Breakdown of frustrated absorption in x-ray sequential multiphoton ionization

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Virtual CFEL-DESY Theory Seminar
April 1, 2020

Collaboration

CFEL-DESY Theory Division

Robin Santra

European XFEL SQS

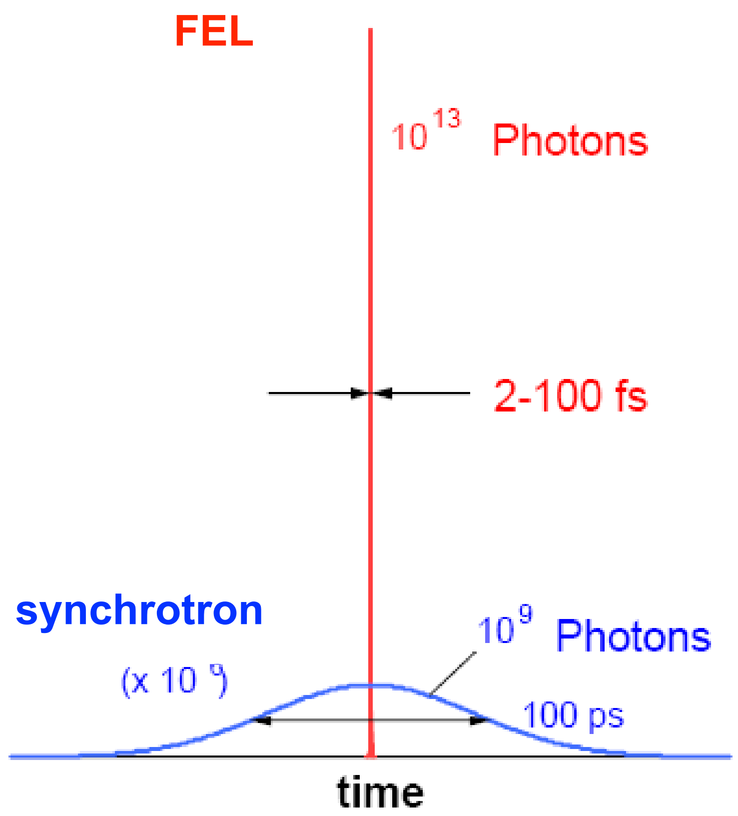
Rebecca Boll

The main part of this presentation has been published in *Phys. Rev. Research.*

Introduction

XFEL: X-ray free-electron laser

- revolutionizing ultrafast science
- photon energy: ~10 keV
  number of photons: ~10^{13}
  focal size: ~10 μm × 10 μm
  pulse duration: ~10 fs
  ➔ peak intensity ~10^{18} W/cm^2
- novel light-matter interaction:
  sequential multiphoton multiple ionization dynamics
- frustrated absorption:
  higher intensity, less ionization

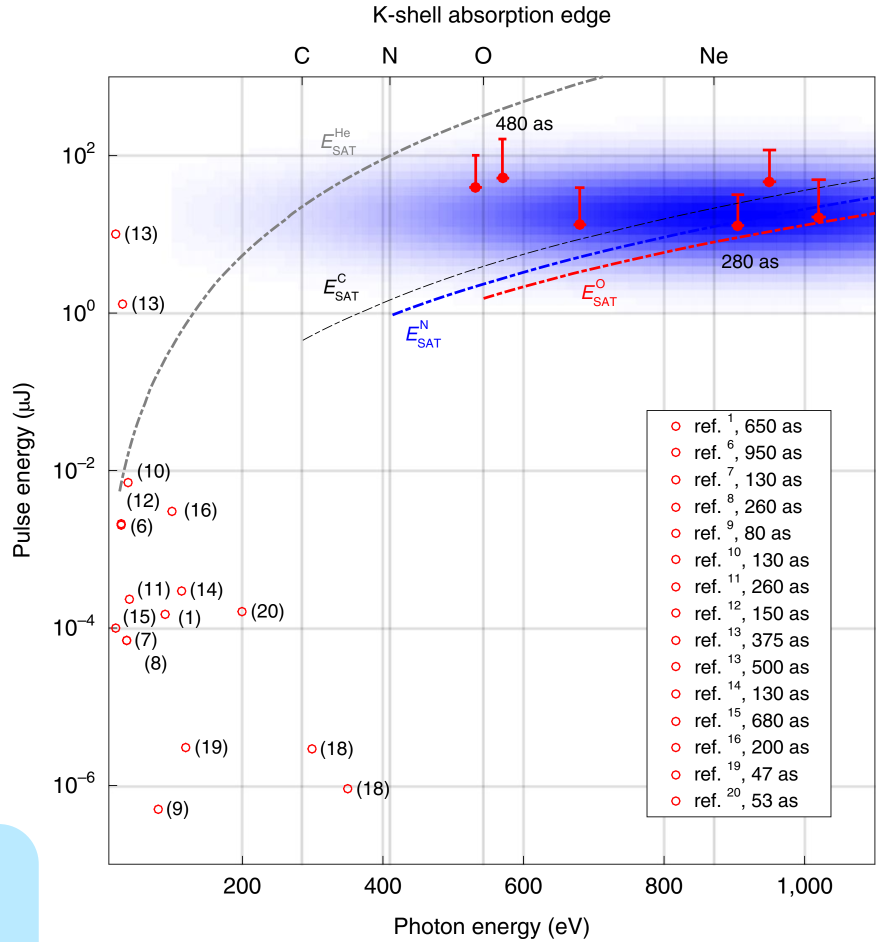


Schneider, *Rev. Accl. Sci. Tech.* **3**, 13 (2010).

