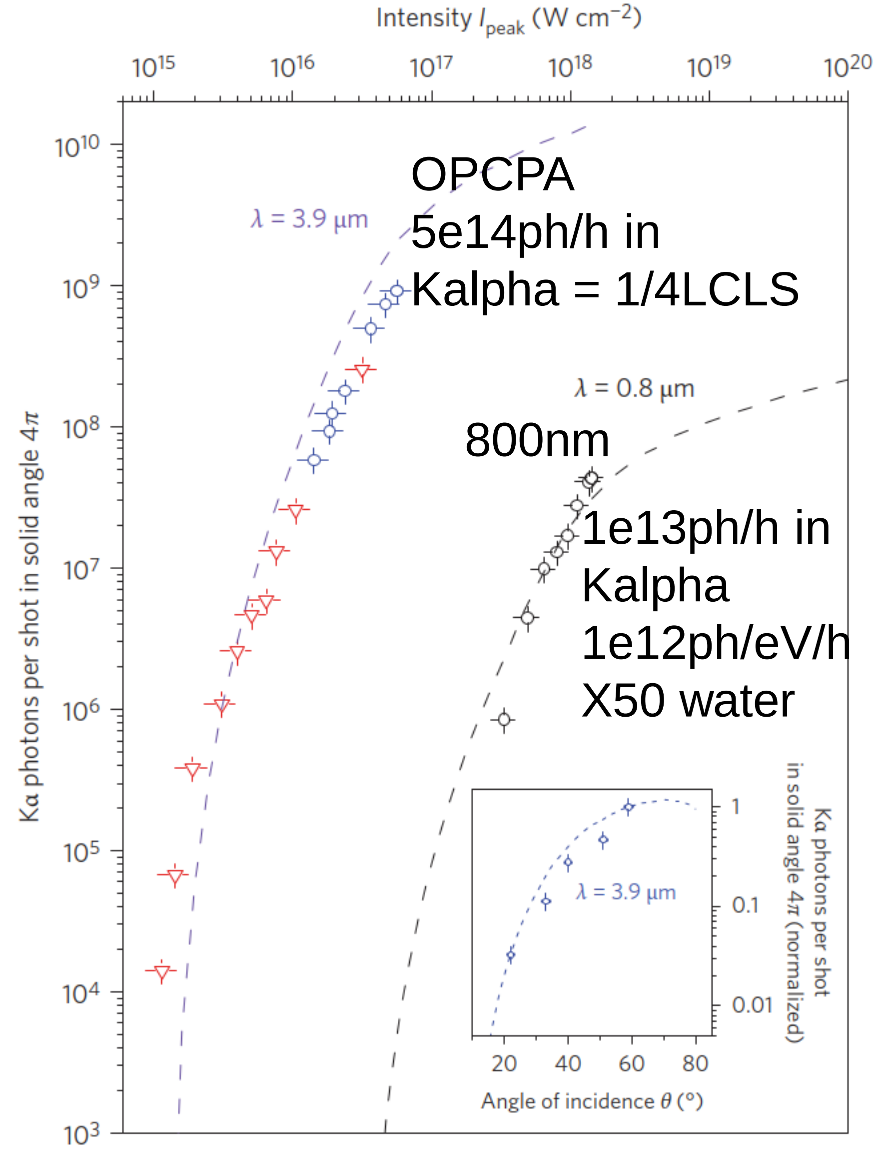


J. Synchrotron Radiat., vol. 28, no. 6, pp. 1778–1785, Nov. 2021, doi: 10.1107/s1600577521008729.

With single crystal detector:
So 2h for steady state Kalpha after reasonable improvement with 100mJ in 2min
With the 16 crystal Zoltan will show you the same time for Kbeta
Differential spectrum x 10 – x100 (20-200min)

Plasma





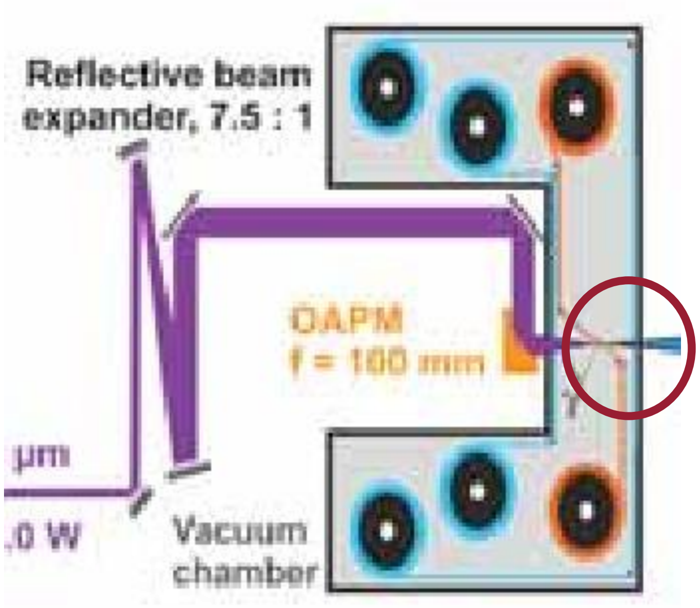
OPCPA
5e14ph/h in
Kalpha = 1/4LCLS

800nm

1e13ph/h in
Kalpha
1e12ph/eV/h
X50 water

Elsaesser, Nature Photonics, vol. 8, no. 12, pp. 927–930, 2014, doi: 10.1038/nphoton.2014.256. (same numbers CLEO 2022)

Using Plasma sources

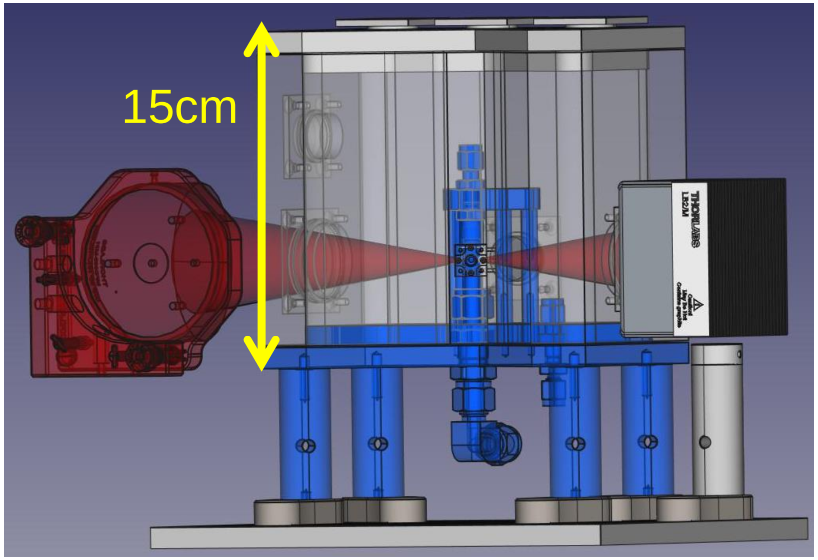


1.5m

Geometry is the key, or: how much of the flux can you catch?
Here 6 cm!

