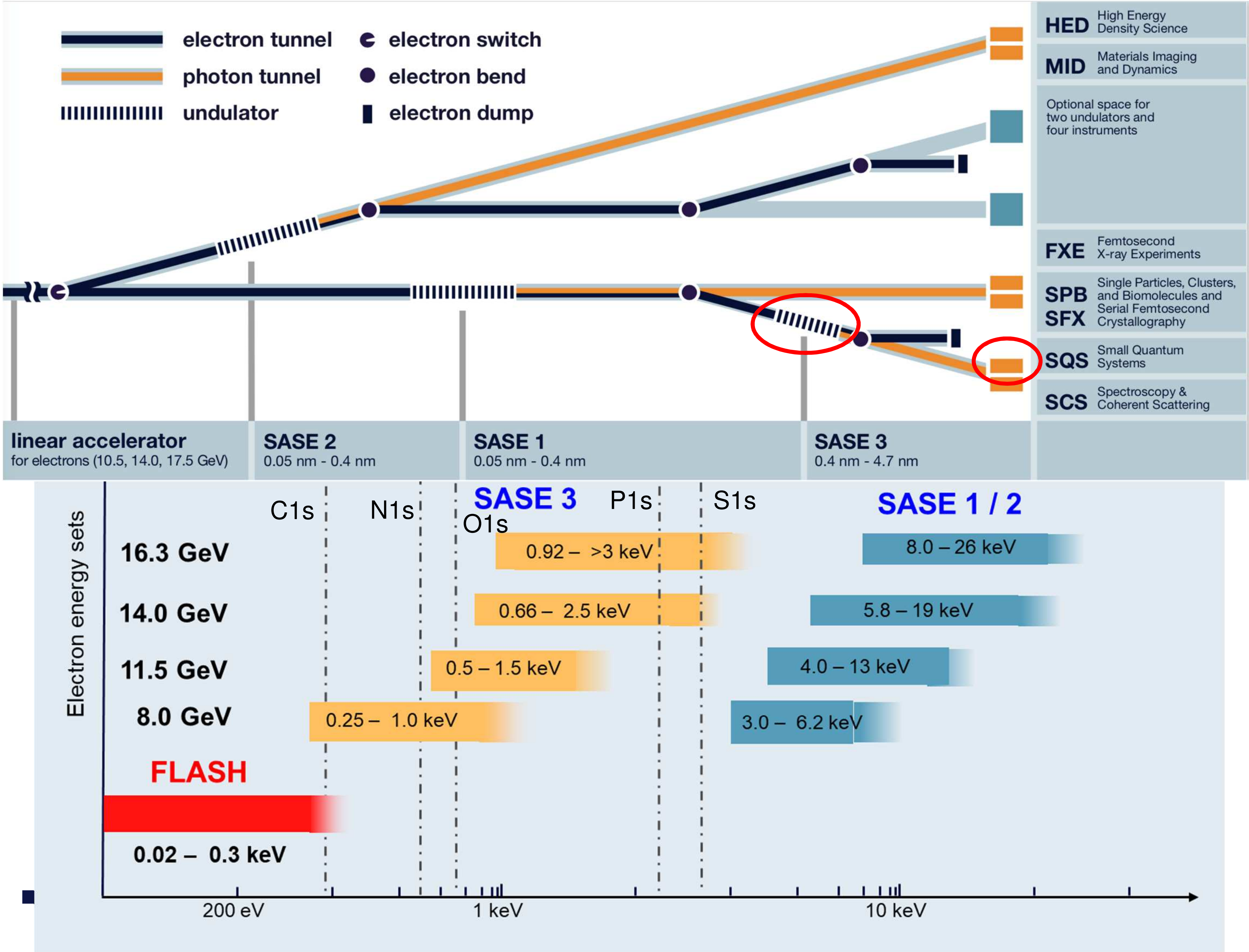
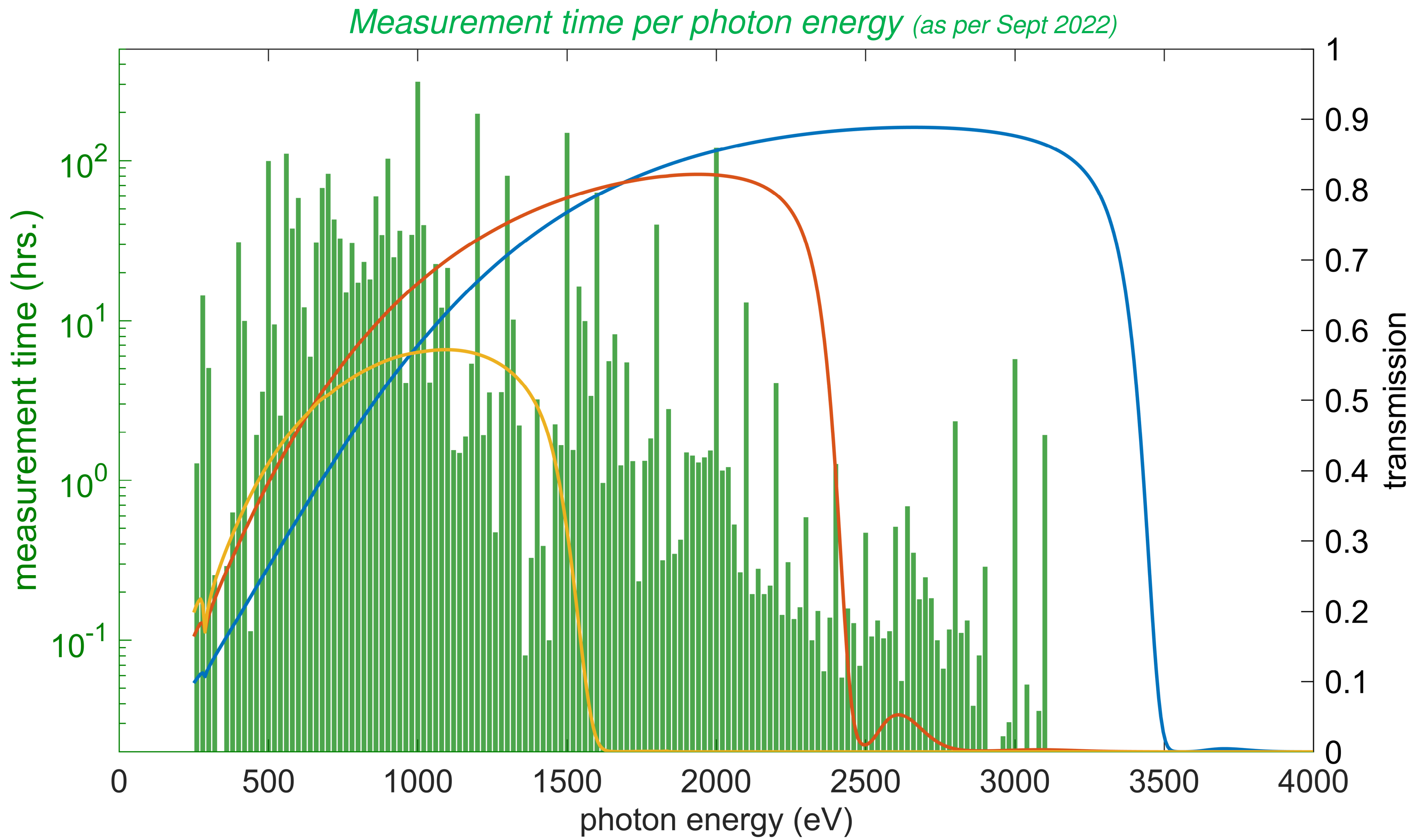
Science at the SQS instrument

Tommaso Mazza
European X-ray Free-Electron Laser Facility
Small Quantum Systems (SQS) Scientific Instrument

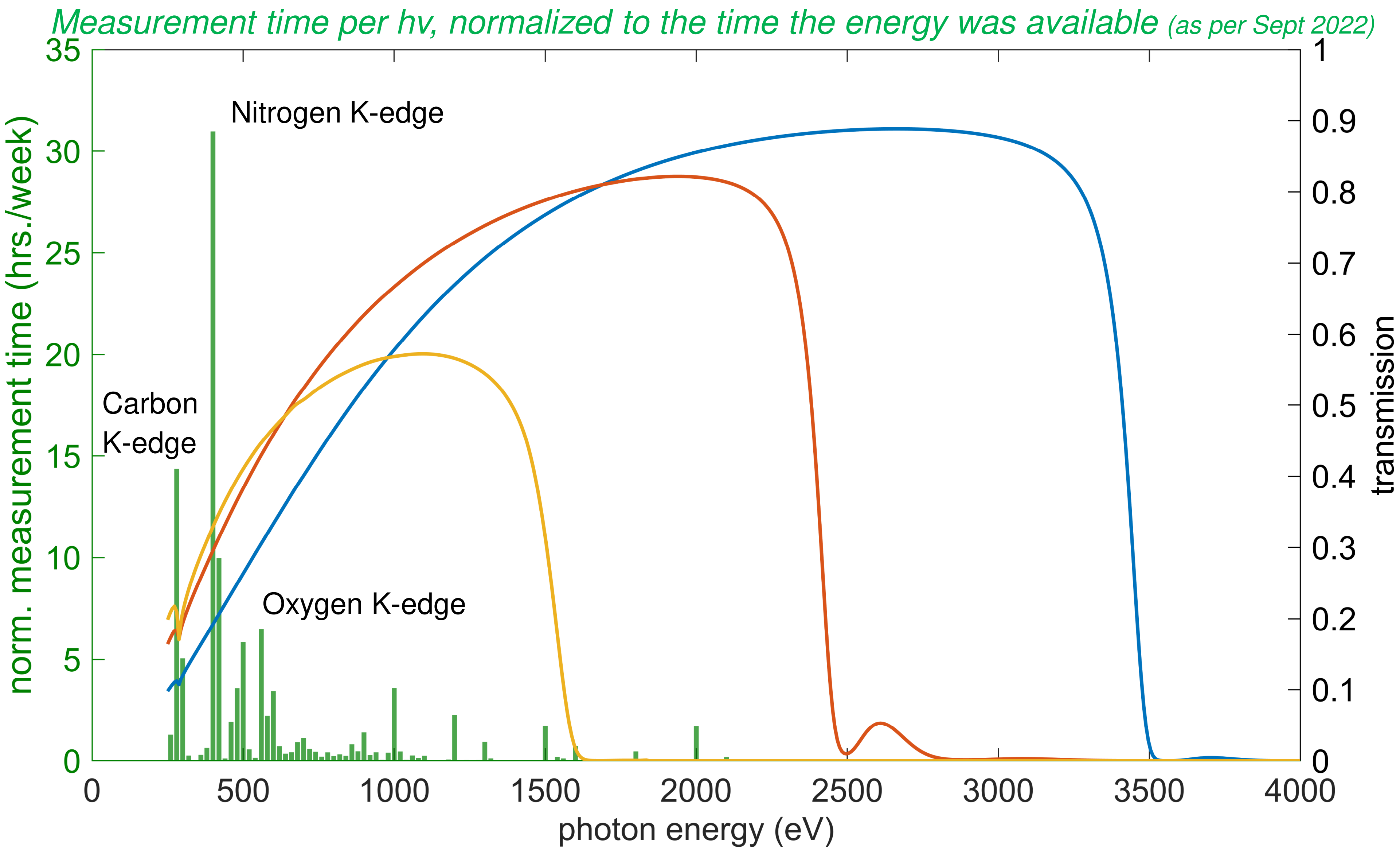
Eurizon 2020+ workshop
FEL linac driver and FEL physics applications
European XFEL Users' Meeting — Satellite meeting


electron tunnel
photon tunnel
undulator
electron switch
electron bend
electron dump
HED High Energy Density Science
MID Materials Imaging and Dynamics
Optional space for two undulators and four instruments
FXE Femtosecond X-ray Experiments
SPB SFX Single Particles, Clusters, and Biomolecules and Serial Femtosecond Crystallography
SQS Small Quantum Systems
SCS Spectroscopy & Coherent Scattering
linear accelerator
for electrons (10.5, 14.0, 17.5 GeV)
SASE 2
0.05 nm - 0.4 nm
SASE 1
0.05 nm - 0.4 nm
SASE 3
0.4 nm - 4.7 nm
Electron energy sets
C1s
N1s
O1s
SASE 3
P1s
S1s
SASE 1 / 2
16.3 GeV
0.92 – >3 keV
8.0 – 26 keV
14.0 GeV
0.66 – 2.5 keV
5.8 – 19 keV
11.5 GeV
0.5 – 1.5 keV
4.0 – 13 keV
8.0 GeV
0.25 – 1.0 keV
3.0 – 6.2 keV
FLASH
0.02 – 0.3 keV
200 eV
1 keV
10 keV

Beamline configurations, photon energies, transmission



Beamline configurations, photon energies, transmission



Scientific scope of SQS

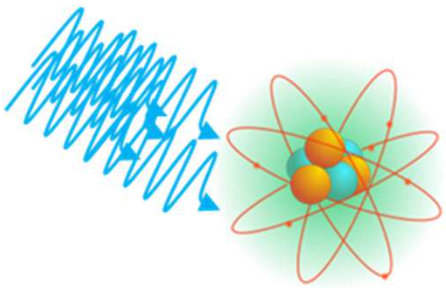
Investigations of the response of atoms, ions, molecules and clusters to intense x-rays and of their ultrafast dynamics