Towards terawatt-attosecond XFEL pulses

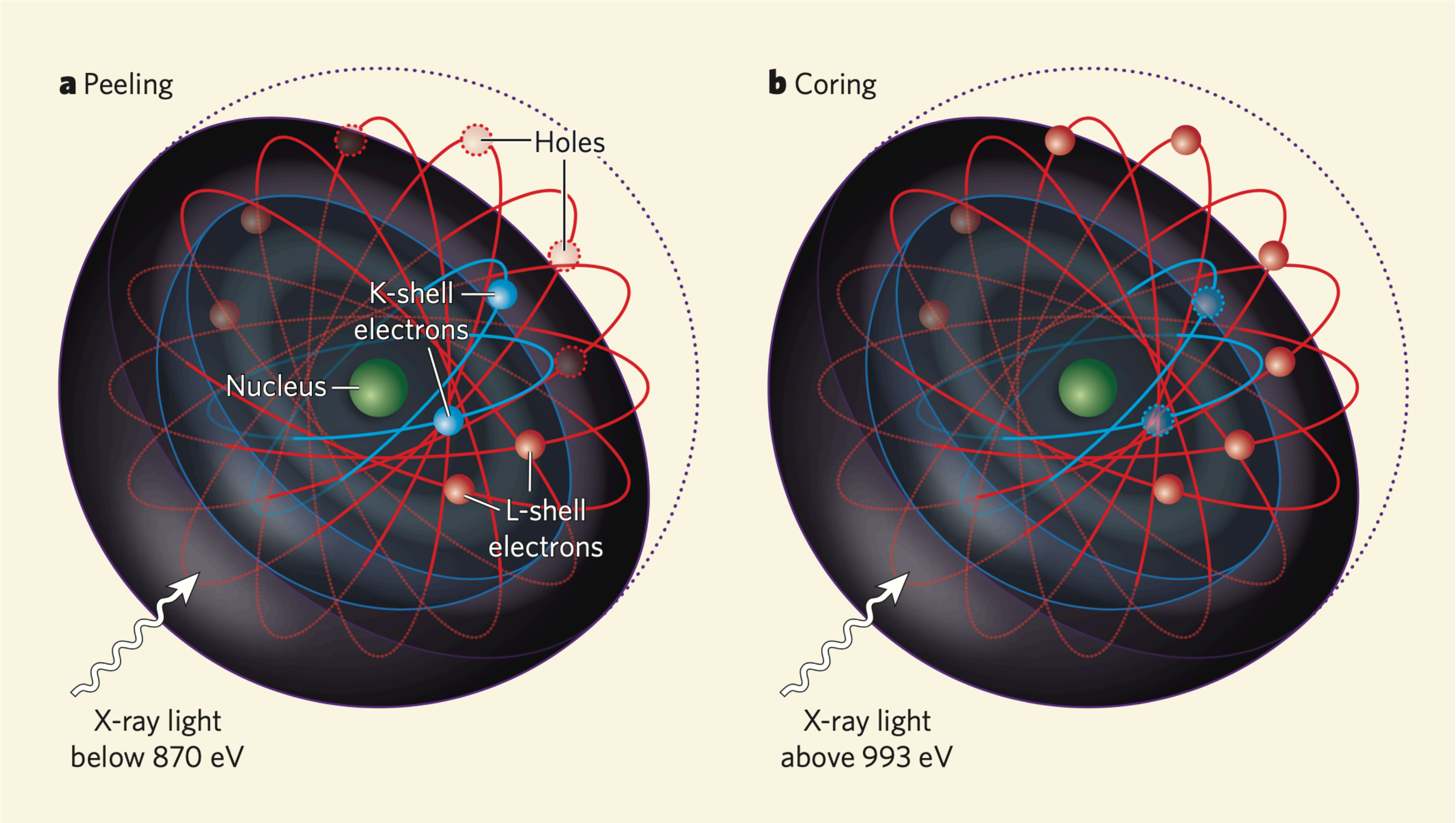
- XLEAP at LCLS: generates tunable isolated attosecond X-ray pulses with gigawatt peak power
- ~10^{12} W tightly focused on 1 μm × 1 μm ➔ 10^{20} W/cm^2
- Why terawatt-attosecond pulses?
  - *ultrashort* pulse: probing electronic dynamics
  - *ultraintense* pulse: single particle imaging

How does matter interact with *ultraintense* and *ultrafast* pulses?



Duris *et al.*, *Nat. Photon.* **14**, 30 (2020).

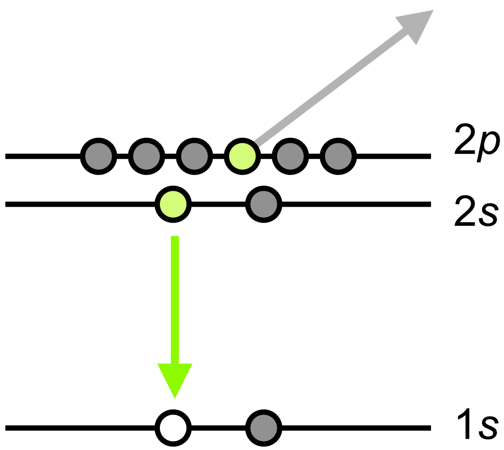
X-ray ionization: peeling or coring



“X-ray laser peels and cores atoms,” Wark, *Nature* **466**, 35 (2010).