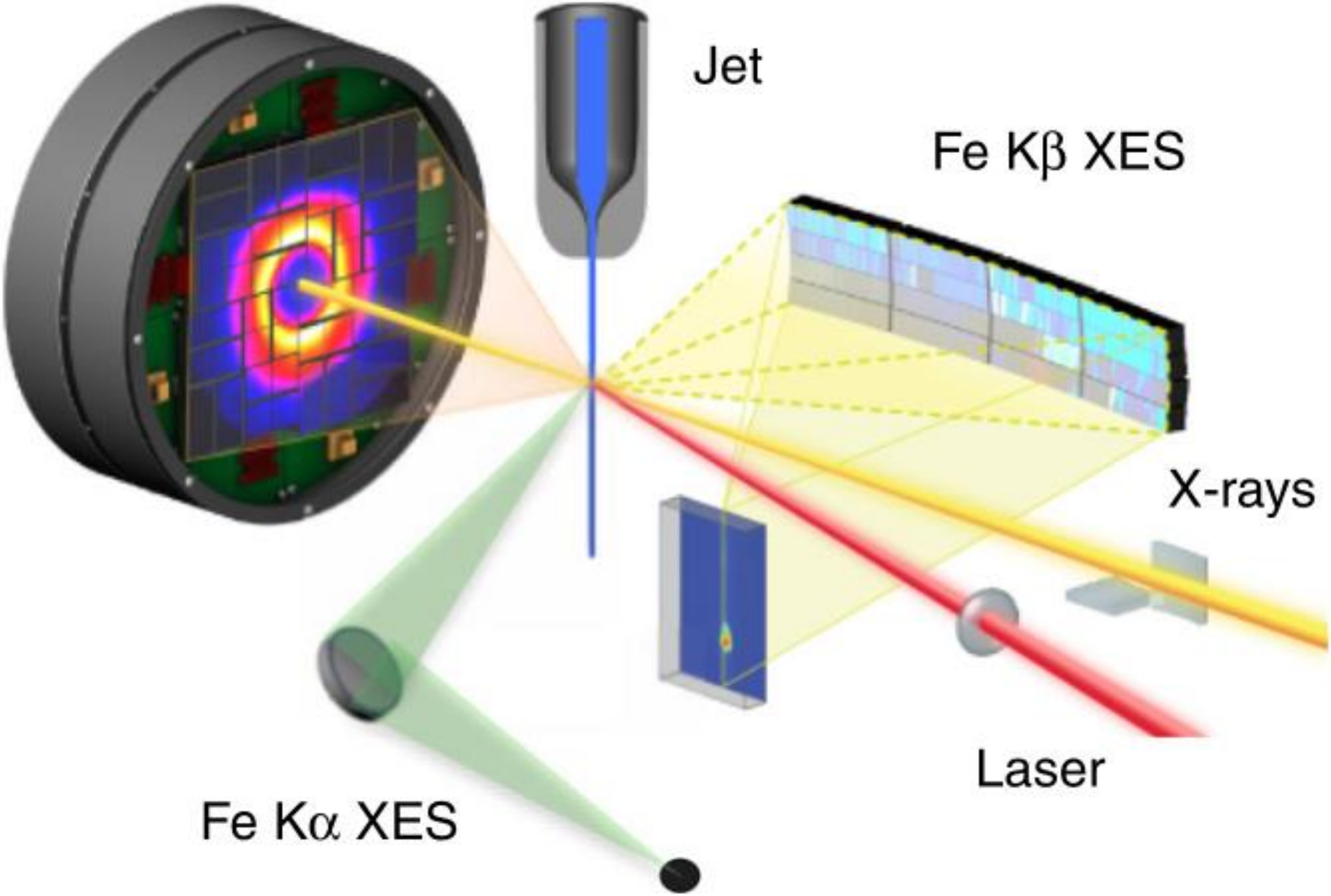
Elaesser, CLEO, IEEE, 2022 JM2E.3

Price 400kEuro,
New bands from BASF, changing bands regularly