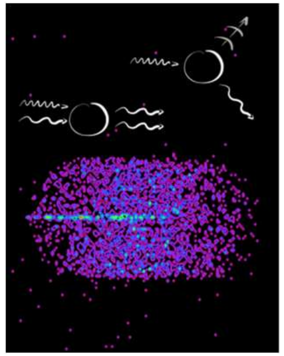
Non-linear phenomena

Intensity > 10^{18} W / cm^2

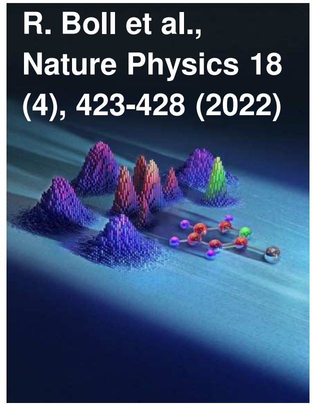
Multiple ionization

Multi-photon processes





U. Eichmann et al., Science 369 (2020) 1630



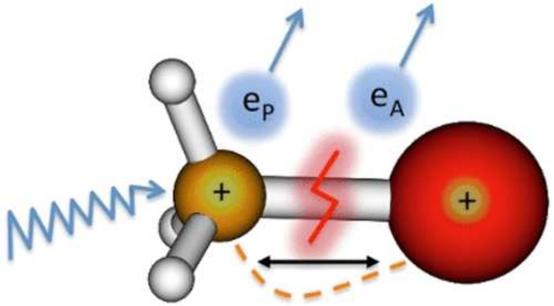
Time-resolved studies

Pulse durations: 2 - 100 fs

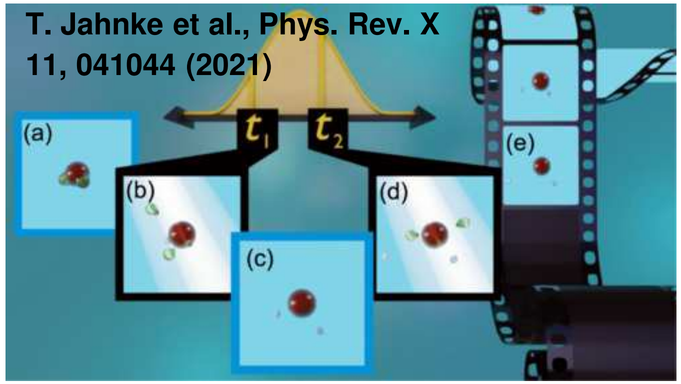
Pump-probe: NIR/XUV, XUV/XUV

Molecular dynamics

Element specific Soft X-rays



J. Phys. Chem. Lett. 2019, 10, 21, 6536–6544

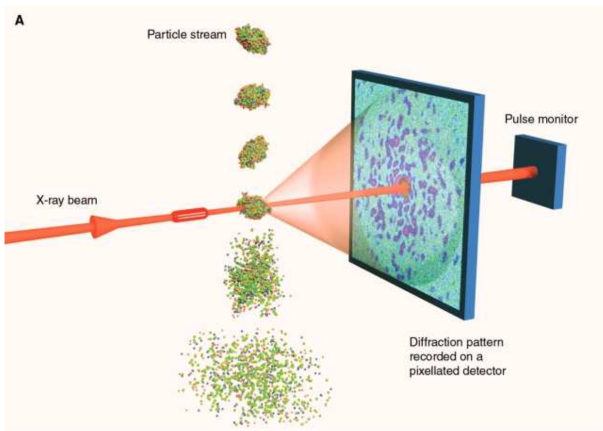


Imaging experiments

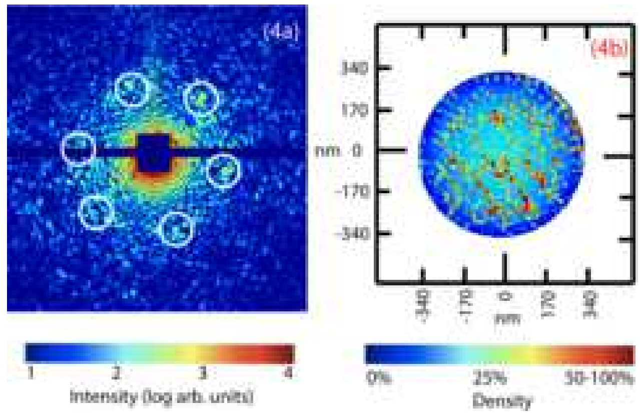
Spatial coherence

Size and shape selection

Cluster dynamics

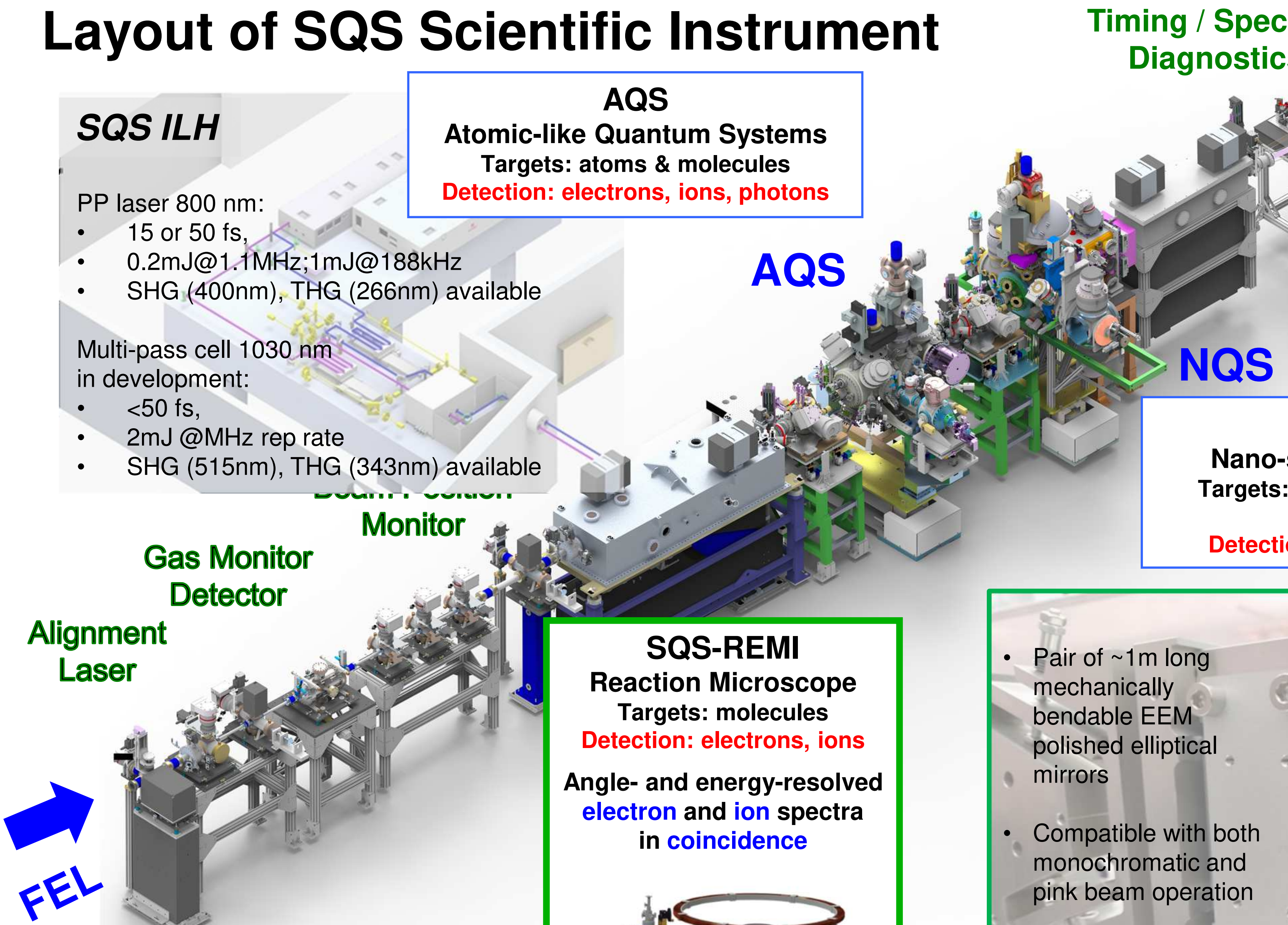


Science 316 (2007) 1444



A. J. Feinberg et al. Phys. Rev. Research 4, L022063

Layout of SQS Scientific Instrument



SQS ILH

PP laser 800 nm:
- 15 or 50 fs,
- 0.2mJ@1.1MHz;1mJ@188kHz
- SHG (400nm), THG (266nm) available

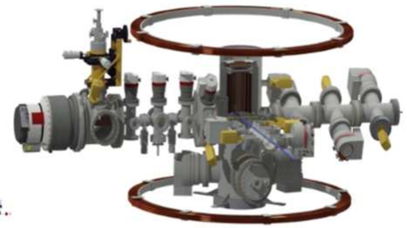
Multi-pass cell 1030 nm in development:
- <50 fs,
- 2mJ @MHz rep rate
- SHG (515nm), THG (343nm) available

AQS
Atomic-like Quantum Systems
Targets: atoms & molecules
Detection: electrons, ions, photons

NQS
Nano-sized Quantum Systems
Targets: Cluster, Nano-particles, bio-molecules
Detection: electrons, ions, photons

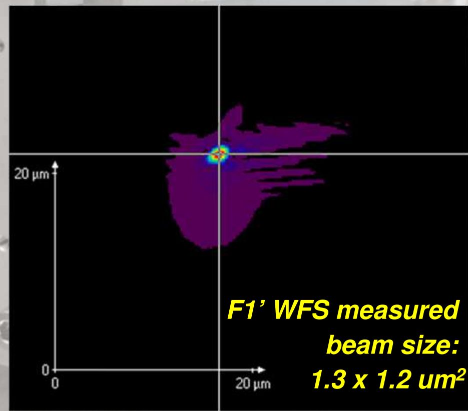
SQS-REMI
Reaction Microscope
Targets: molecules
Detection: electrons, ions

Angle- and energy-resolved electron and ion spectra in coincidence



KB Optics

- Pair of ~1m long mechanically bendable EEM polished elliptical mirrors
- Compatible with both monochromatic and pink beam operation
- Microfocus can be induced in 3 different interaction regions



Atomic-like Quantum Systems (AQS) Endstation

Non-linear interaction with X-rays and ultrafast dynamics of atoms and small molecules

Six electron time of flight spectrometers (eTOF)

- 0.14% of 4π, max rep. rate = 4.5MHz
- E/ΔE > 10000, E_{max}=3000 eV.

A. De Fanis et al., J. Synchr. Rad. 29, 755 (2022)

Ion spectrometer

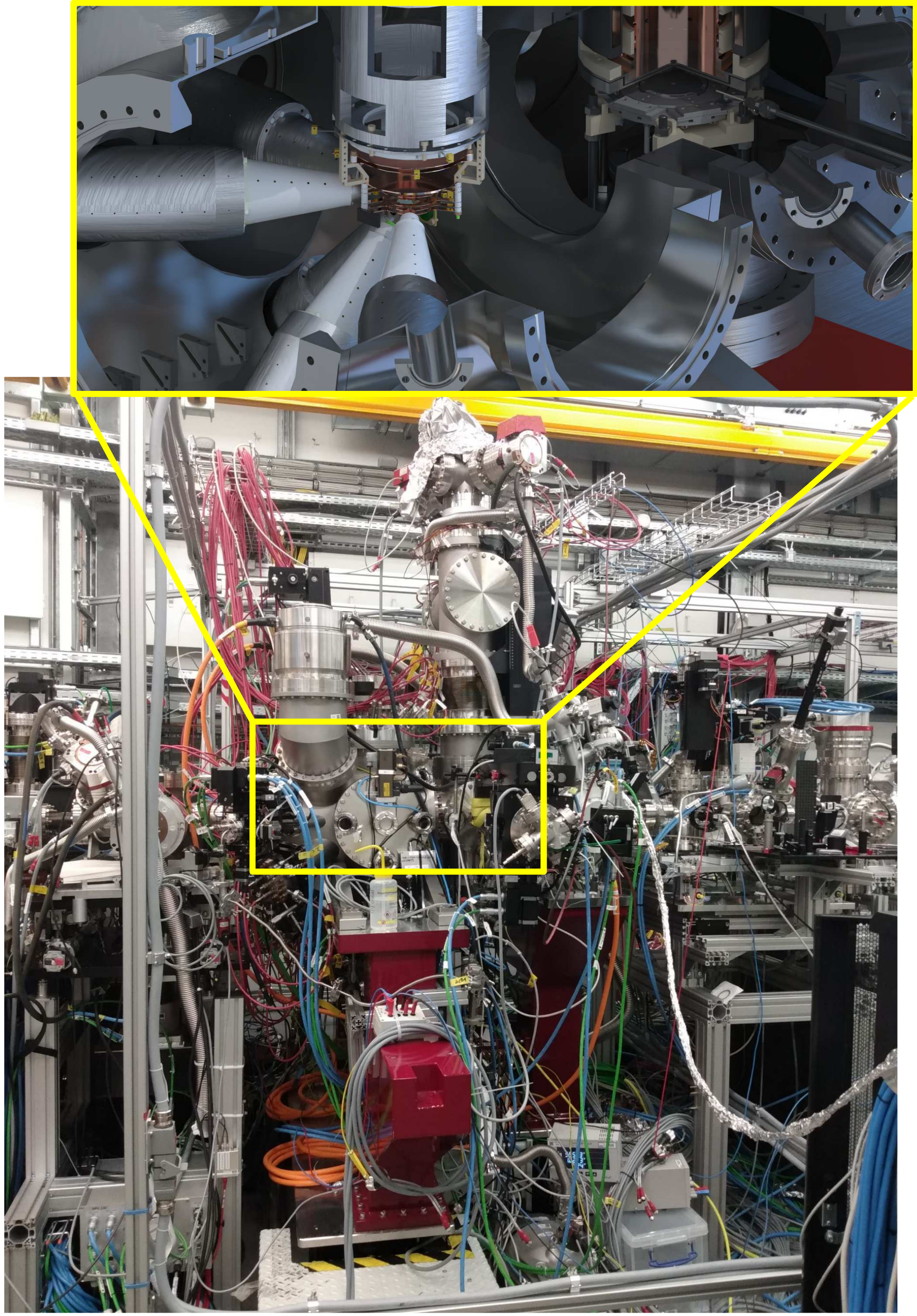
- m/Δm > 200, Electrostatic focus tuning for KER resolution

Velocity map imaging spectrometer (VMI)

- 4π collection, 10 Hz (Timepix integration planned);
- E/ΔE >50, E_{max}=1200 eV

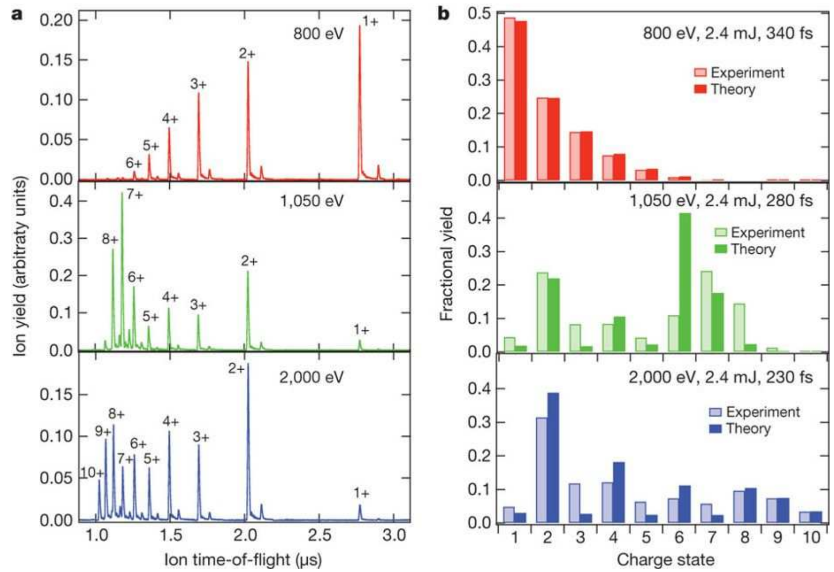
Magnetic bottle electron spectrometer

- 4π e-collection + short i-TOF
- coincidence/covariance studies

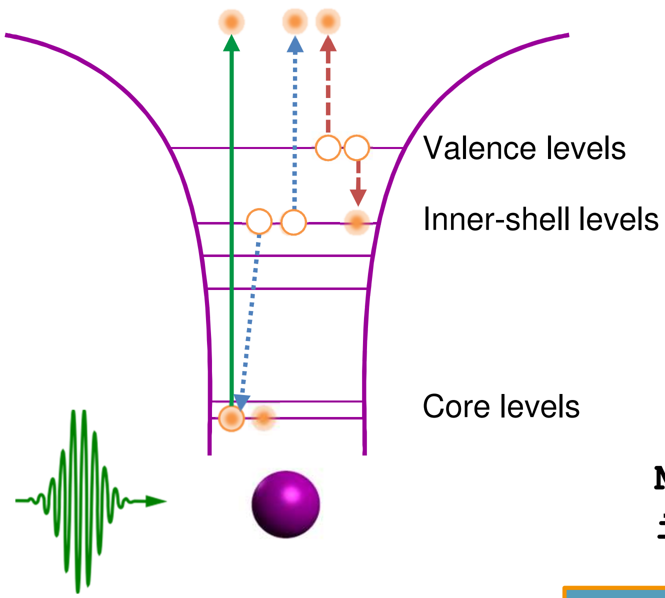


The interaction of atoms with intense x-ray pulses

- The response of atoms (and molecules and clusters) to intense x-ray pulses **has been the first target of FEL investigations**; even for the simplest systems ***the response dynamics is yet to be understood*** under several aspects!
- **Ground observation:** The non-linear interaction of atoms with x-rays (many photons on the same atom!) results in the production of high charge states [Case of Neon, Young 2010].