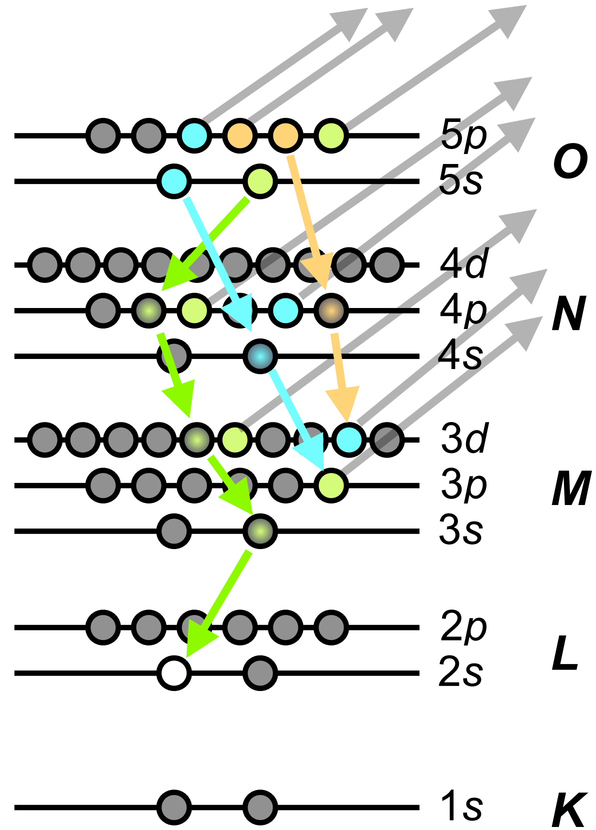
Relaxation of inner-shell vacancy

$Ne^{+}(1s^{-1})$



Auger decay (or fluorescence) after core ionization

Xe^{+} $(2s^{-1})$



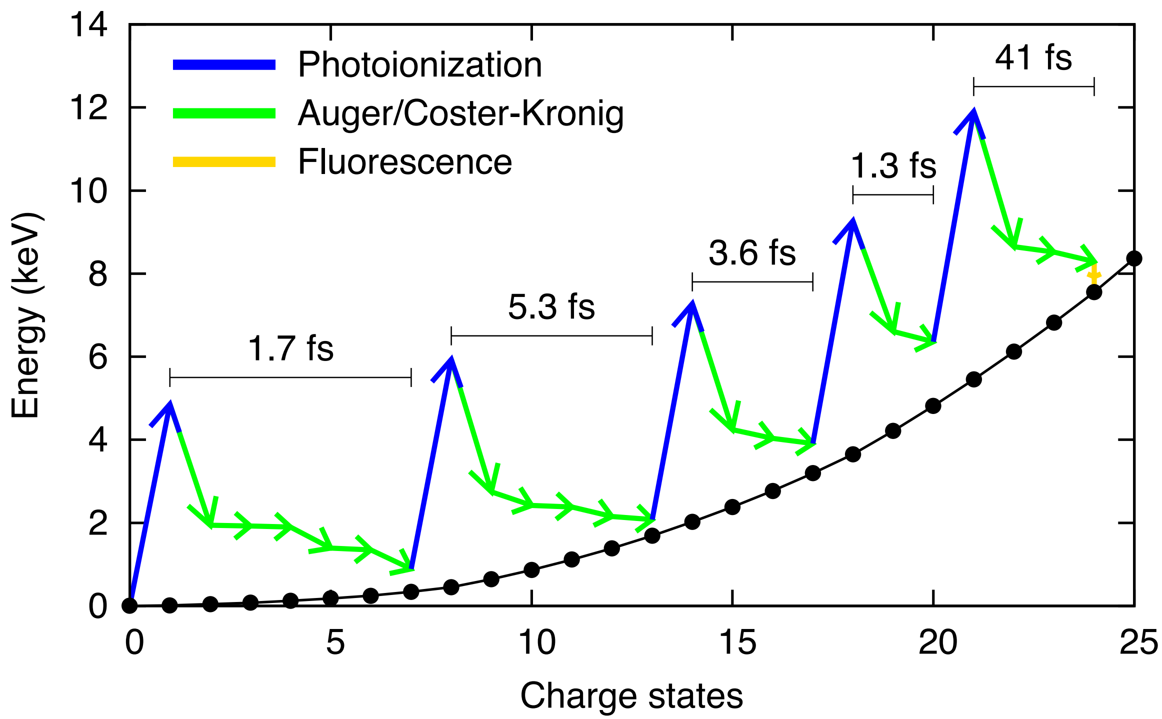
Auger cascade makes complex ionization dynamics

Sequential multiphoton multiple ionization

- At high x-ray intensity, the system can absorb many photons
- Direct multiphoton absorption: negligible if one-photon abs. is available

Doumy *et al.*, *Phys. Rev. Lett.* **106**, 083002 (2011).

- A sequence of one-photon absorption and accompanying relaxations

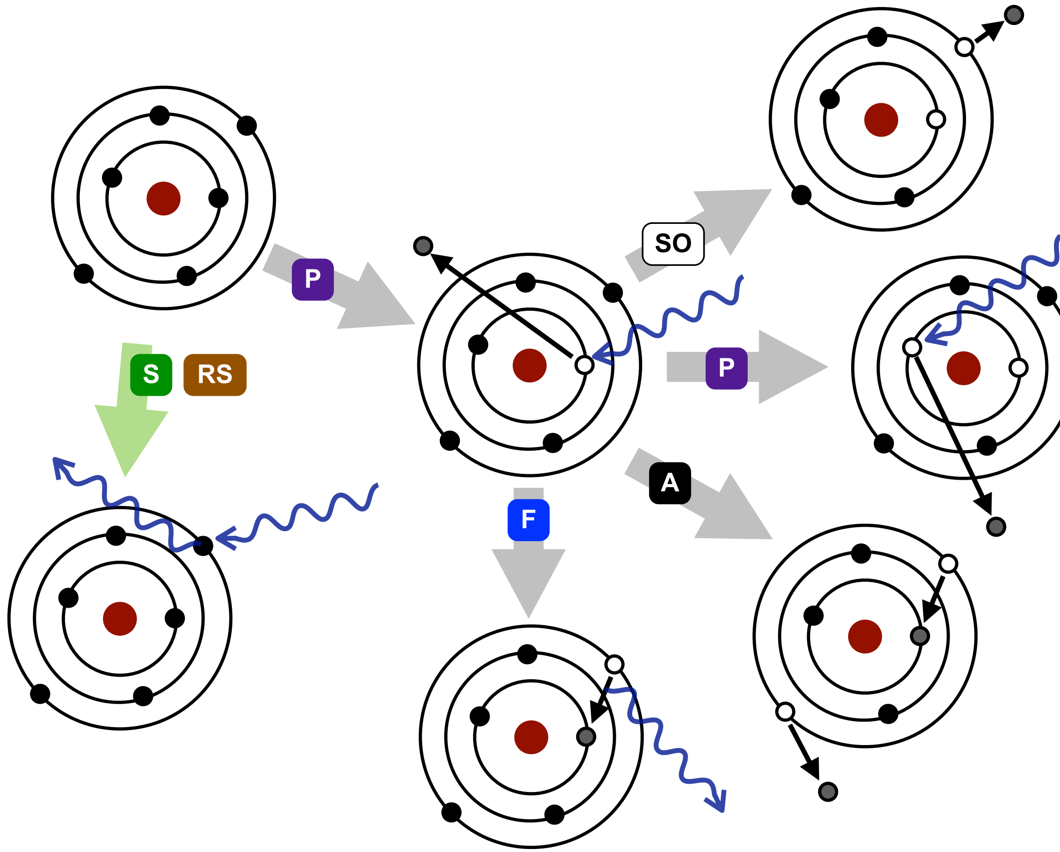


Xe@5.5 keV

Fukuzawa *et al.*, *Phys. Rev. Lett.* **110**, 173005 (2013).