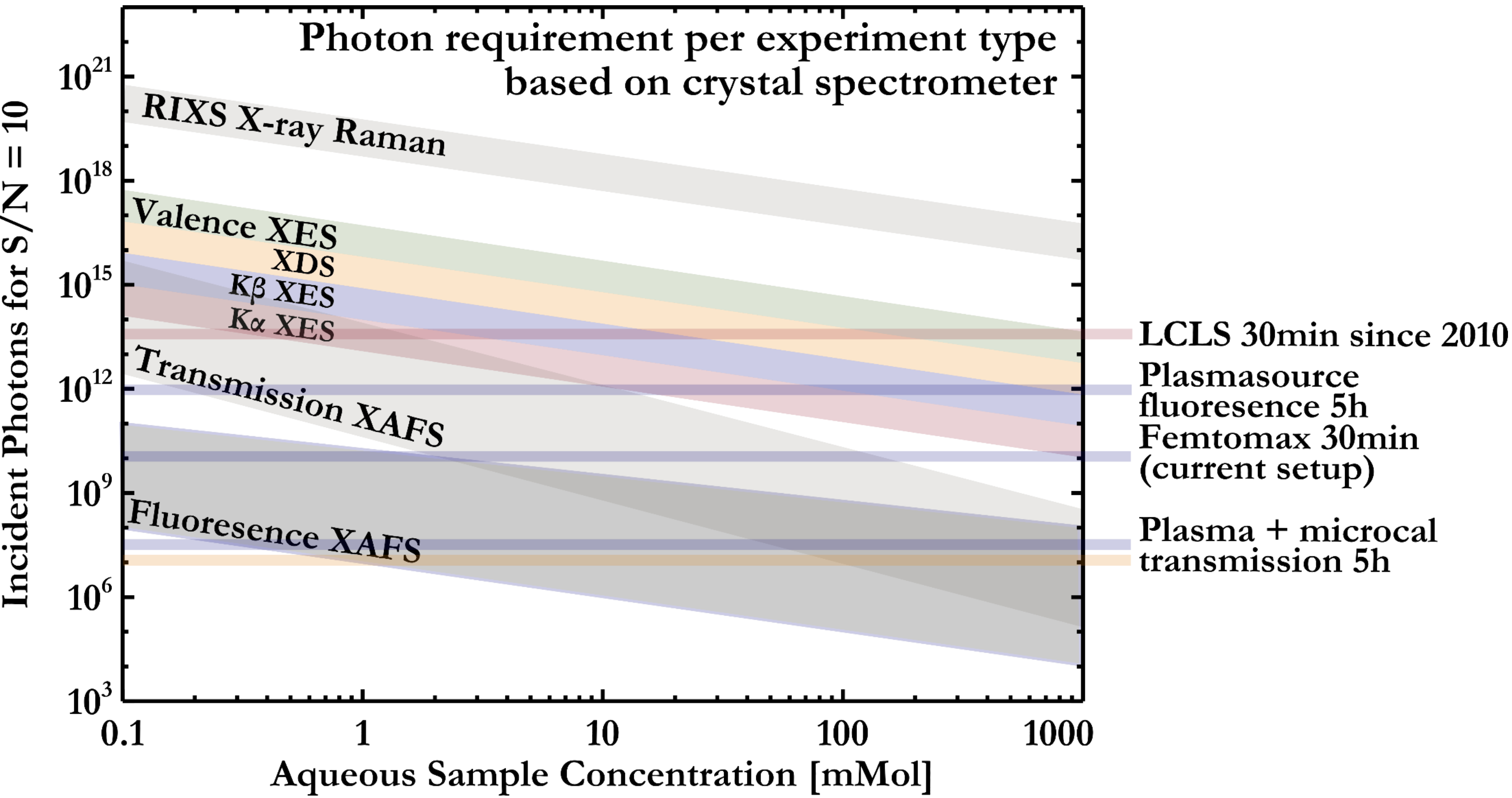


Here 1.8 cm = 11x the used flux

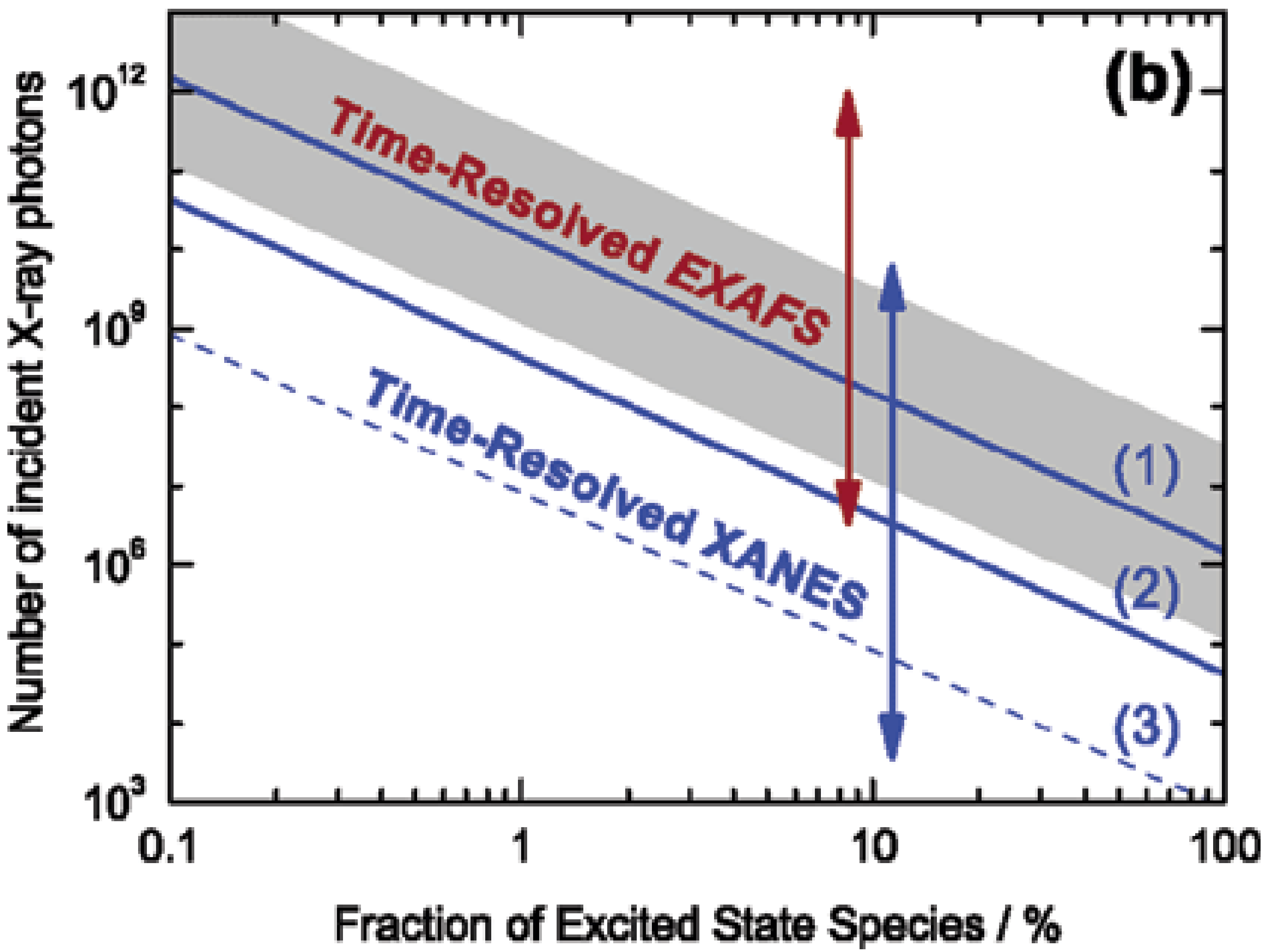
As seen with x-ray eyes. Simultaneous XES/XDS and slow XAS