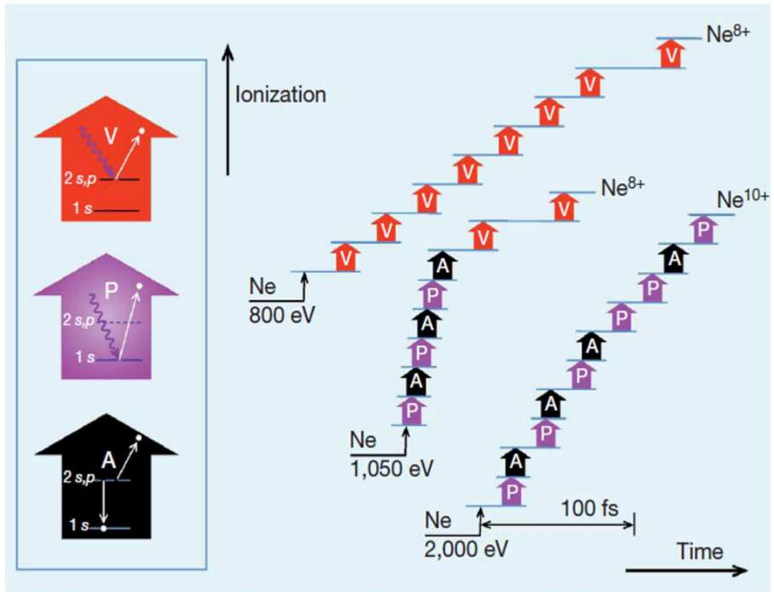


Young et al., Nature 466, 56 (2010)

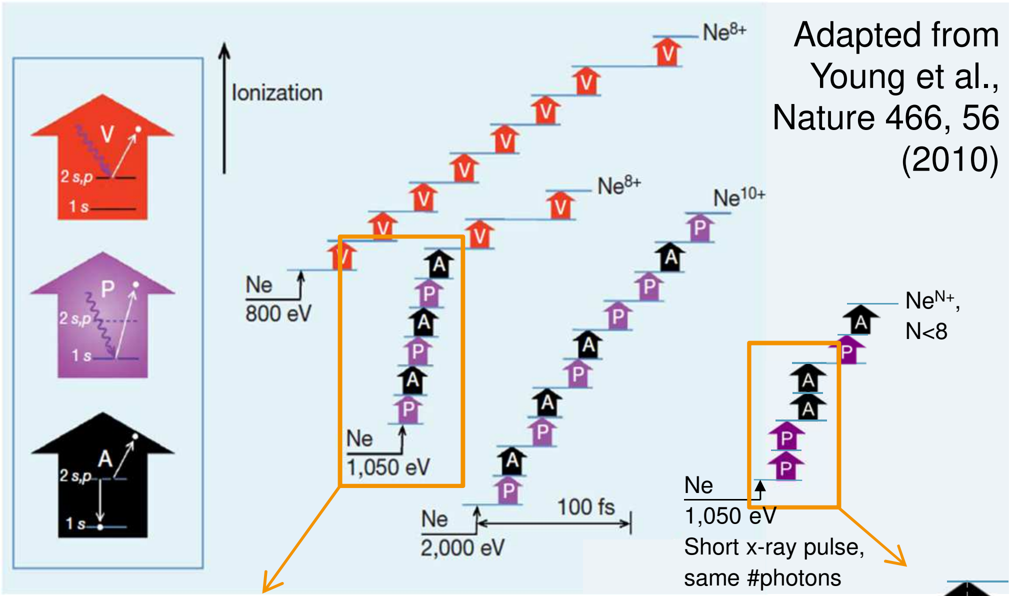


MPI: repeat in sequence

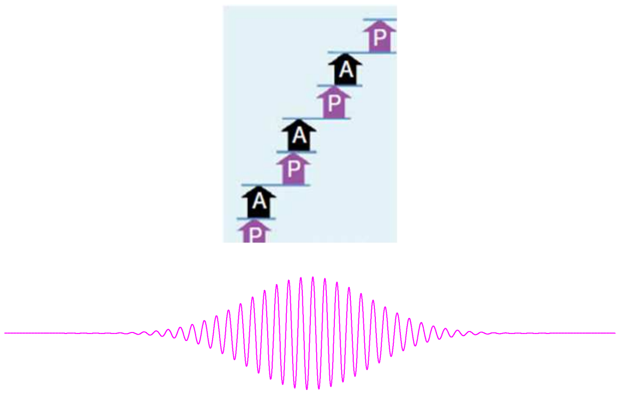
Is this all what we have to tell about it?



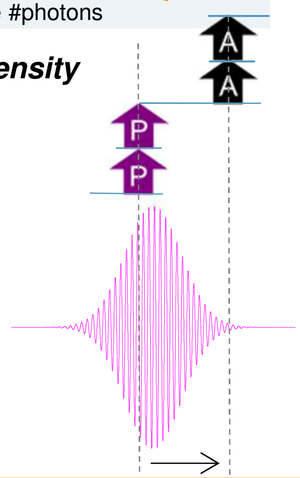
Competition between sequential ionization and ultrafast Auger decay: hollow atoms



Low intensity



High intensity



Sequential ionization occurs during the whole pulse duration

Multiple core hole ionization → hollow atom → induced transparency

core hole lifetime vs. intensity dependent ionization rate

- Lifetime Ne1s: 2.4 fs
- *$Ne^{+}1s^{1}2s^{2}2p^{6}$ → $Ne^{2+}1s^{0}2s^{2}2p^{6}$* cross section @ 1.05keV ~ 100kbarn
- DCH rate:

$$r = \sigma[cm^2] \cdot I\left[\frac{W}{cm^2}\right]/h\nu[J]$$

$$\approx 0.7 \cdot 10^{14} Hz\ per\ 10^{18}\frac{W}{cm^2}$$

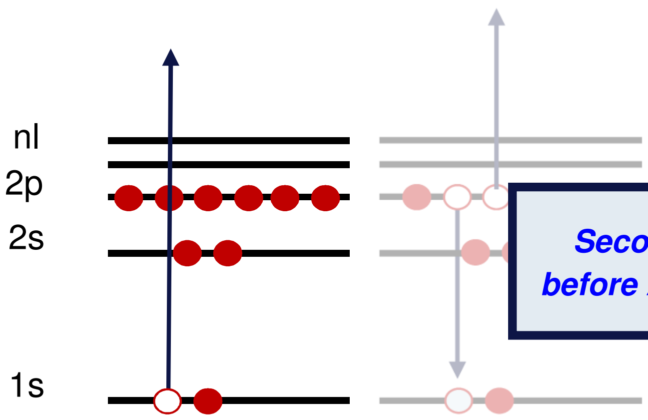
► This means: $\frac{1}{r} = 2.4fs$

with $I = 6 \cdot 10^{17}\frac{W}{cm^2}$ $\sim \mathbf{10^{18}\frac{W}{cm^2}}$

***CDI**: hollow atoms make X-ray scatter without creating damage*

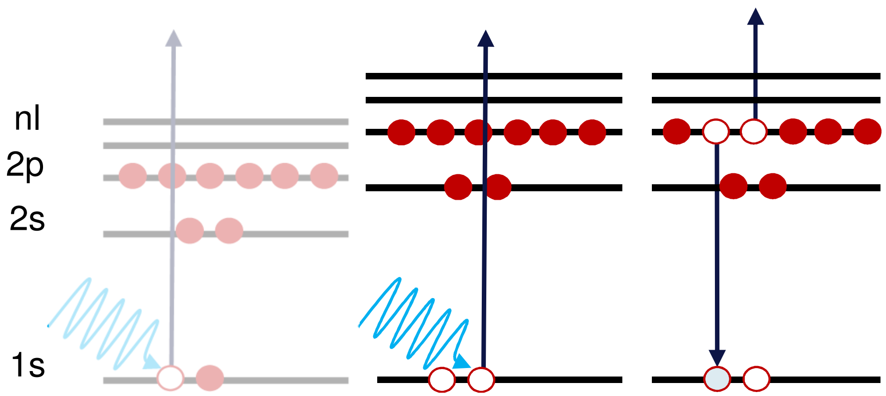
***Enhanced chemical sensitivity** of double core hole photoemission*

Competition between sequential ionization and ultrafast Auger decay: probing the transient ion



Core level ionization + Auger decay (linear regime)

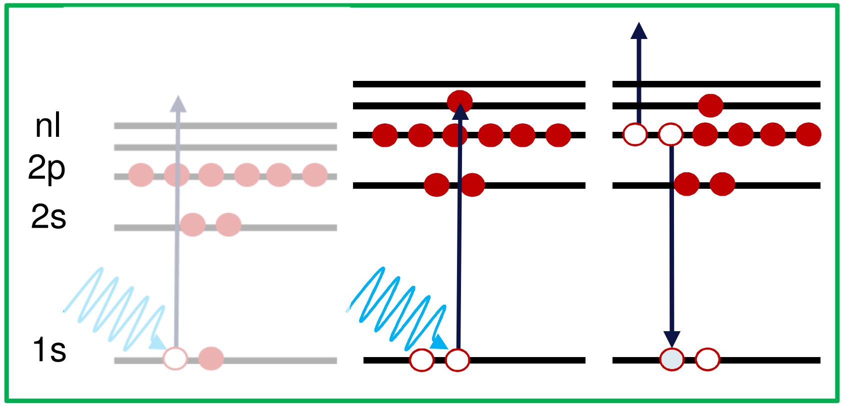
Second photon before Auger decay



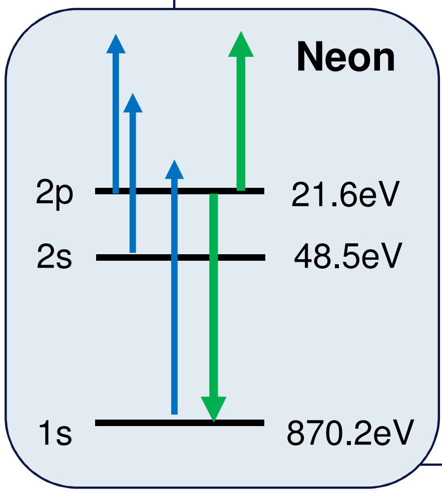
A second photon makes the atom hollow:

It can be a probe on the transient electronic structure

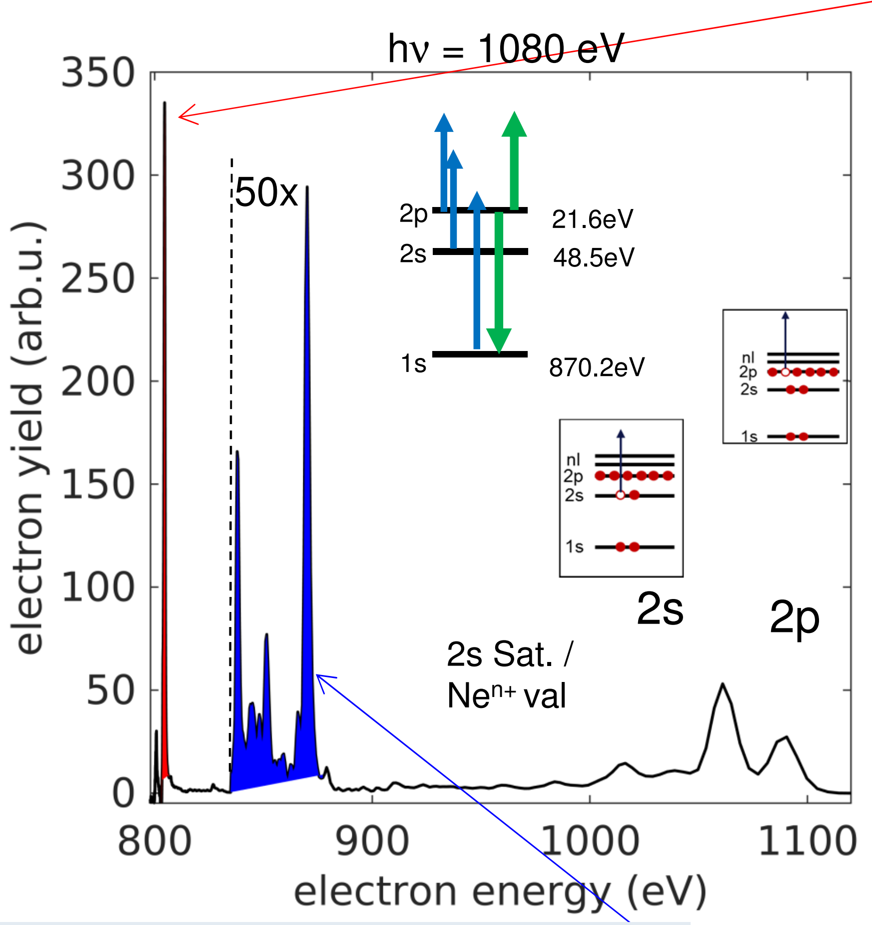
Tuning the second photon to the ion resonant excitation