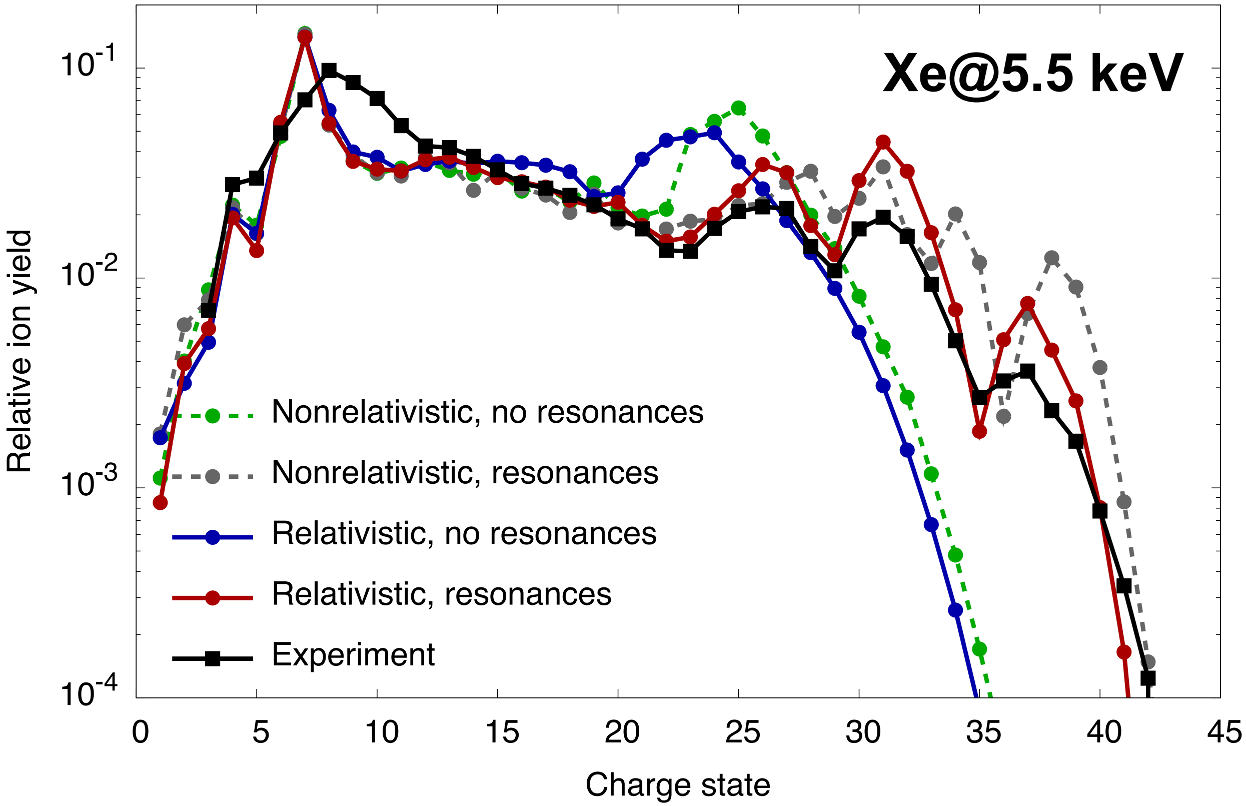
XATOM: all about x-ray atomic physics

- X-ray-induced atomic processes calculated for any given element and configuration
- Ionization dynamics solved by a rate-equation approach
- Sequential ionization model has been tested by a series of atomic XFEL experiments



Son, Young & Santra, *Phys. Rev. A* **83**, 033402 (2011).
Jurek, Son, Ziaja & Santra, *J. Appl. Cryst.* **49**, 1048 (2016).
Download executables: http://www.desy.de/~xraypac

Comparison with experimental CSD



LCLS experiment

Daniel Rolles at KSU

Artem Rudenko at KSU

Benedikt Rudek at PTB

XATOM development

Koudai Toyota

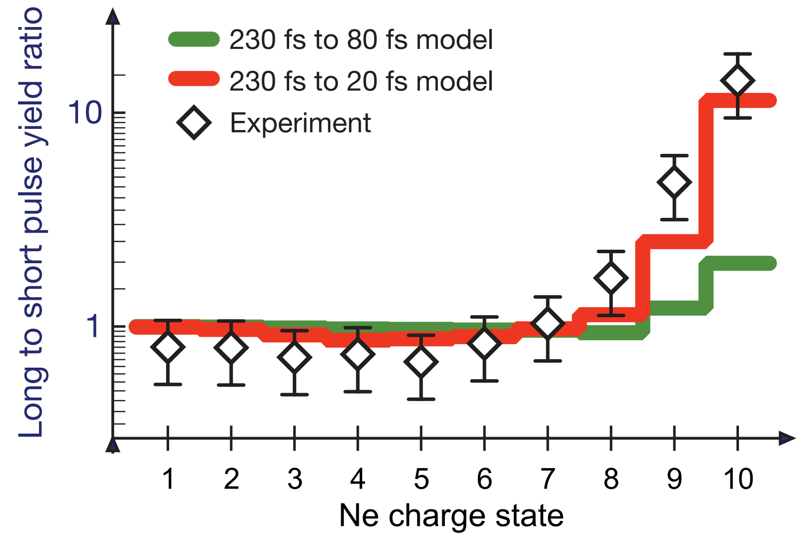
Rudek *et al.*, *Nat. Commun.* **9**, 4200 (2018).

- CSD (charge-state distribution): outcome of ionization dynamics
- Sequential multiphoton multiple ionization model verified
- Structured CSD: interplay of resonance and relativistic effects