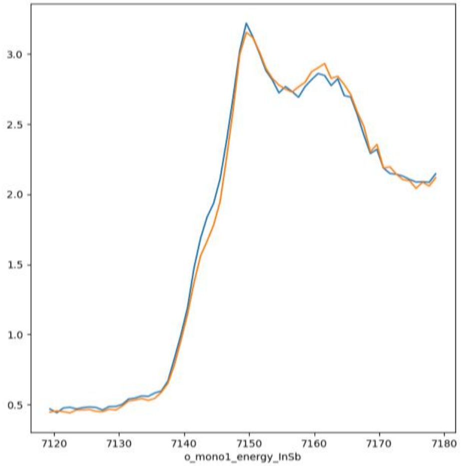
Flux for different pump-probe techniques

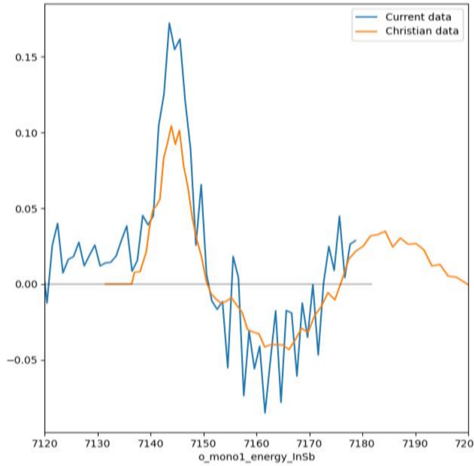


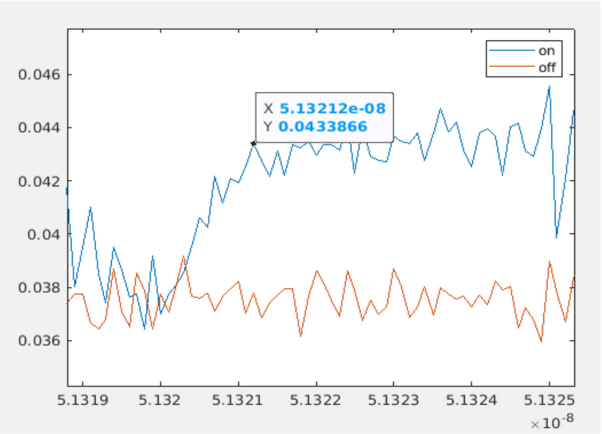
Flux for different pump-probe techniques



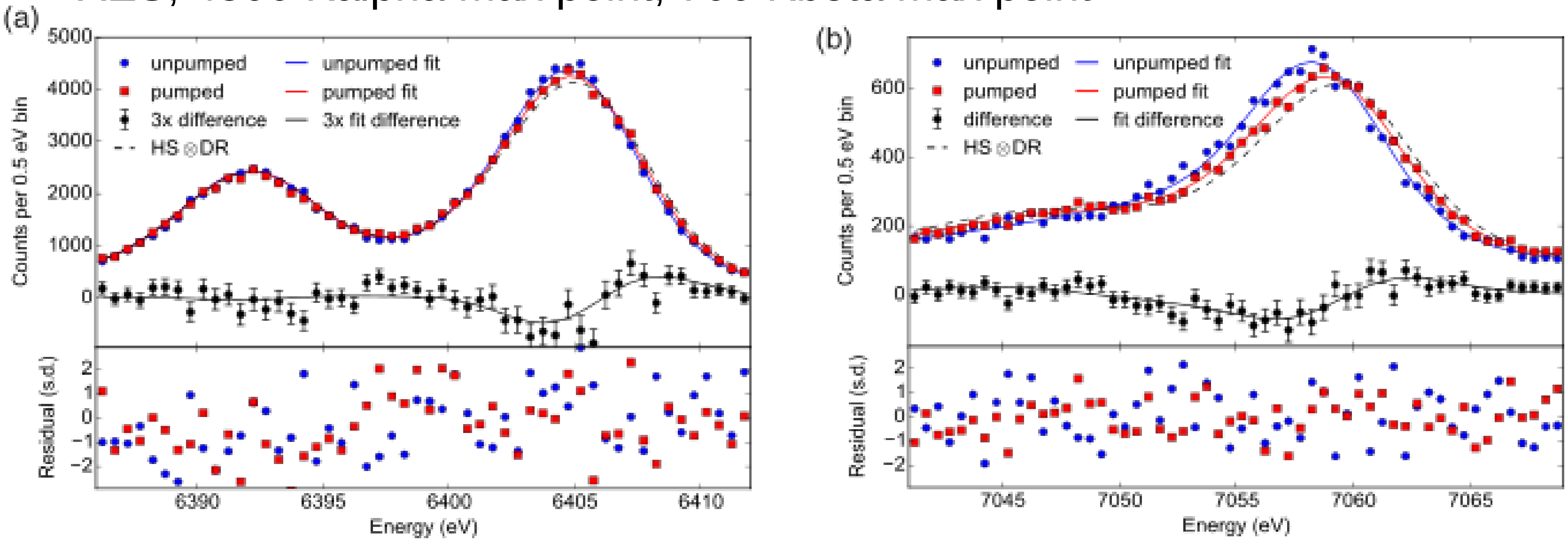
Numbers from Experiments:







XANES, need for SN=4 3.2e6 photons/point, for f=20%,
XES, 4500 Kalpha max point, 700 Kbeta max point

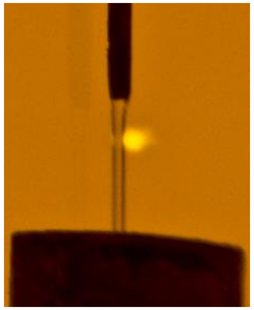


Physical Review X, vol. 6, no. 3, Sep. 2016, doi: 10.1103/physrevx.6.031047.