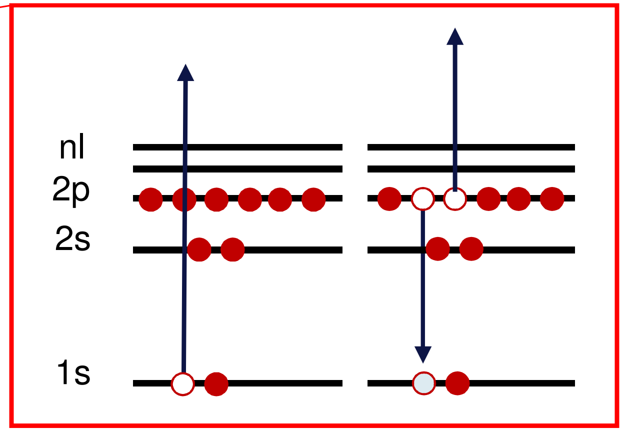


Spectroscopically study the transient core-excited ion by mapping its resonant electronic structure



Sequential 2-core hole ionization electron spectroscopy



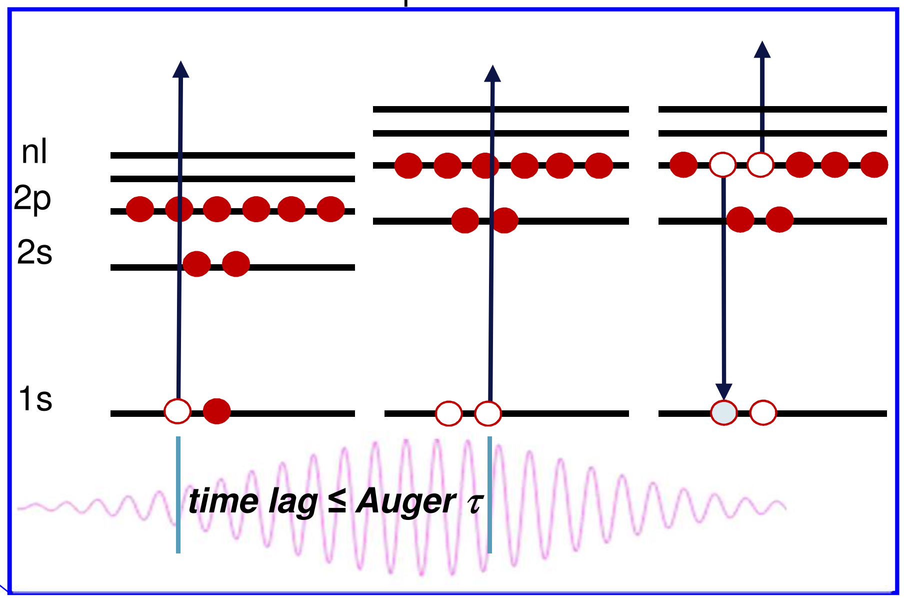


KLL Auger

Single Core Hole: Ne $1s^2 2s^2 3p^6$ + hν → Ne$^+$ $1s^1 2s^2 3p^6$ +e

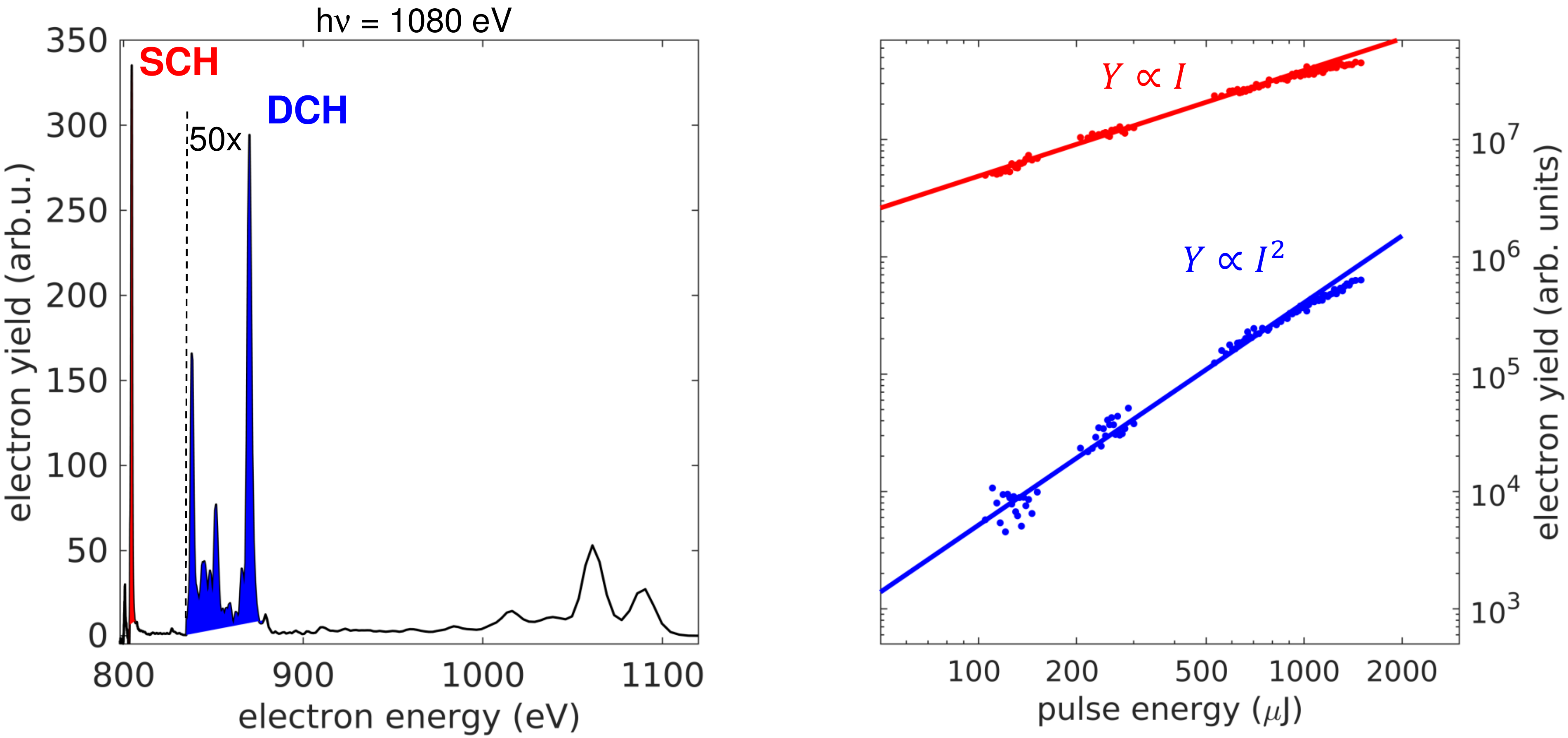
Double Core Hole: Ne $1s^2 2s^2 3p^6$ + 2hν → Ne$^+$ $1s^0 2s^2 3p^6$ +e

Double core hole Auger



hv =	800-1200 eV
Focus =	1.5 um^2,
Pulse energy @sample =	1mJ,
Pulse duration (guess) =	20fs
Estimated intensity =	**1 10^{18} W/cm^2**

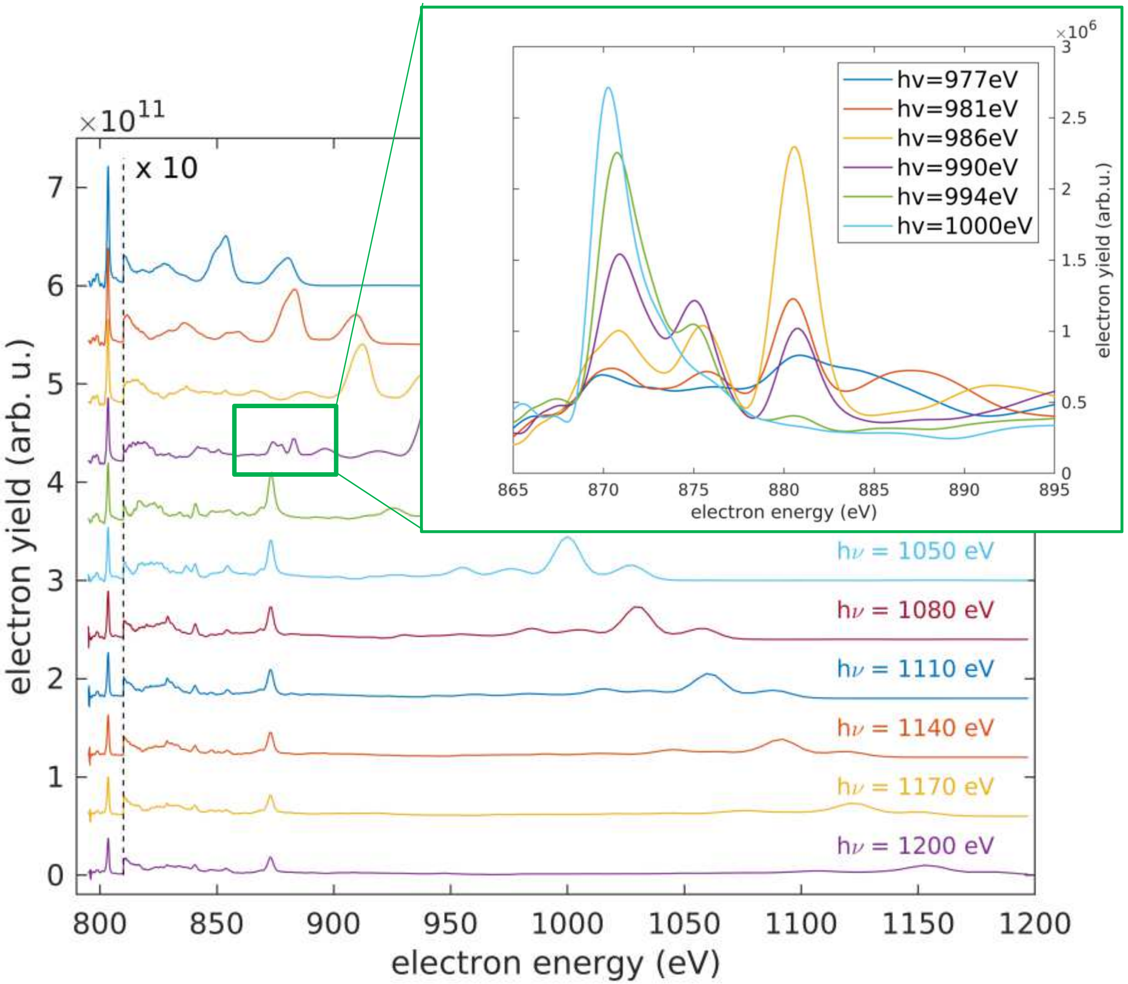
Sequential 2-core hole ionization electron spectroscopy: intensity dependence



SCH: Single Core Hole: Ne **1s**^{2}2s^{2}3p^{6} + **hν** → Ne^{+}**1s**^{1}2s^{2}3p^{6} + **e**$^{-}$

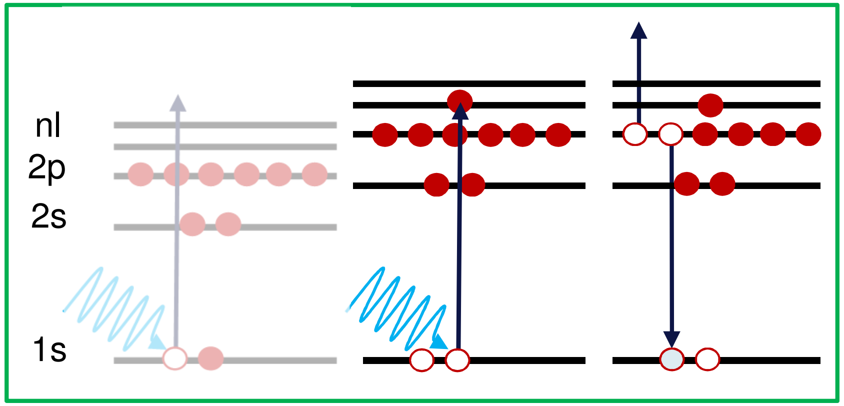
DCH: Double Core Hole: Ne **1s**^{2}2s^{2}3p^{6} + **2hν** → Ne^{+}**1s**^{0}2s^{2}3p^{6} + **e**$^{-}$

Sequential 2-core hole ionization electron spectroscopy: photon energy dependence



$Ne1s^1 2s^2 2p^6 + hv \rightarrow$

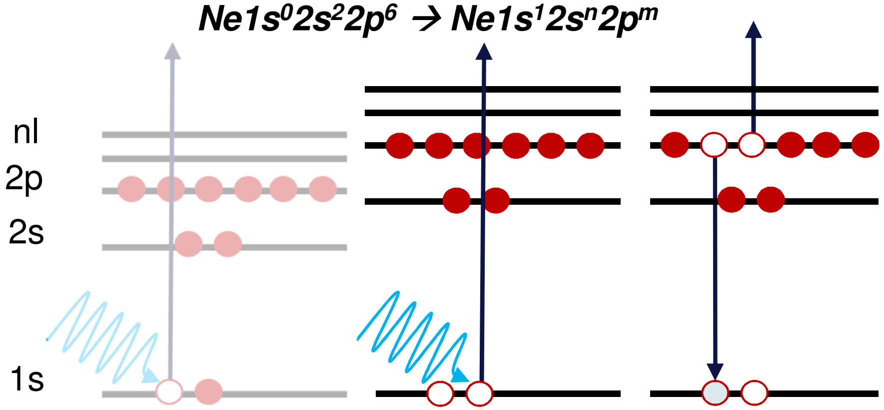
$Ne1s^0 2s^2 2p^6 np \rightarrow Ne1s^1 2s^m 2p^{m'} np^{m''}$