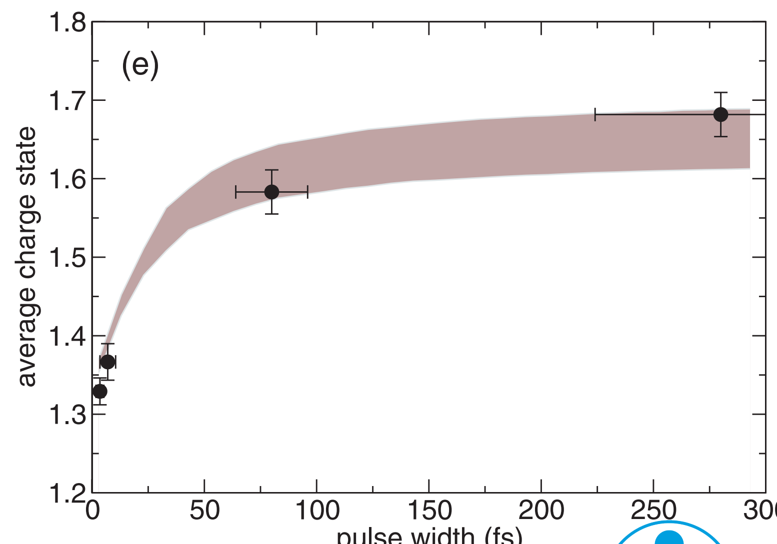
Frustrated absorption

Pioneering works for Ne and N_2

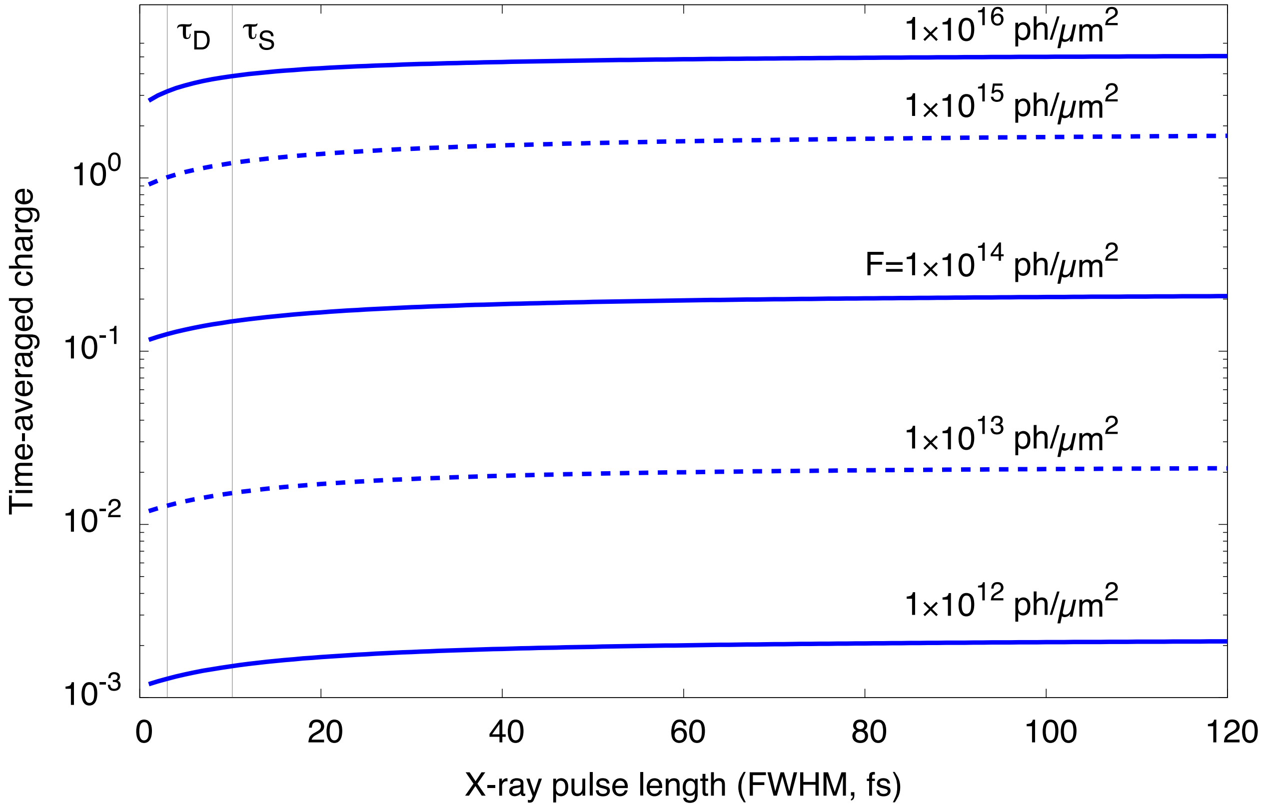
- Ne atom
  - Young *et al.*, *Nature* **466**, 56 (2010).
  - long to short pulse yield ratio > 1: longer pulse gives higher yields for high charges
  - *intensity-induced x-ray transparency*



- N_2 molecule
  - Hoener *et al.*, *PRL* **104**, 253002 (2010).
  - mean charge reduced for shorter pulses
  - *frustrated absorption*



Frustrated absorption: carbon

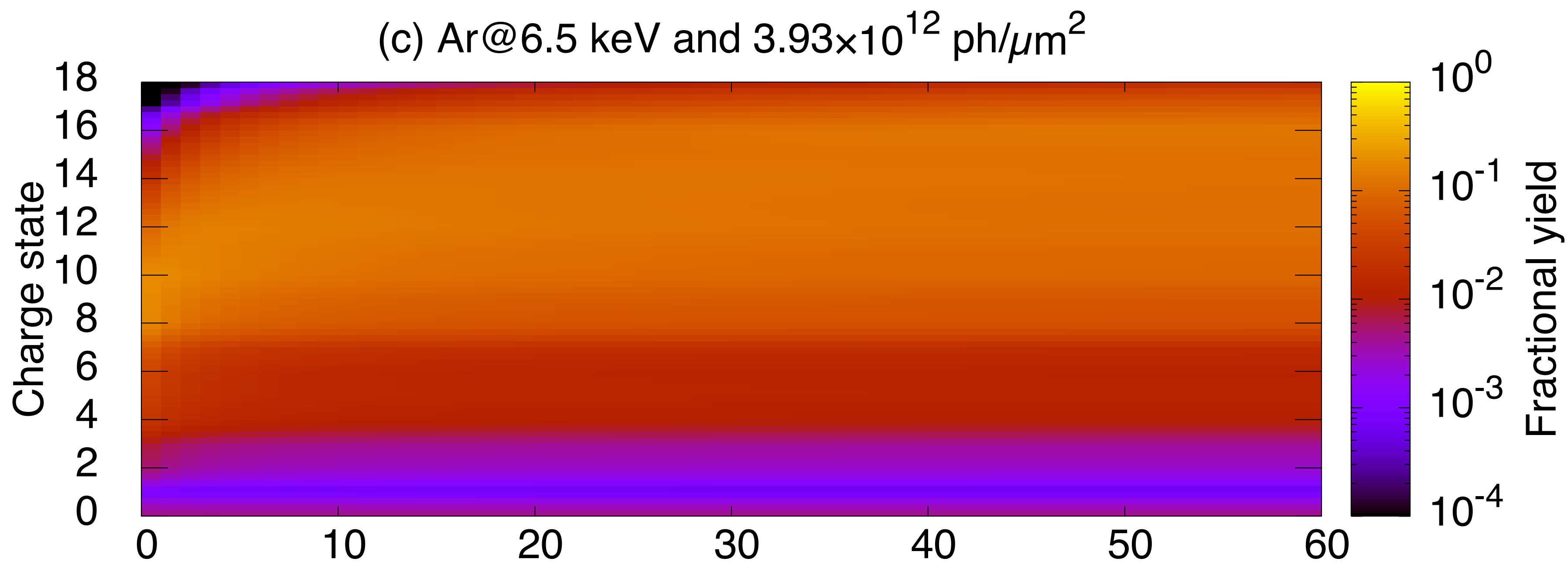


C@12 keV

Son, Young & Santra, *Phys. Rev. A* **83**, 033402 (2011).

- For pulse duration > SCH lifetime, almost no dependence
- For pulse duration < SCH/DCH lifetime, average charge is reduced

Frustrated absorption: argon



Toyota *et al.*, *J. Synchrotron Radiat.* **26**, 1017 (2019).