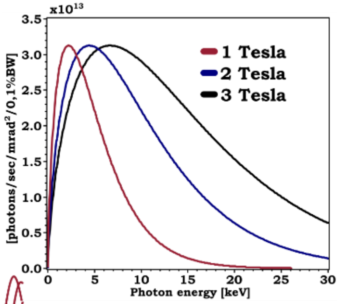
Source comparison

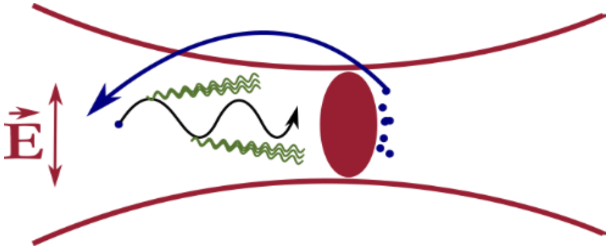
XAS: 200eV
XES: 1500eV

OPCPA:
5e14ph/h/str in Kalpha = 1/4LCLS
800nm:
1e12ph/eV/h/str broadband copper
2e9/ph/eV/h/str broadband water

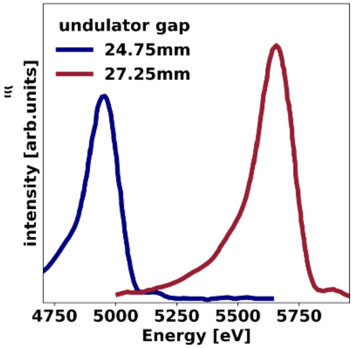


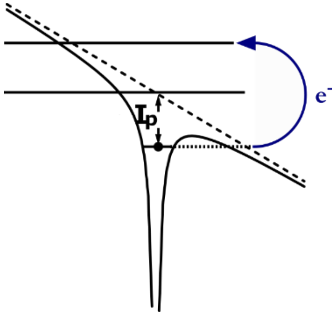
APS 7keV (approx.)
1e14 ph/s @ 6MHz mono chrome
2e13 ph/s @ 1.25 MHz
7e16 ph /h monochrome





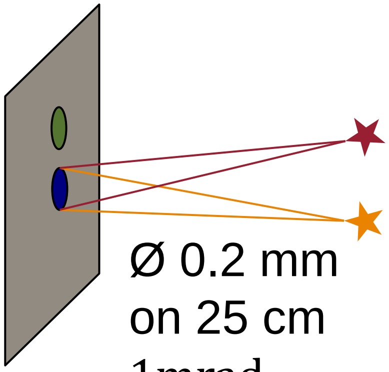
LCLS 7keV (approx.)
1e10 ph/shot @ 60Hz
2e15 ph /h polychrome
2e13 ph /h monochrome





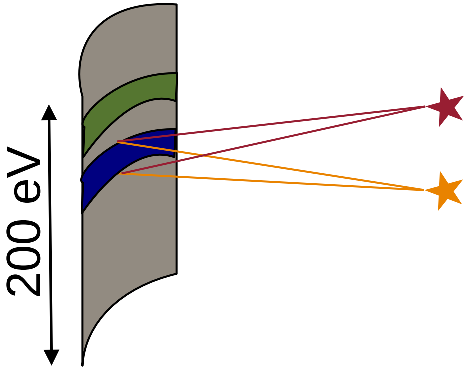
Crystal geometries

Flat



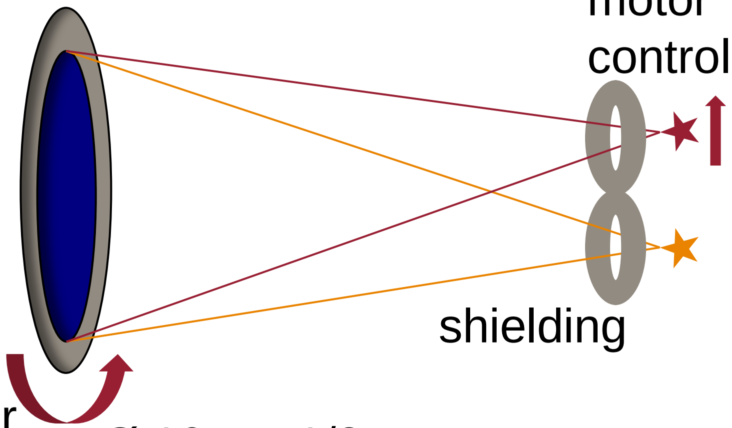
Ø 0.2 mm
on 25 cm
$1\mathrm{mrad}$
$\approx 1 \times 10^{-7}$