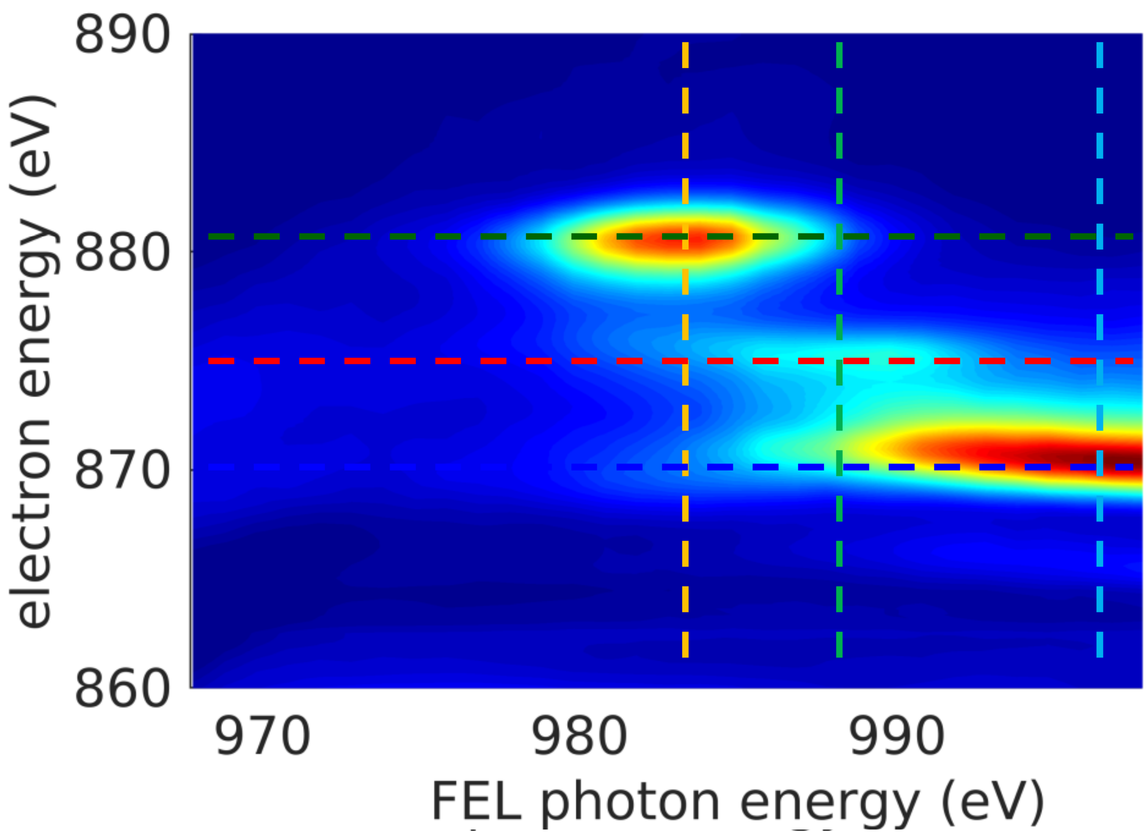
$Ne1s^2 2s^2 2p^6 + hv \rightarrow Ne1s^1 2s^2 2p^6$;

$Ne1s^1 2s^2 2p^6 + hv \rightarrow$

$Ne1s^0 2s^2 2p^6 \rightarrow Ne1s^1 2s^n 2p^m$



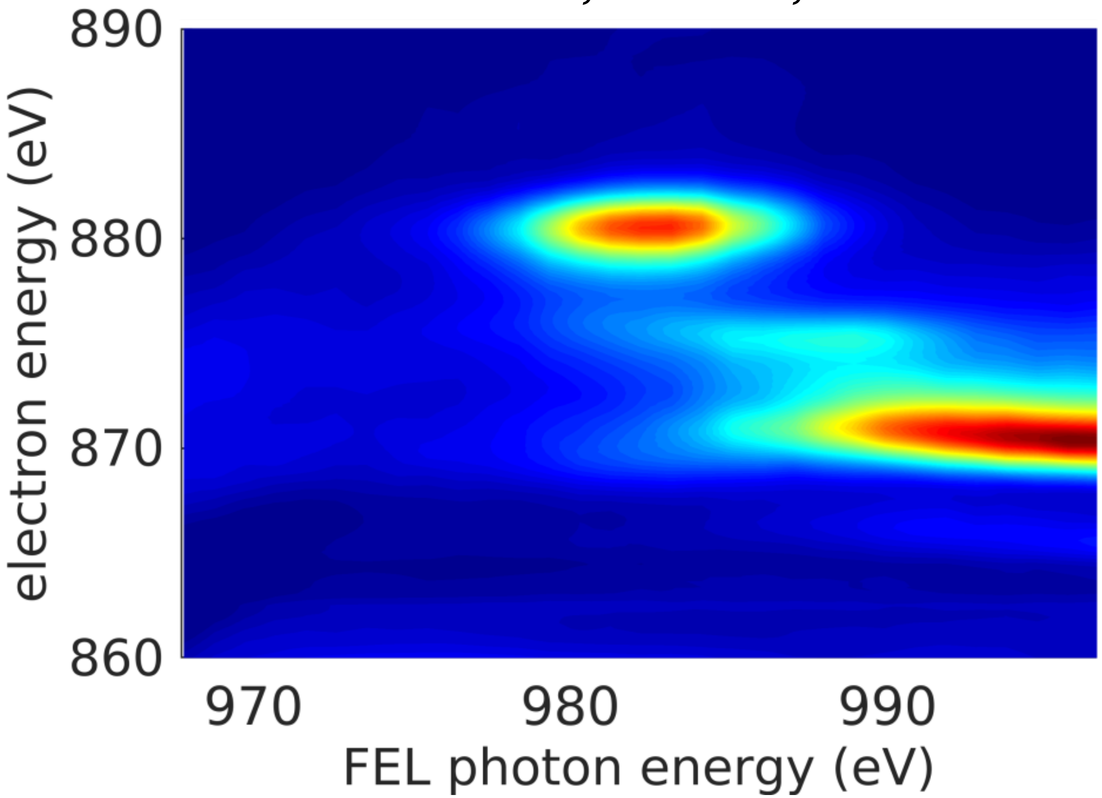
Double core hole resonance in Neon: probing the electronic structure of the core-excited ion

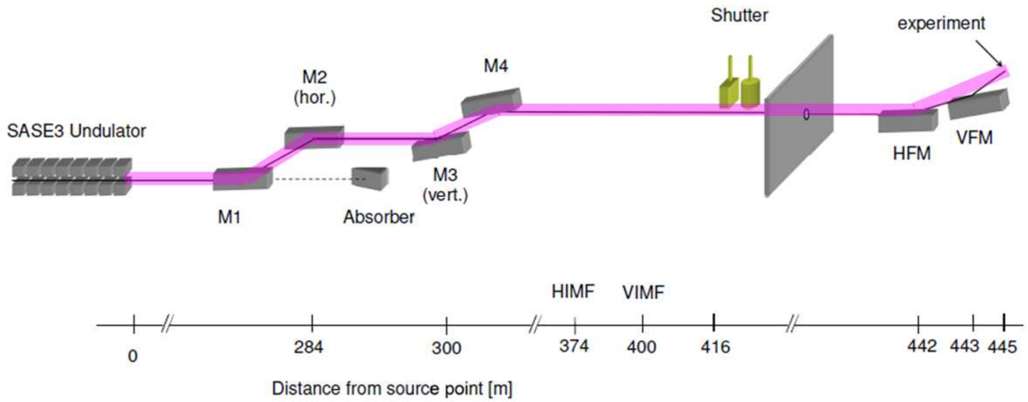


T. Mazza et al., PRX 10, 041056 (2020)

Next step: Non-linear interaction with monochromatic X-rays

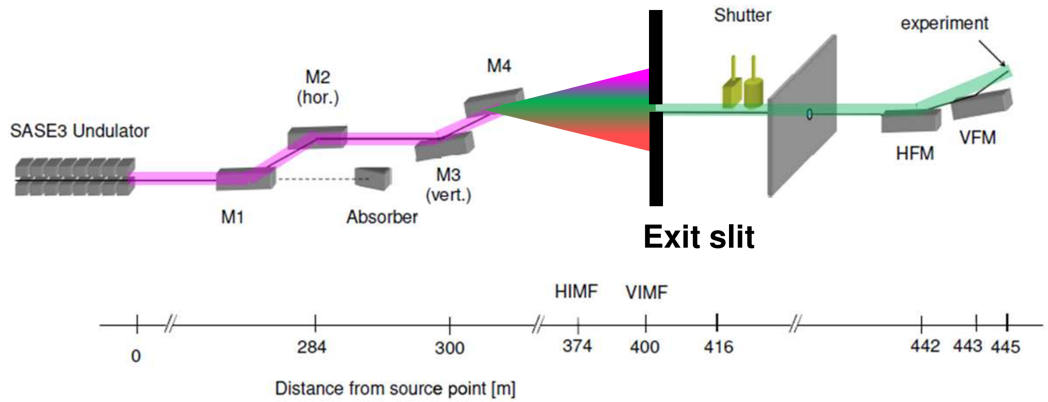
SASE beam, bw ~ 1%, T~65%





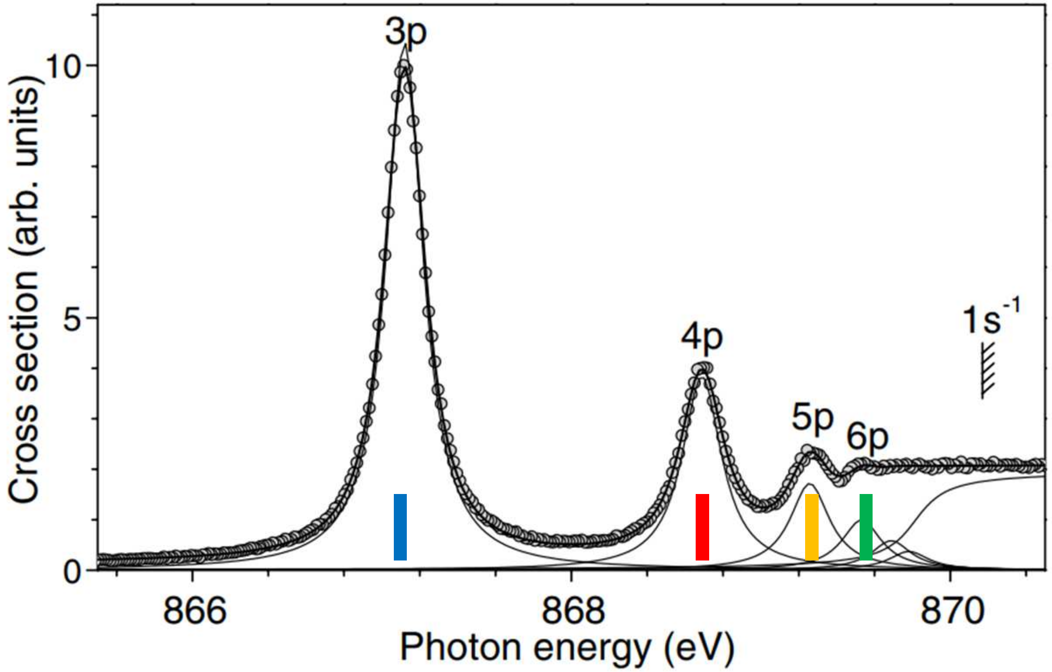
Mono beam, bw ~ 0.05%, T~0.2%

unpublished



Ne1s^{0}2s^{2}2s^{6}np resonances positions and linewidths

neutral Ne (A. De Fanis et al., PRL 89, 243001 (2002))



	Γ (meV)	E_0 (eV)
1s^1 3p	254	867,12
1s^1 4p	246	868,69
1s^1 5p	250	869,27
1s^1 εp	271	870,17

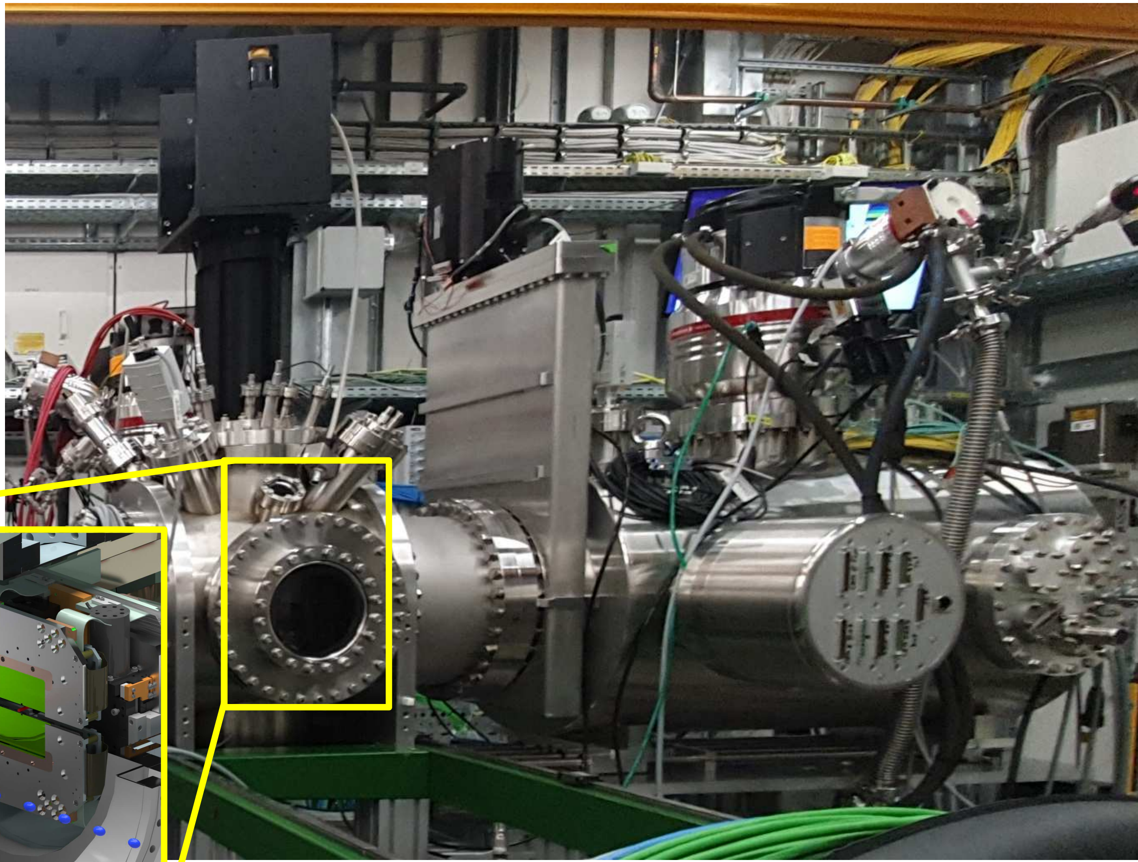
from Coreno et al., PRA 59 2494 (1999)

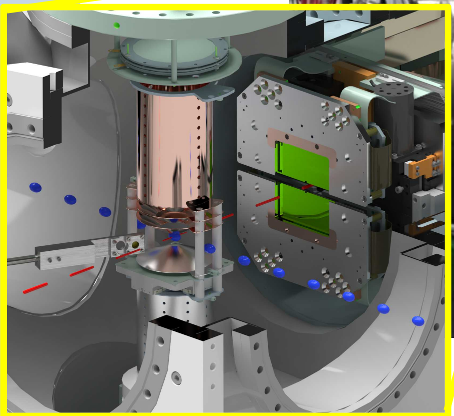
Core excited Ne+ with mono

unpublished

Nano-size Quantum Systems (NQS)

Structure and ultrafast dynamics of clusters, nanoparticles and biomolecules





iTOF + VMI:

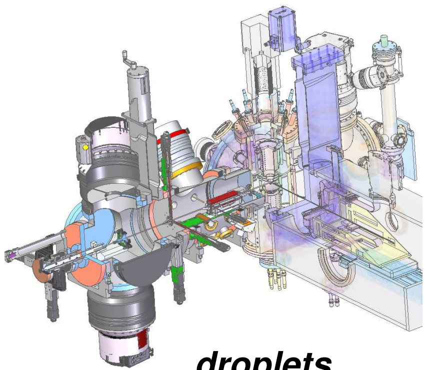
- iToF mass resolution m/Δm:
  - 450 for thermal ions in VM&iTOF mode
  - 1000 for thermal ions in iTOF mode
- VMI mass resolution m/Δm:
  - 200 for thermal ions
  - 30 for 400eV kinetic energy ions
- Electron VMI:
  - Energy range E<850eV
  - energy resolution 3%