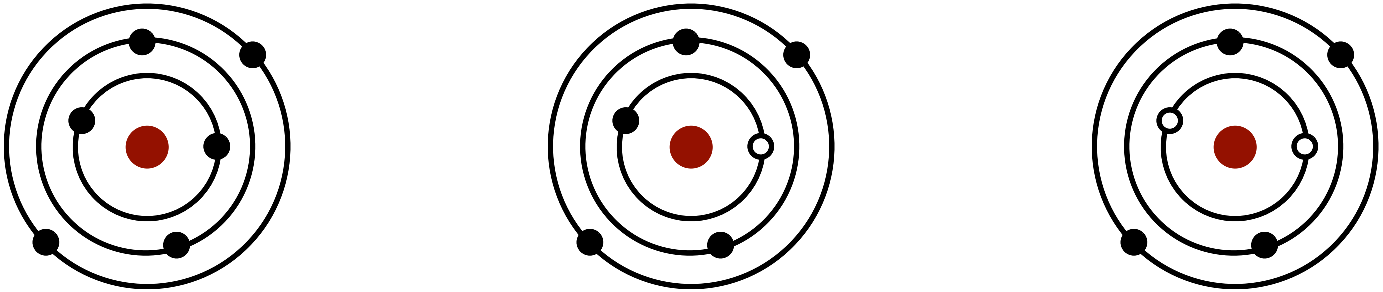
Ar core-hole lifetime:
$Ar^{+}(1s^{-1})$ ~ 1 fs
Ar^{15+} ($1s^{1}\ 2s^{2}$) ~ 15 fs

- CSD (charge-state distribution) as a function of pulse duration
- No dependence except pulse duration shorter than core-hole lifetime

Implication of frustrated absorption

- Mechanism of frustrated absorption
  - hollow-atom formation: another photoionization before core-hole relaxation ➔ cross section reduced ➔ less ionization
  - shorter pulse, less radiation damage ➔ advantage for molecular imaging

C@12 keV

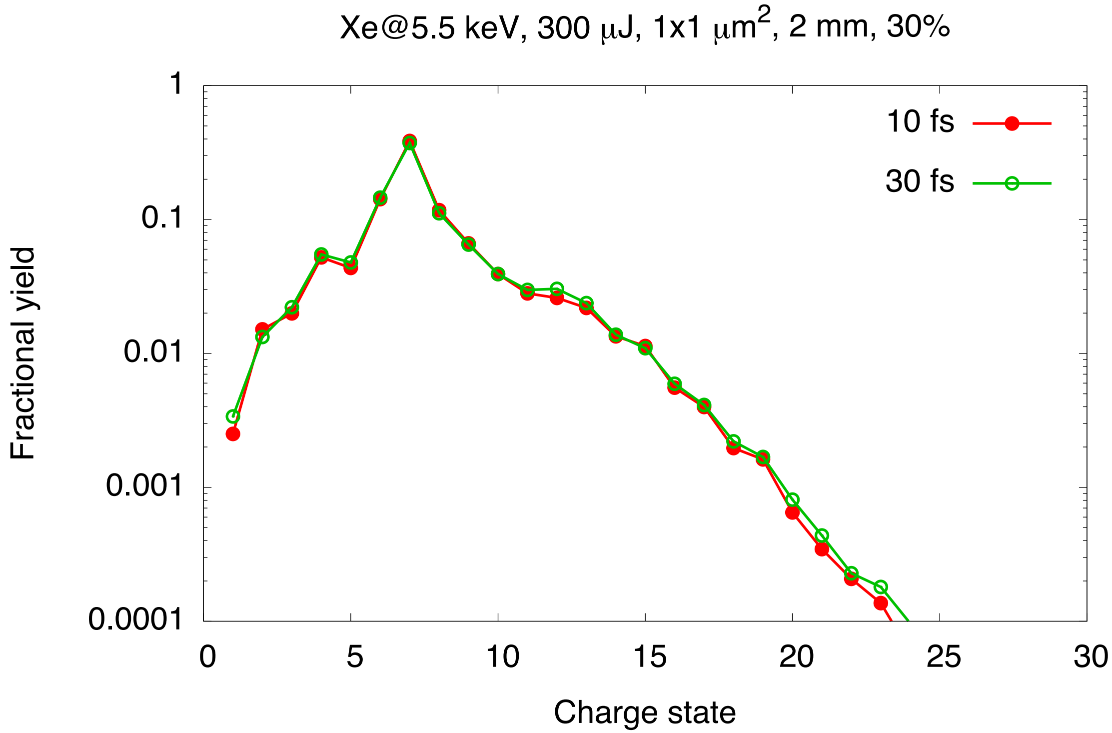


neutral	single-core-hole	double-core-hole
σ_{abs} = 23 barns	13 barns	2 barns
σ_{sc}/σ_{abs} = 0.057	0.075	0.305

For scattering (sc), resolution=1.7 Å is assumed.

Frustrated absorption for heavy atoms

- Frustrated absorption has been verified for light atoms
- What happens for heavy atoms? Core-hole lifetime is even shorter
- Previous studies: almost no dependence of Xe on pulse duration