cylindrical bend
von-Hamos



0.2 mm x 10 cm
on 25 cm
$\approx 3 \times 10^{-5}$

Spherical bend,
Johan/Johannson



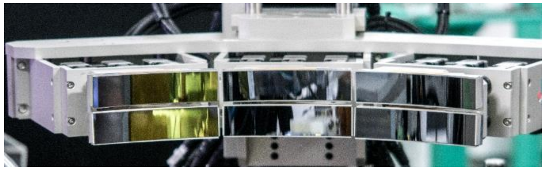
motor
control

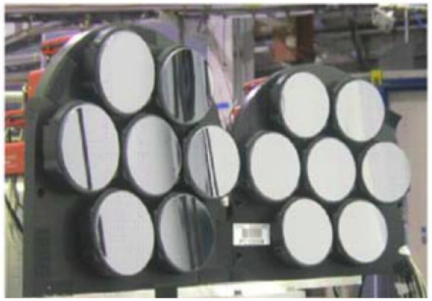
shielding

motor
control

Ø 10 cm 1/3
on 50 cm
$\approx 8 \times 10^{-4}$

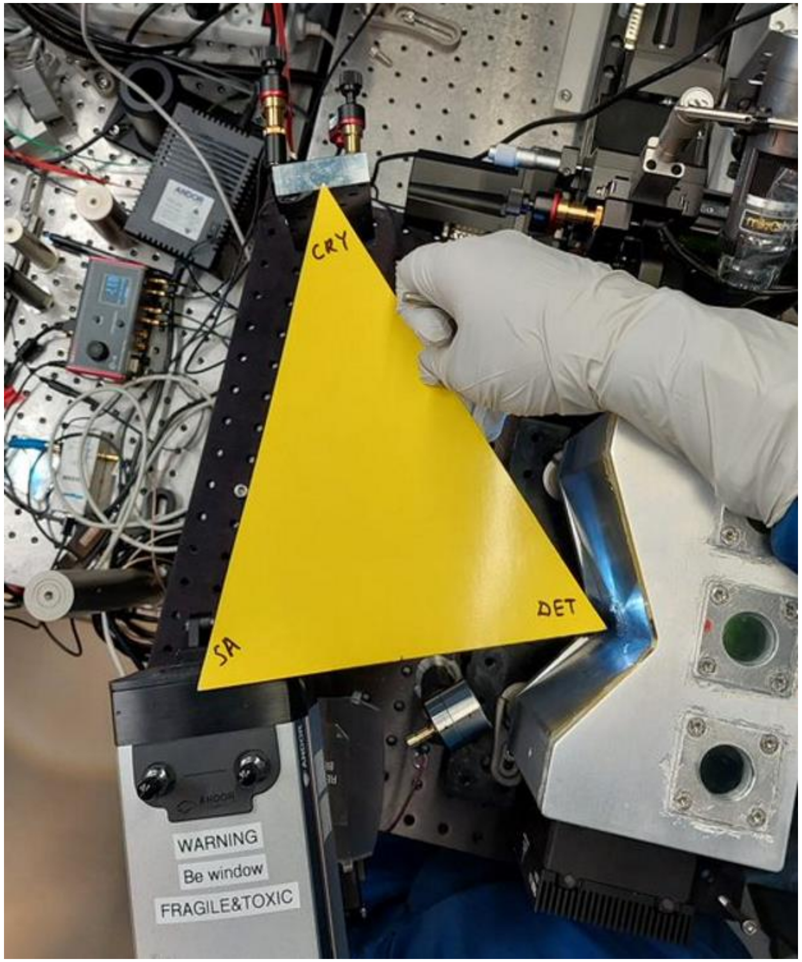
0.5 eV
scanning



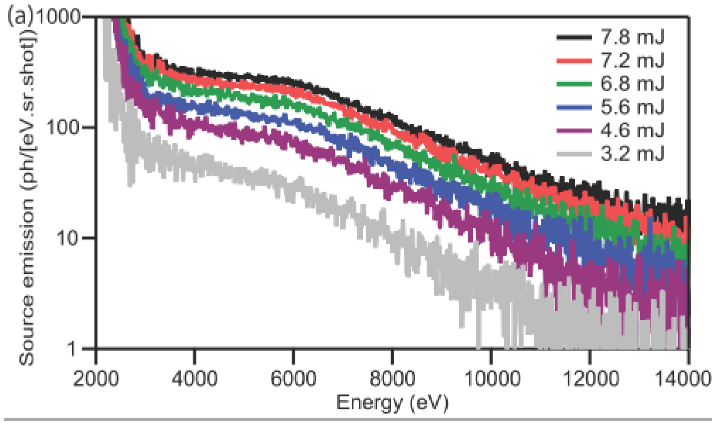


Gerry Seidler Global XAFS club
https://www.youtube.com/watch?v=3lJ9uE7Xvcg&t=101s

high efficient crystal detectors: efficiency <1e-7

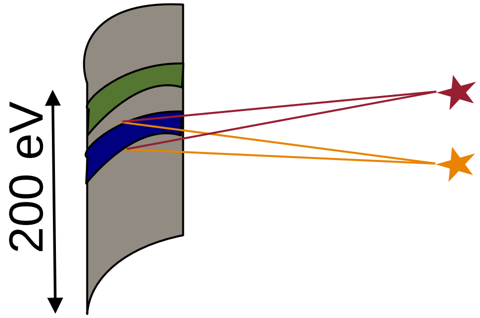


How to use all these numbers:



XAS
cylindrical bend von-Hamos

XES



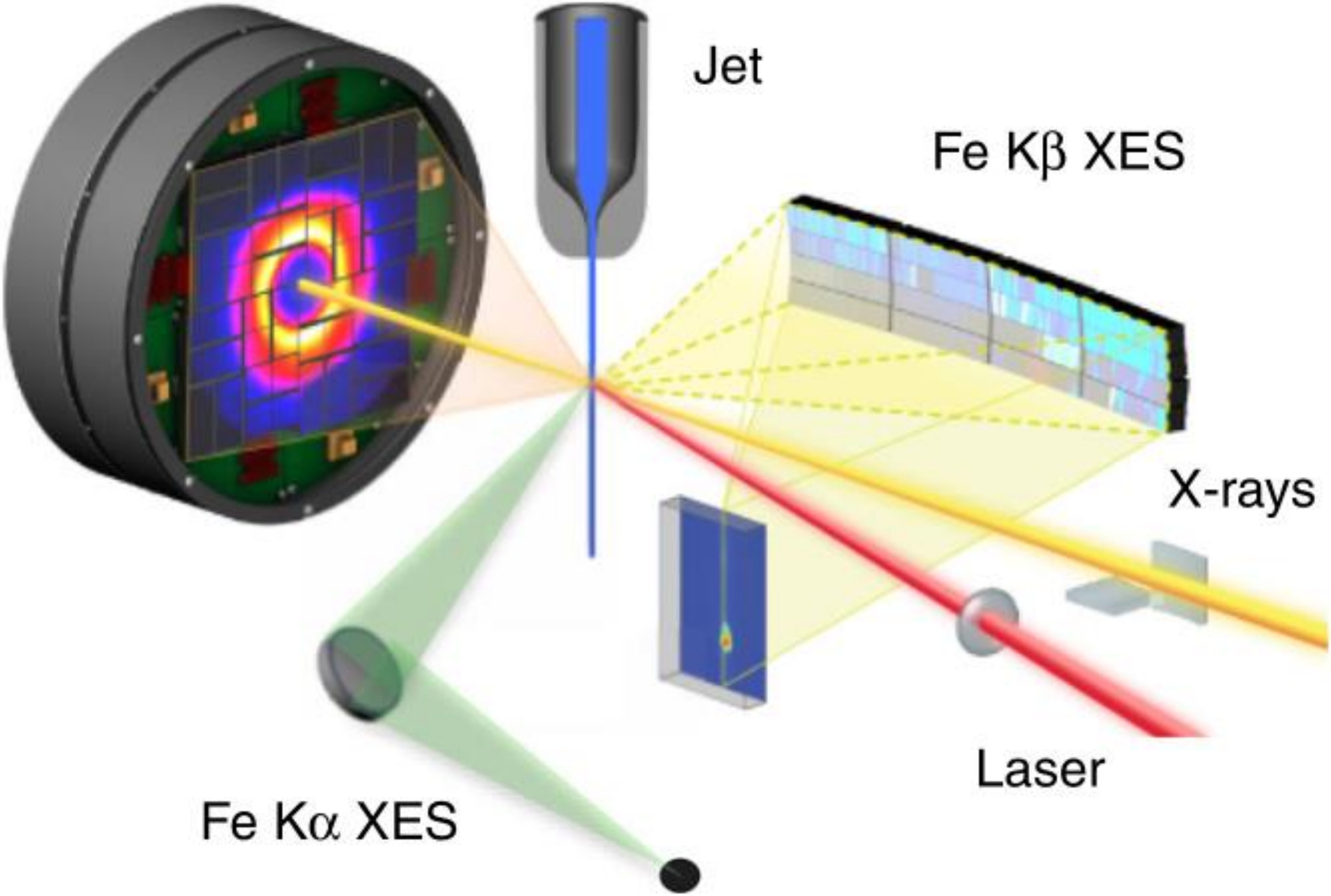
Source to sample 2cm,
300 micron spot
str= 3e-5 str area