Scattering detectors

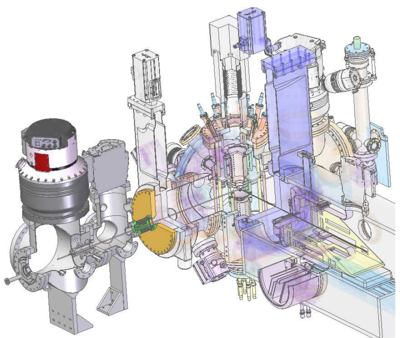
- pnCCD (10Hz operation, high dynamic range)
- Mini-SDD (DSSC, MHz rep rate, lower dynamic range)

Sample delivery

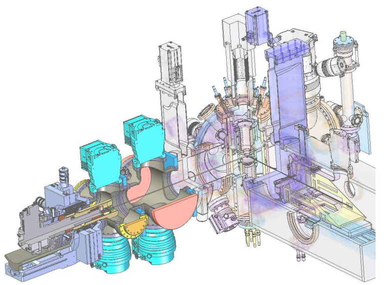
- Rare gas clusters / He droplets
- Aerosol source
- COntrolled MOlecules set up
- Pulsed Microplasma Cluster Source



droplets



PMCS

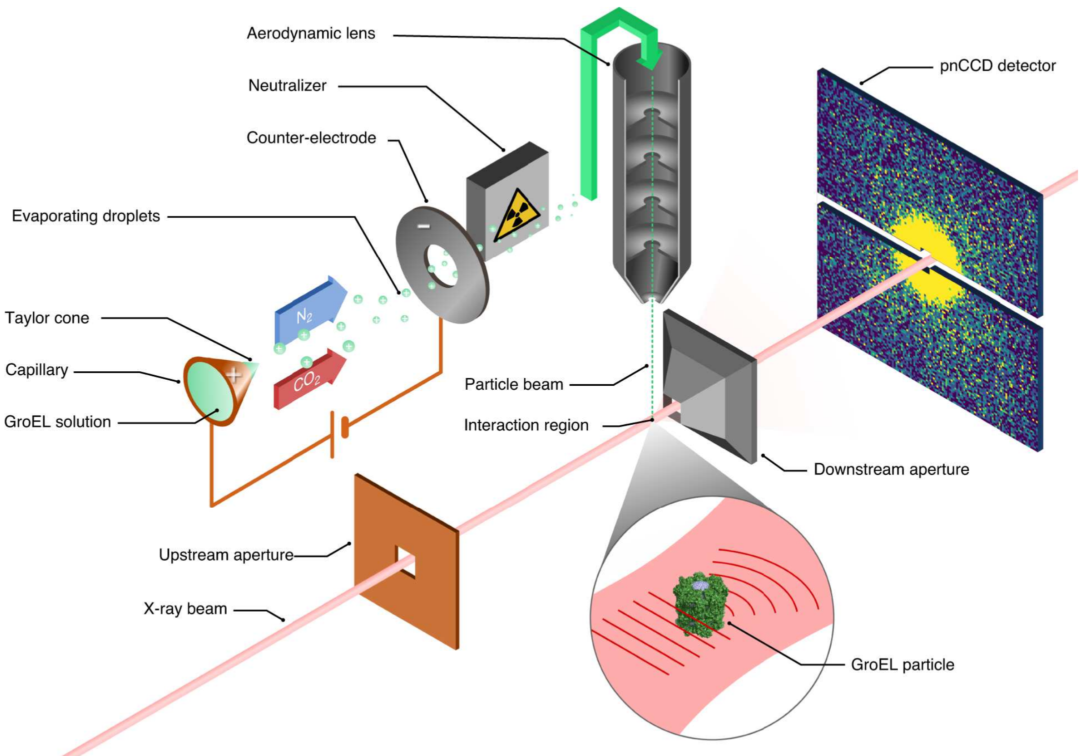


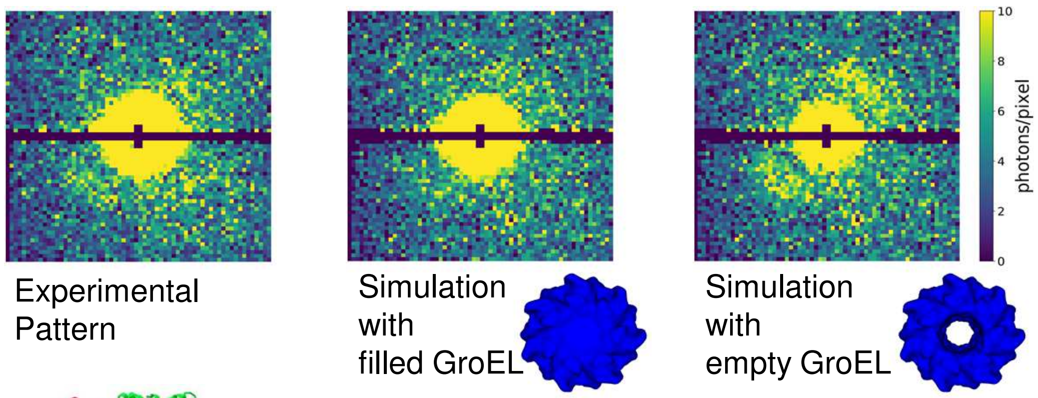
COMO

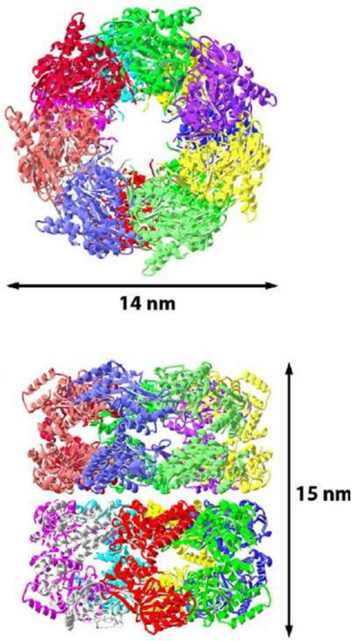
Coherent Diffraction Imaging Experiments at SQS

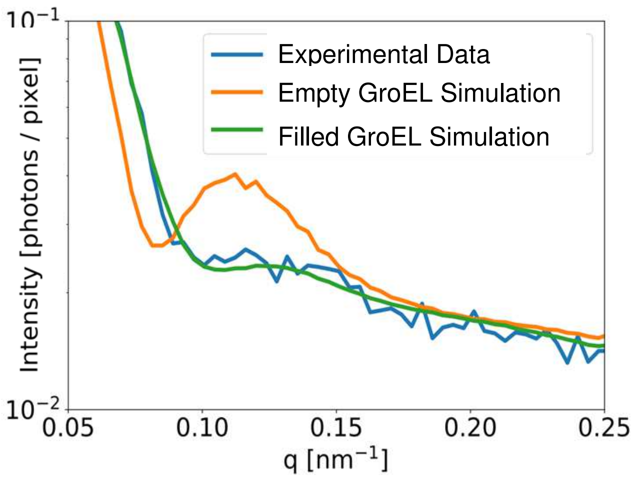
PI: F. Maia (U Uppsala)

X-ray Imaging of Single Proteins









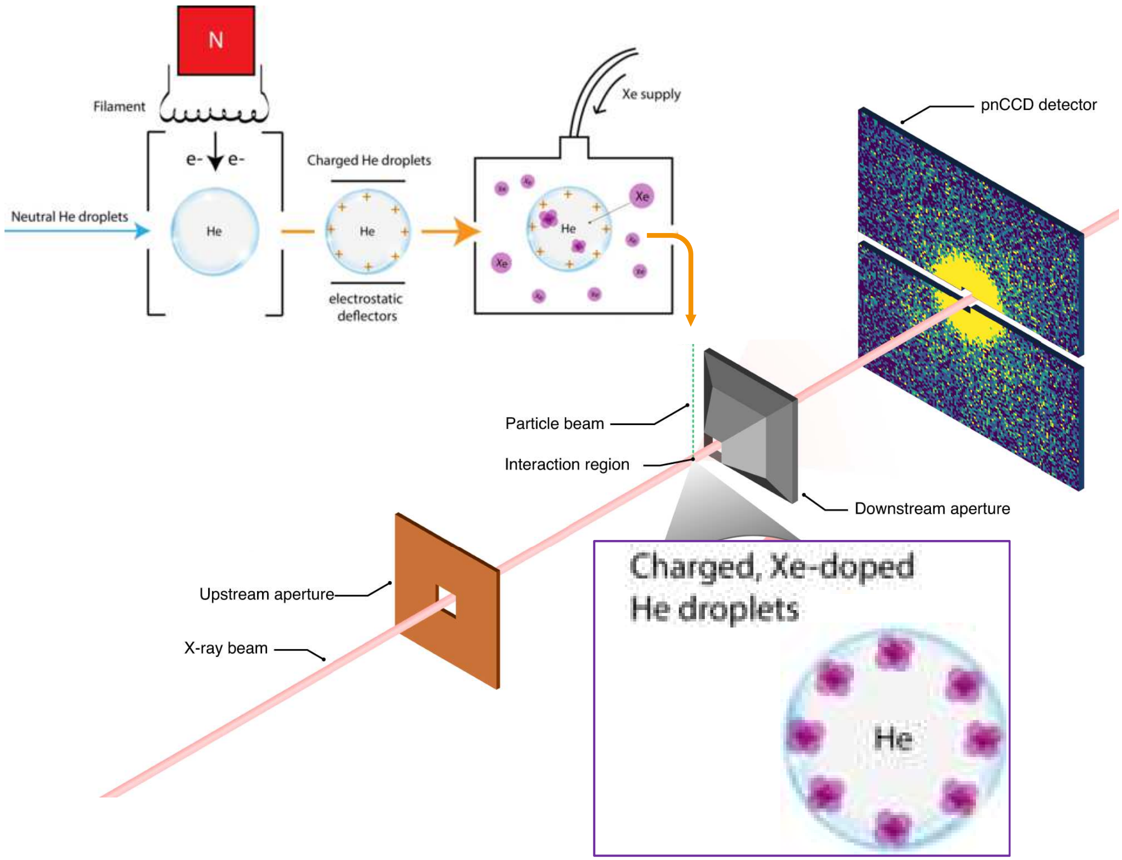
- "Diffraction before destruction" concept proven
- Approximate orientation of the protein determined
- Probing the hydration condition of the protein

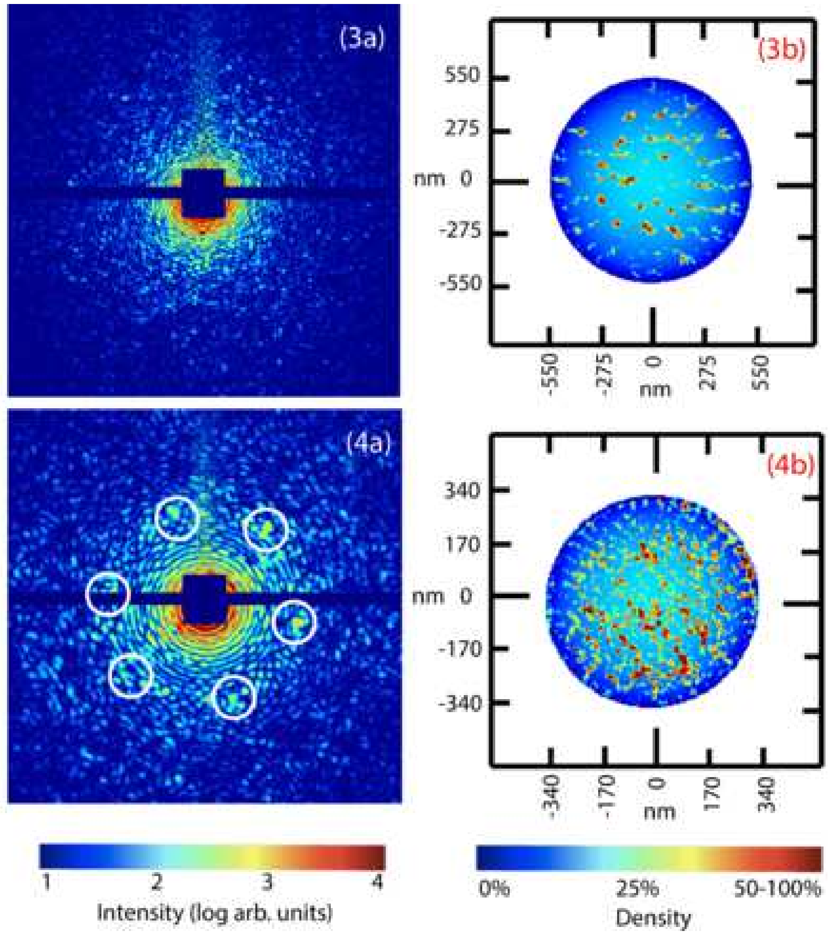
T. Ekeberg et al., Light: Science & Applications (2024)13:15

Coherent Diffraction Imaging Experiments at SQS

PI: A. Vilesov (U Southern California)

Imaging highly charged superfluid He nanodroplets





- General problem: determination of the minimum energy configuration of charges on a sphere;
- The sphere is a He droplet; the charges are “marked” with dopant atoms (Xe)

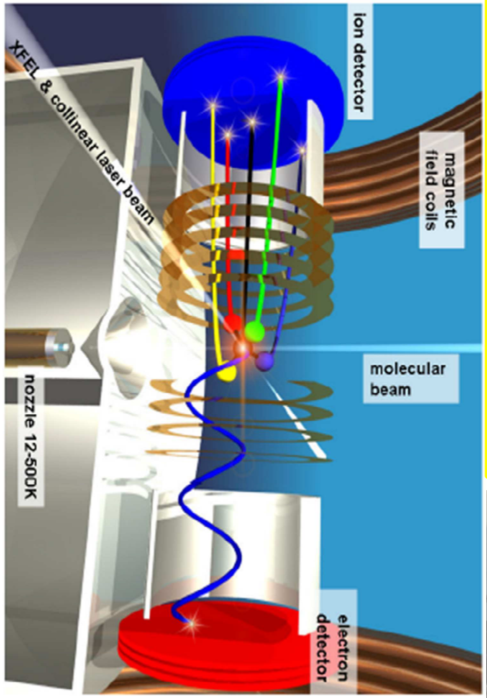
A. Feinberg et al., Phys. Rev. Research 4, L022063 (2022)