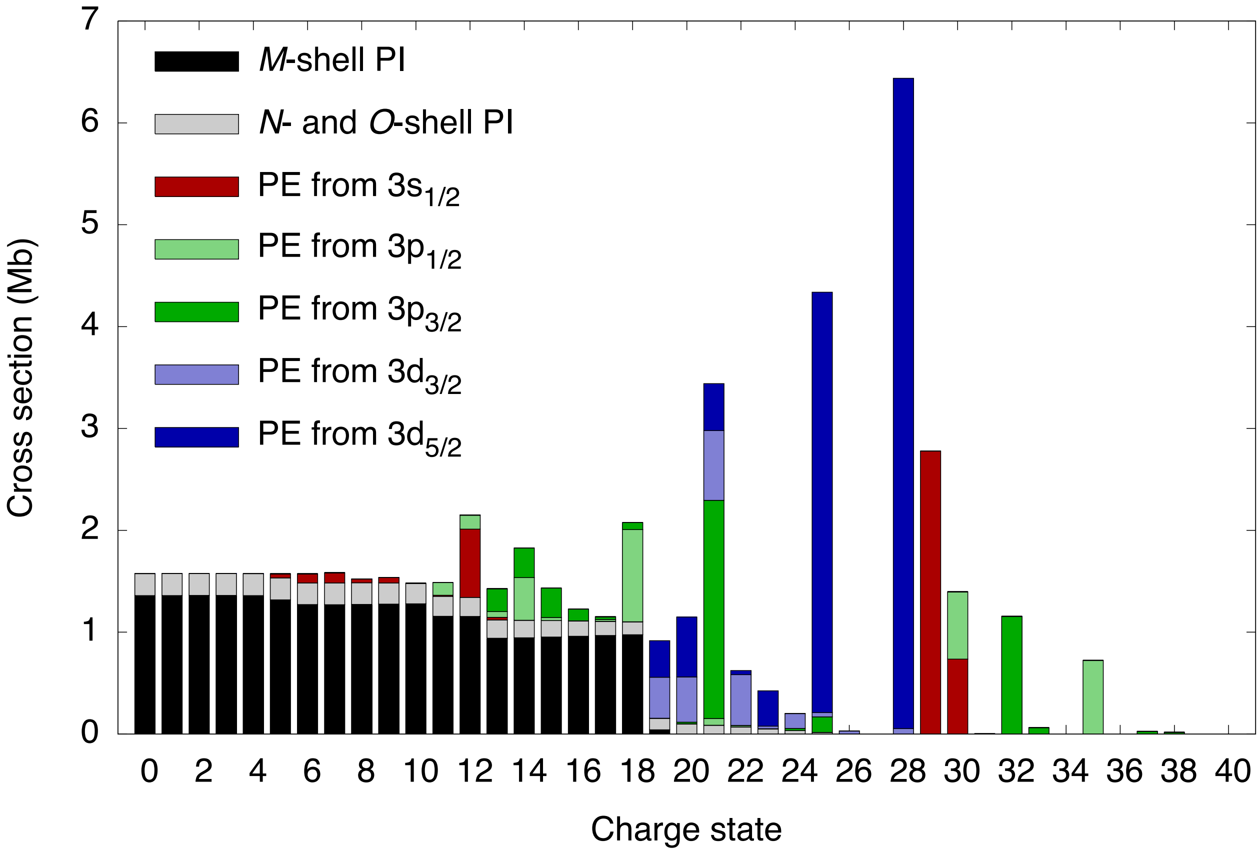
Test calculation for SACLA Xe@5.5 keV

Breakdown of frustrated absorption: a study of Xe@1200 eV

Son, Boll & Santra,

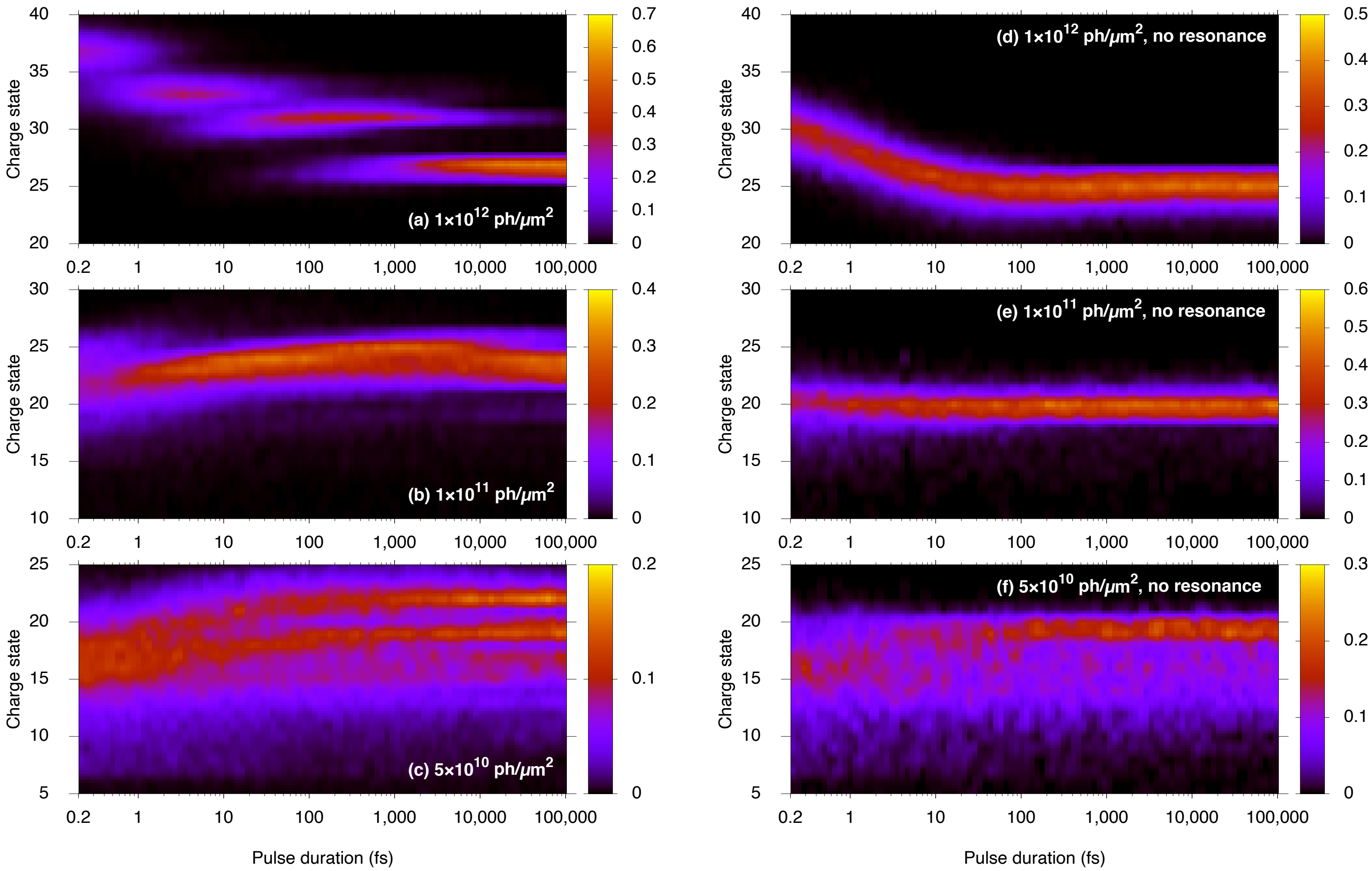
Phys. Rev. Res. **2**, 023053 (2020)