0.2 mm x 10 cm
on 25 cm ≈ 3×10^{-5}

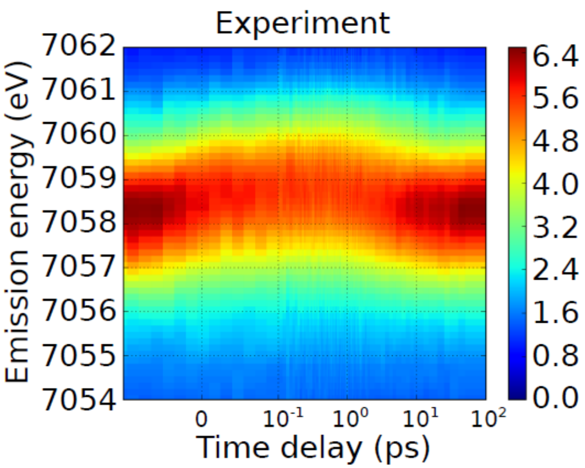
2e9ph/str/eV/h @8mJ
XAS: 200eV x 2e9ph/str/eV/h
x 3e-5 str =6e4/h we need 3e6 = 50h
XES: 1500eV x 2e9ph/str/eV/h = 3e12/h/str useful from the source
x 3e-5 str = 9e7/h hit sample and are absorbed
x 0.3 emission prob = 2.7e7/h/str emitted into 4pi
x 3e-5 str = 810/h detected Kalpha (1eV)
need 4500 = 5.5h
= 40/h detected Kbeta (all spectrum)
need 700 = 17 h

What can we improve? More crystals(x6)
Direct detection = 5000/s in theory, measured, 6h for small array, Factor 10 possible