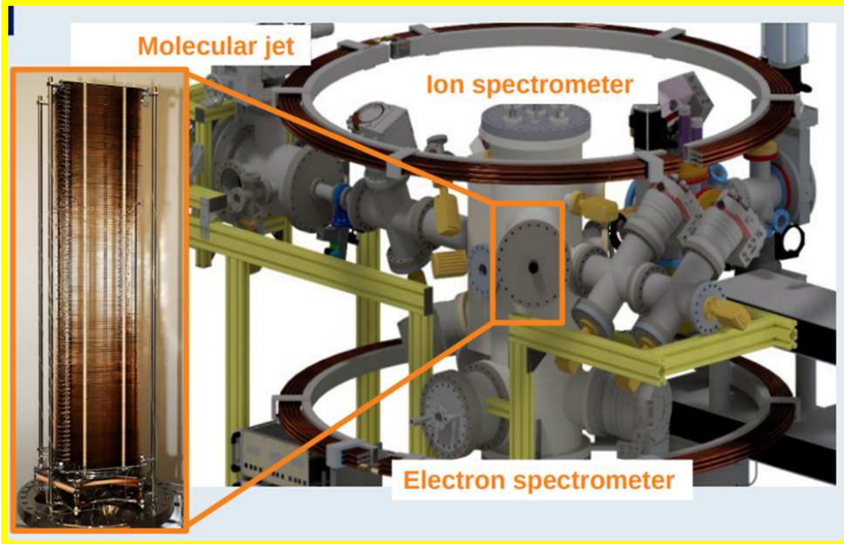
Reaction Microscope (REMI)

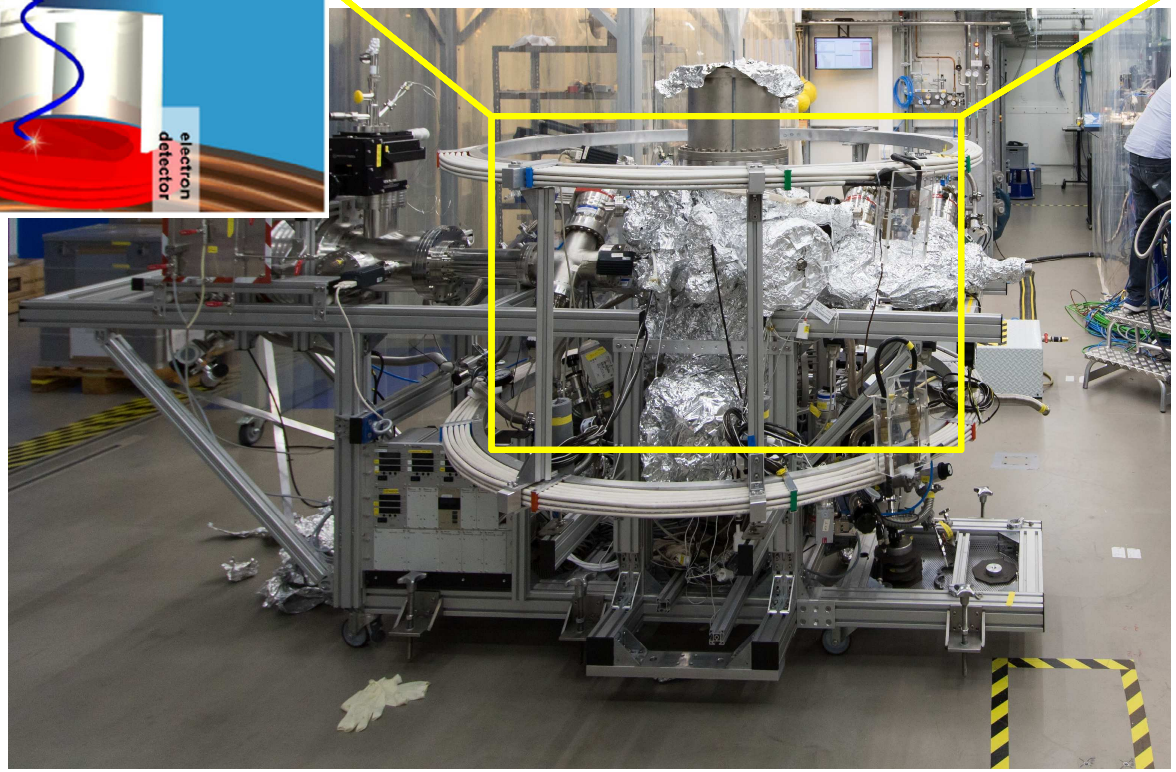
COLTRIMS experiments on atoms and molecules

Ions and electrons in coincidence
- Kinetic energies: 0-50 eV (ions); 0-500 eV (electrons);
- momentum resolution: 1%;
- mass resolution: 1%;
- max rep. rate ~ 200kHz

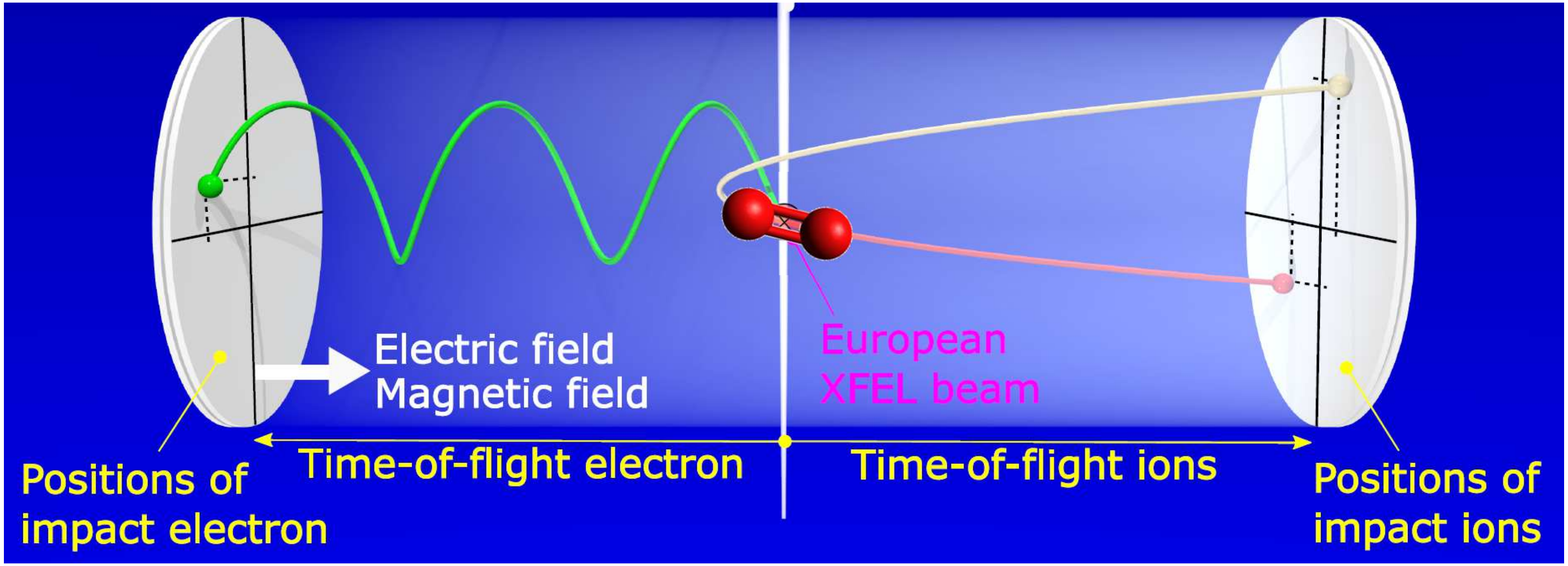
Sample: supersonic jet, 5-450K







Reaction Microscope (REMI)

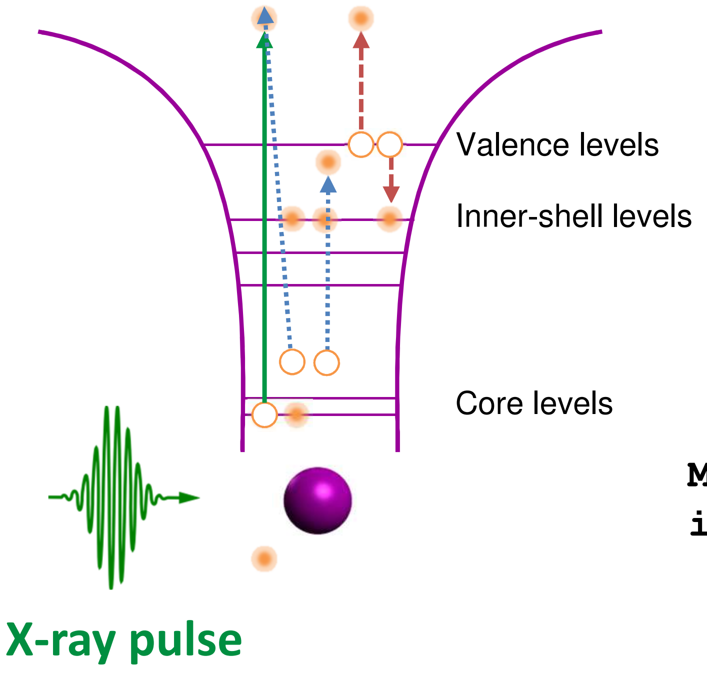


- record flight time as well as (x,y) position of each ion and electron
  - reconstruct 3d momentum
  - coincidence measurements possible if at most 1 molecule ionized per pulse
- gas-phase molecules are randomly oriented!
  - but: measured 3d ion momenta in coincidence allow to “align” them in the analysis

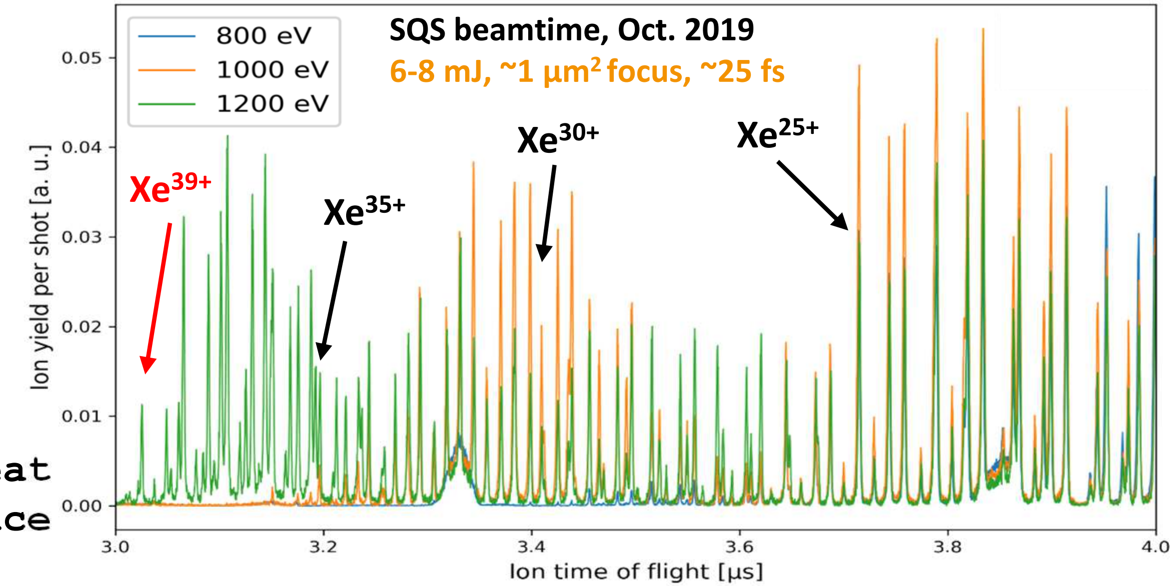
Slide courtesy R. Boll, T. Jahnke

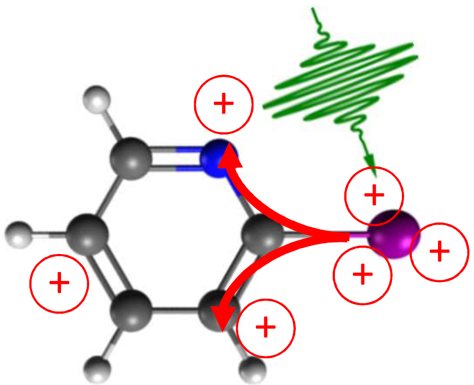
G. Kastirke et al., Phys. Rev. X **10**, 021052

fs-MPI in molecules enabling Coulomb explosion imaging



MPI: repeat in sequence

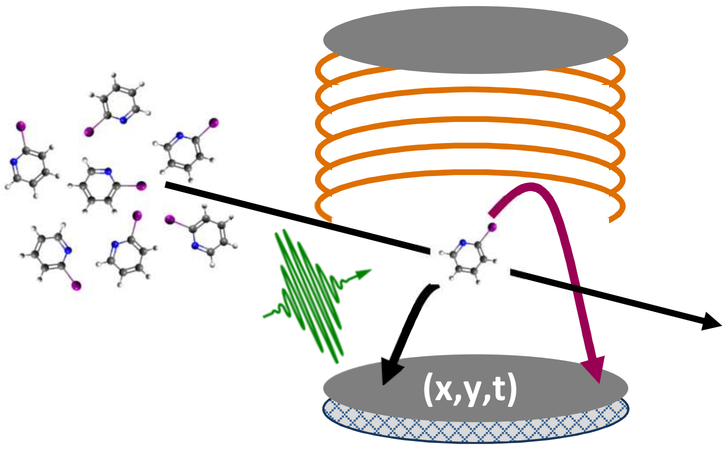




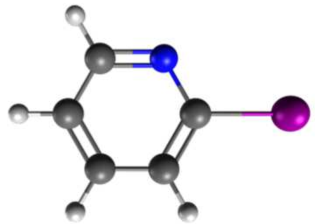
C_5H_4NI

high charges and efficient redistribution leads to prompt Coulomb explosion

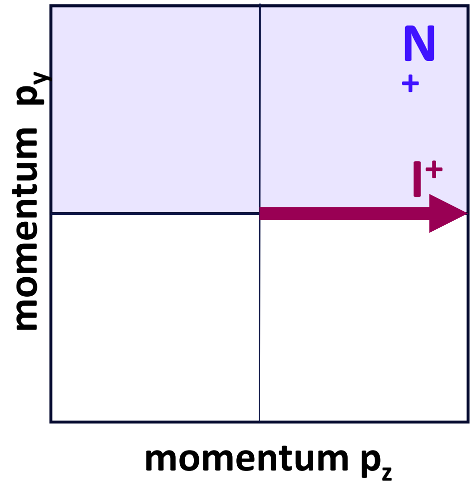
Multi-ion coincidence analysis



2-iodopyridine

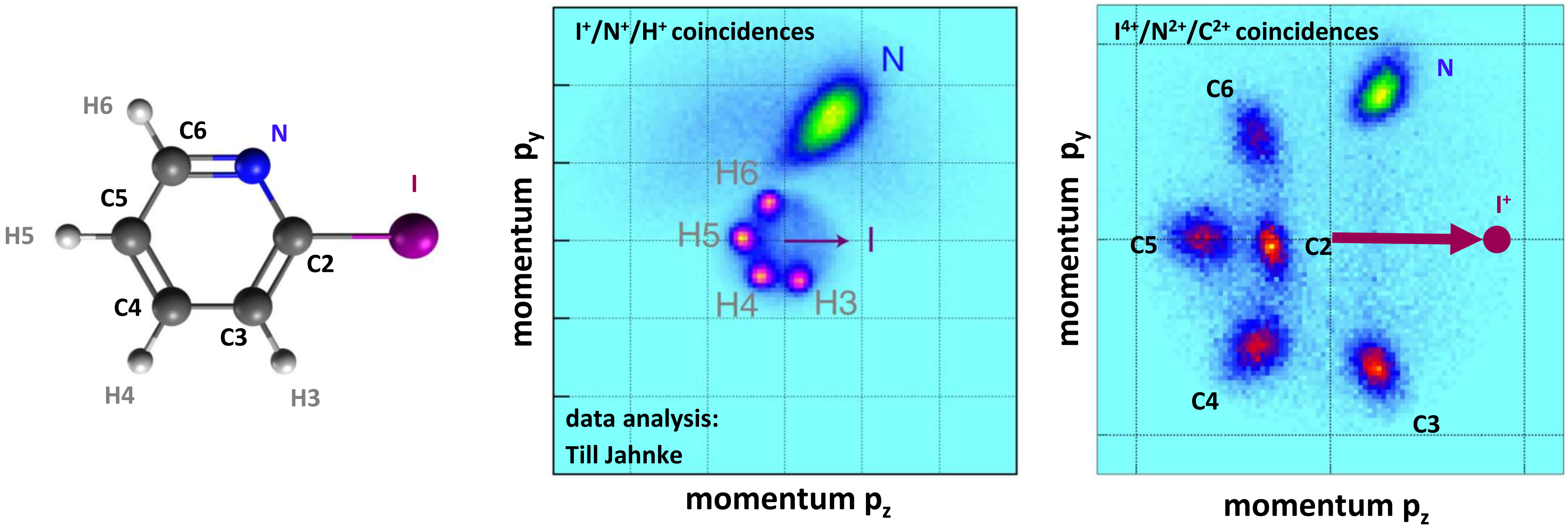


C_5H_4NI



- image the fragmentation following X-ray ionization in the molecular frame
- gas-phase molecules are randomly oriented!
- but: measured 3d ion momenta in coincidence allow to “align” them in the analysis
- create Newton plot of 3 (or more) ions recorded in the same FEL shot
  - make iodine momentum point towards $p_x = p_y = 0$, $p_z = 1$
  - make nitrogen momentum point towards $p_x = 0$, $p_y > 0$
  - plot momentum of any third particle in this coordinate frame

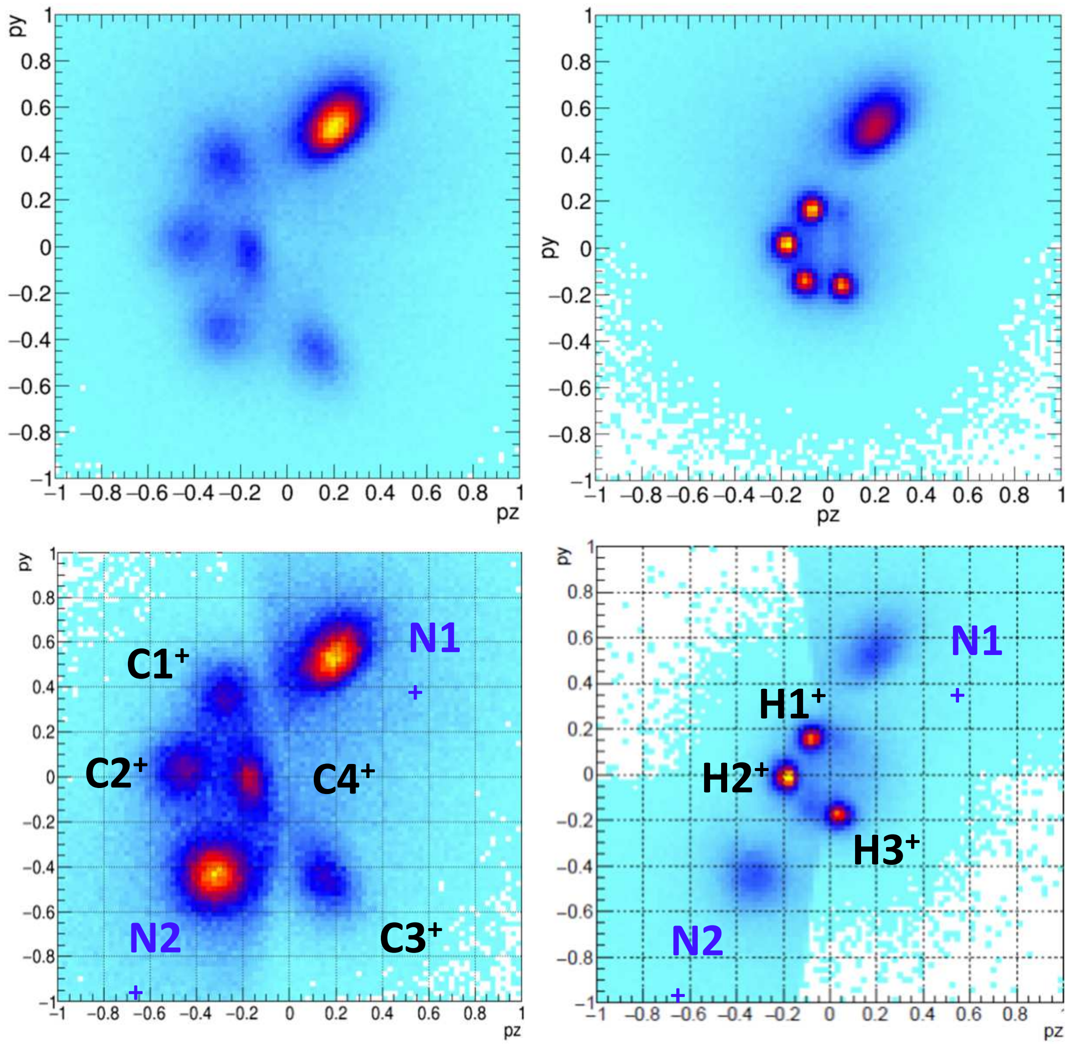
Complete Coulomb Explosion Imaging

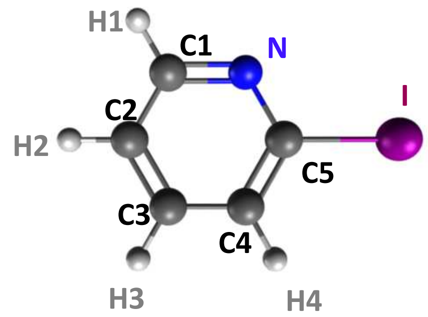


- molecular structure is very well reflected in measured momenta
- no evidence of deformation or rotation before breakup
- 3-fold ion coincidences are sufficient to image the entire molecule
- no need to record all 11 ions in coincidence, as long as all atoms are charged up fast enough

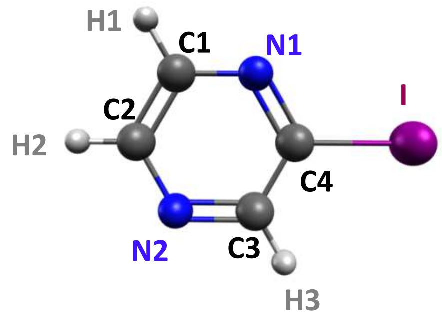
Boll et al., Nature Physics 18, 423 (2022)

Molecular fingerprints





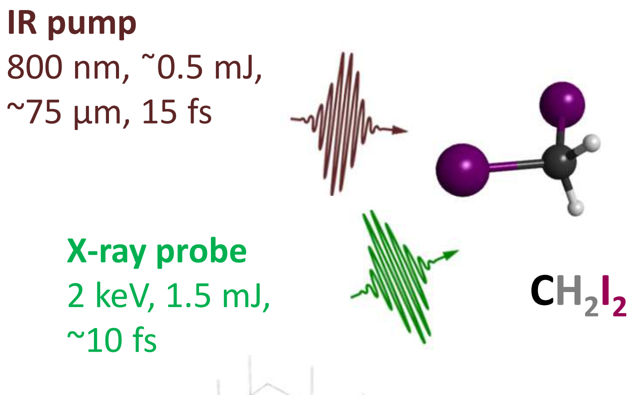
molecular structure can be clearly identified!



these can be the first frames of a molecular movie!

Boll et al., Nature Physics 18, 423 (2022)

Coulomb explosion imaging goes time-resolved!



unpublished

- one of the main objectives of the technique: image molecular dynamics
- first pump-probe experiment: IR-induced vibration and dissociation in CH_2I_2
- here: full 5-fold coincidences, 3D imaging
- direct link between bond distances/angles and ion momenta?
  → modelling!

X. Li et al., to be submitted

Coulomb explosion imaging goes time-resolved!

unpublished

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unpublished

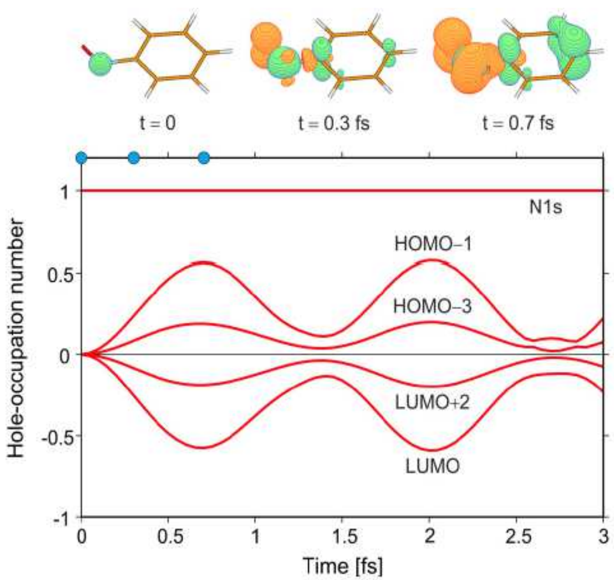
X. Li et al., to be submitted

Outlook: Time-resolved (attosecond) Spectroscopy

Charge migration in the valence opon core ionization

Molecular electron dynamics decoupled from nuclear motion

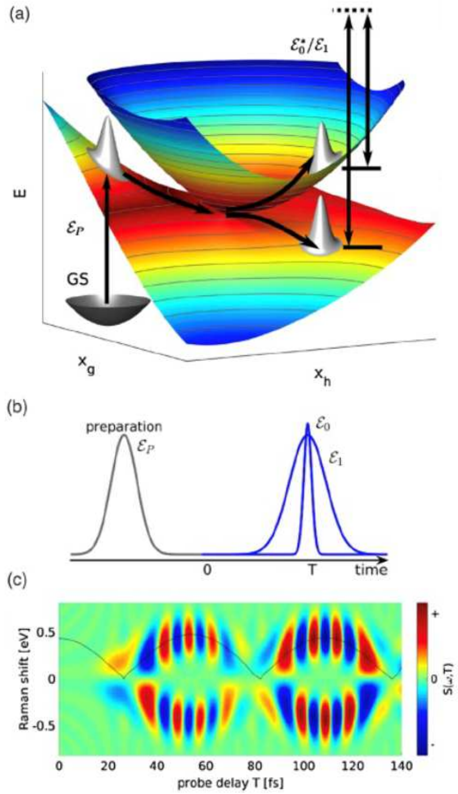
N $1s^{-1}$ in nitrobenzene



A. Kuleff et al., PRL **117**, 093002 (2016)

Time-resolved Impulsive X-ray Raman Scattering

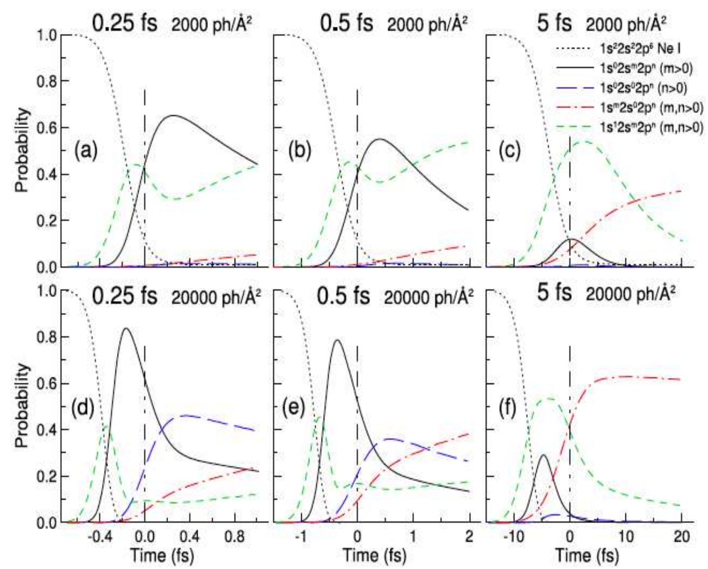
Conical Intersections in photochemical processes



M. Kowalewski,…,S. Mukhamel, PRL **115**, 193003 (2015)

Nonlinear Attosecond Spectroscopy

Double-core hole formation upon photoionization in Ne



S. Serkez…. A. Grum-Grzhimailo, J. Opt. **20** (2018)

Time-resolved (attosecond) Spectroscopy: New developments at SQS

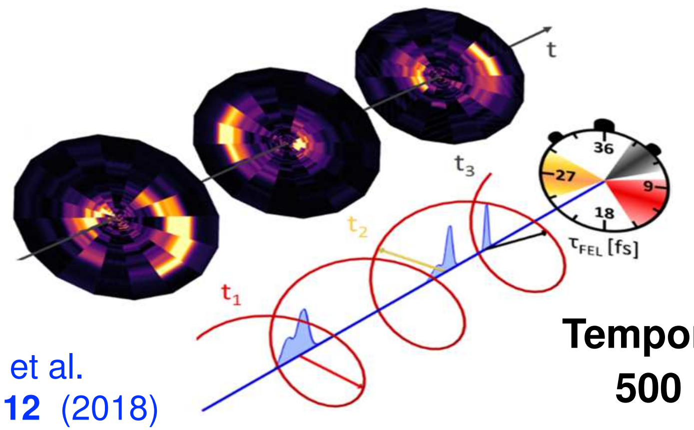
Single-spike lasing at SASE3

Demonstration at 16.5 GeV with dedicated (strong compression combined with electron beam orbit dispersion) operation mode

unpublished

Angular streaking for as-diagnostics

Circularly polarized field → angular distribution



λ = 10.6 μm (35 fs)

Temporal Resolution 500 as (→ 50as)

Hartmann et al.
Nat Phot. **12** (2018)

unpublished

U Dortmund, DESY, U Kassel, SQS, FEL R&D

SQS group, 16.1.2024

Thank you for your attention !!!

European XFEL