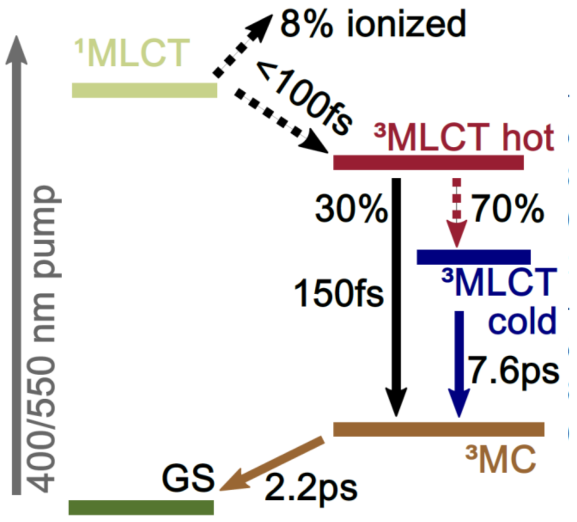
Plan experiment

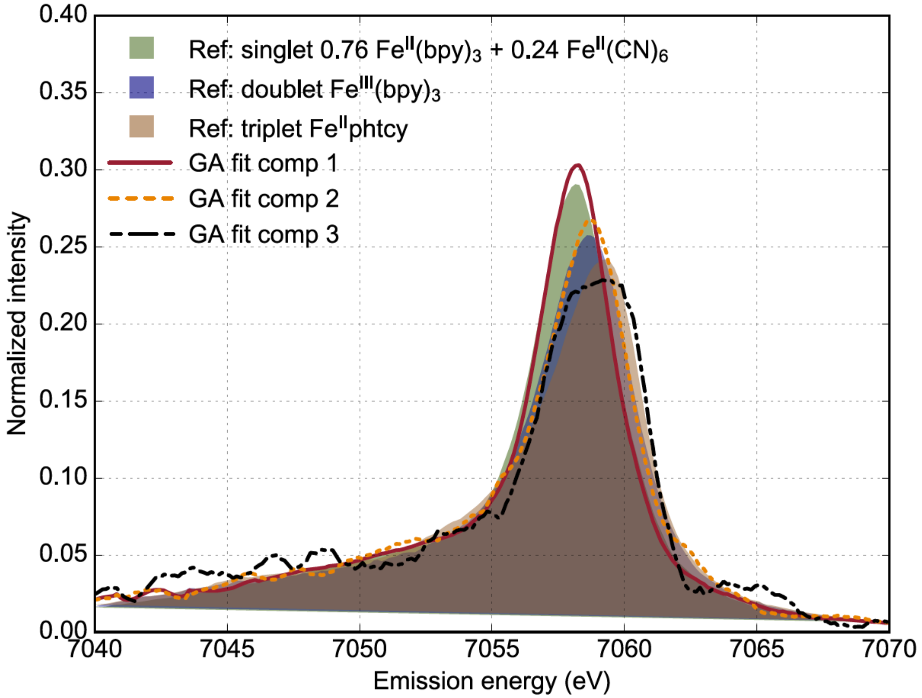


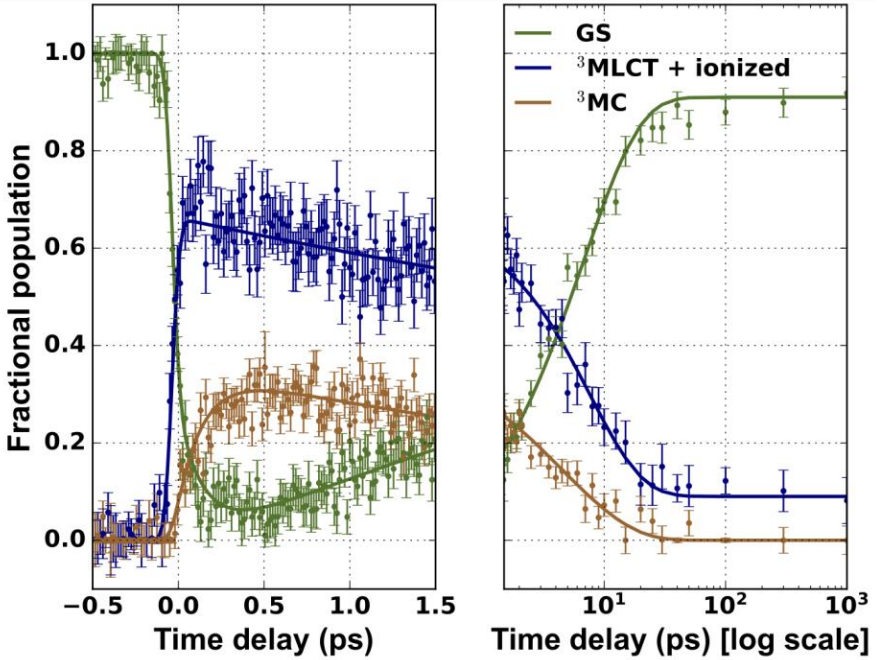
Global analysis



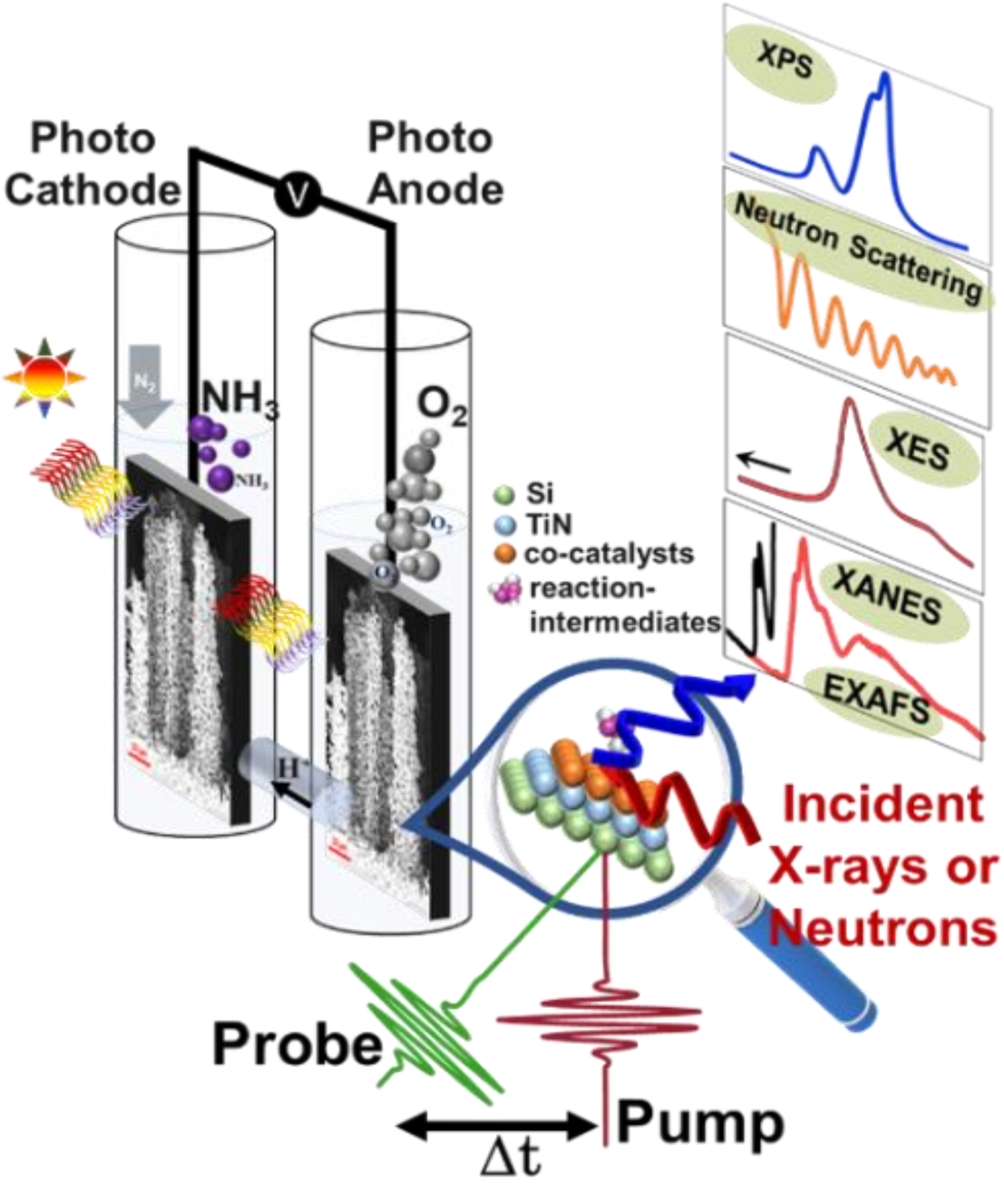
KiMo Pack



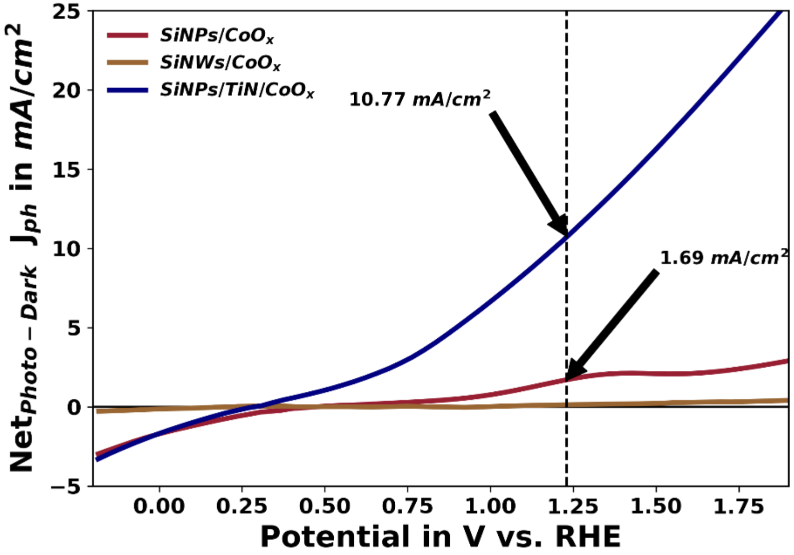




The path towards a highly efficient and scalable photo-catalysis using silicon nanowires



“World record”
13.3% Photons to Hydrogen efficiency
At 1 sun



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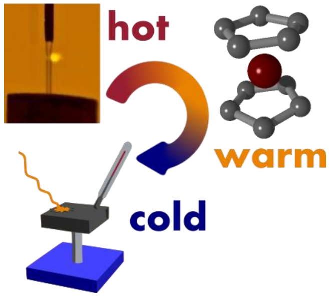
KiMo Pack

Care for your photons



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Thanks for your attention