Photoabs. cross section of Xe at 1200 eV

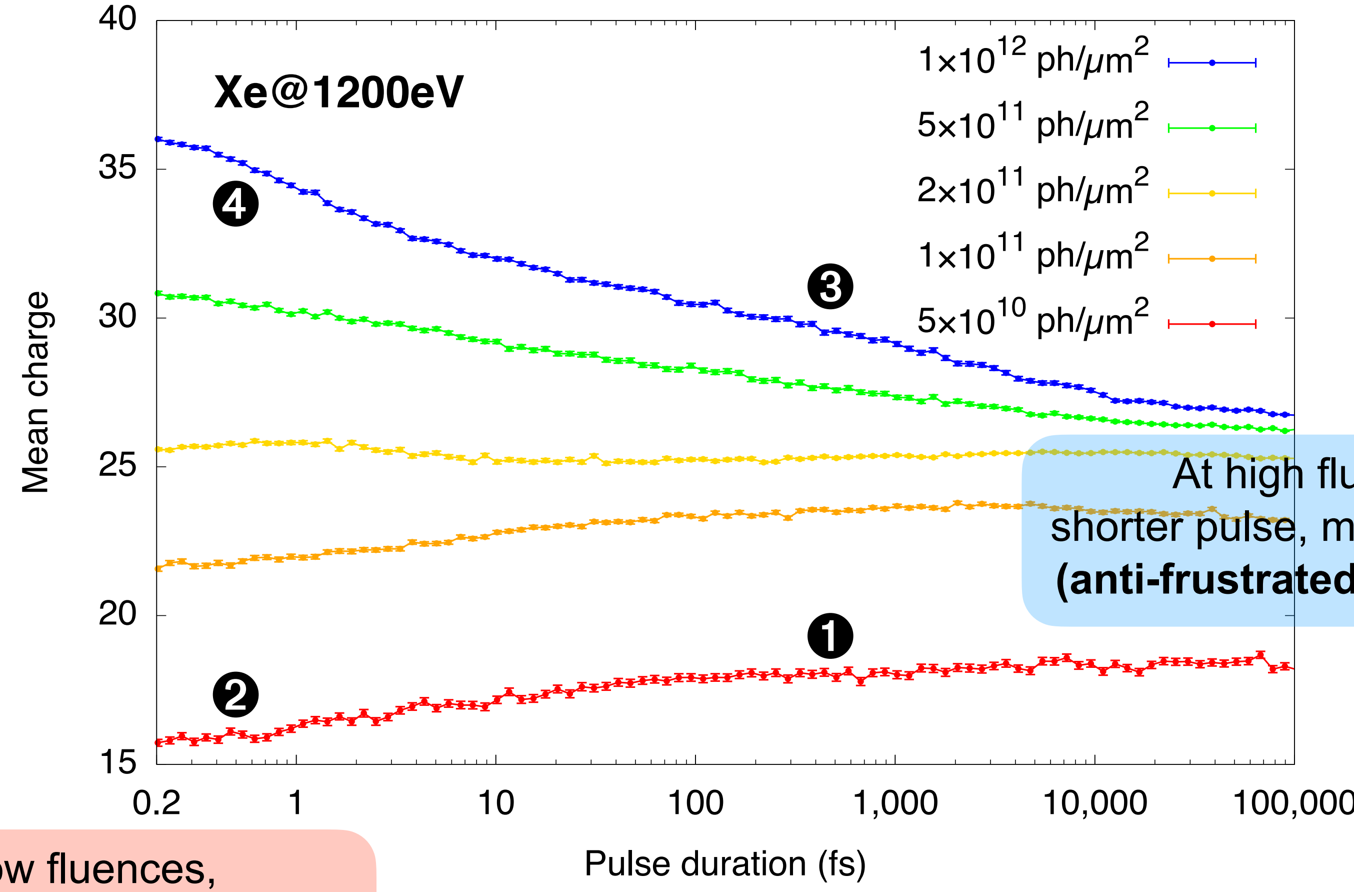


- PI: photoionization (bound→continuum)
- PE: resonant photoexcitation (bound→bound, $3 \leq n \leq 30$ & $0 \leq l \leq 7$ in calculation)
- M-shell ionization / excitation is dominant

Pulse-duration dependence of Xe CSD



Breakdown of frustrated absorption

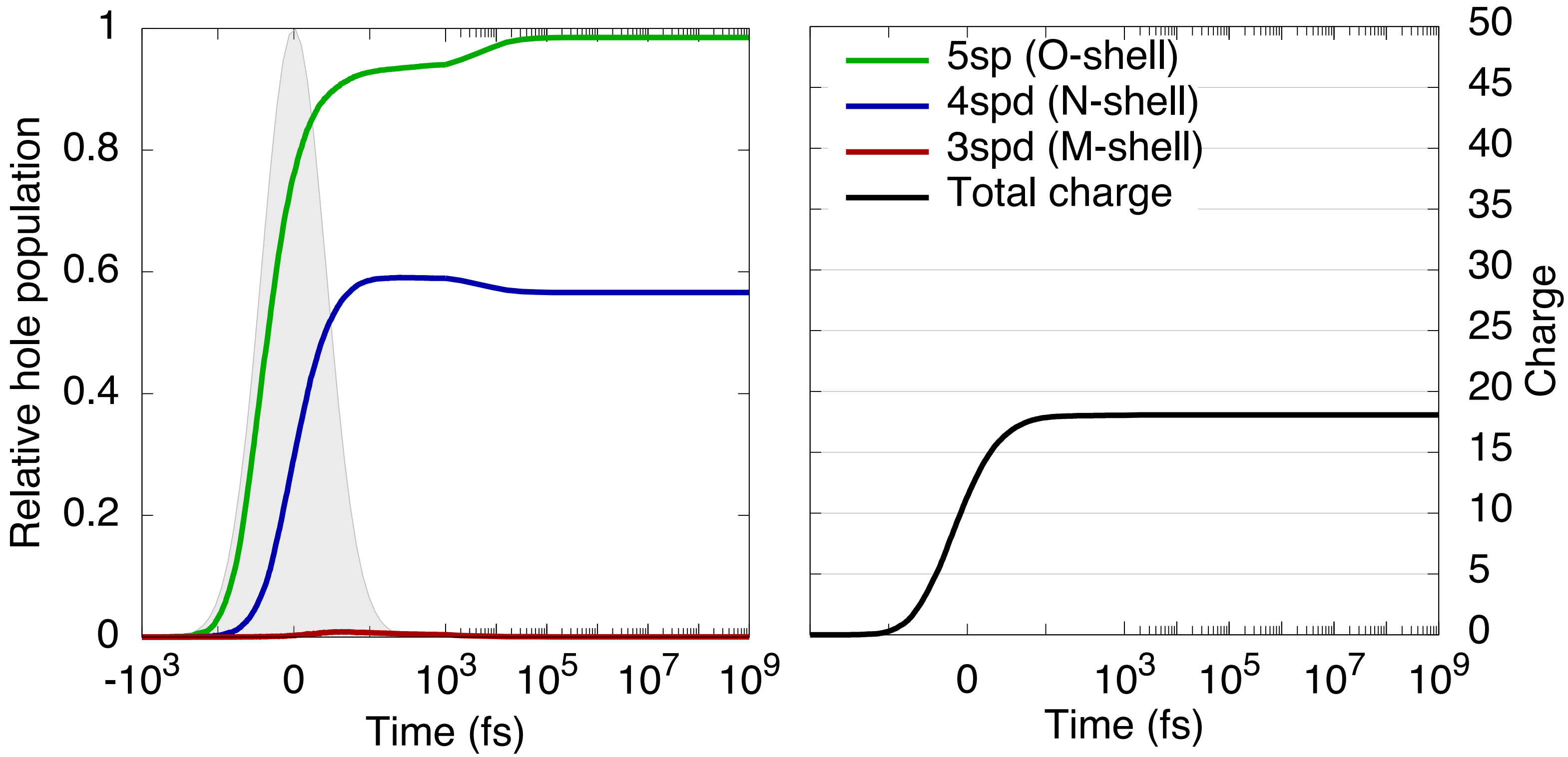


At high fluences,
shorter pulse, more ionization
(anti-frustrated absorption)

At low fluences,
shorter pulse, less ionization
(frustrated absorption)

Hole population dynamics: *low F, long τ*

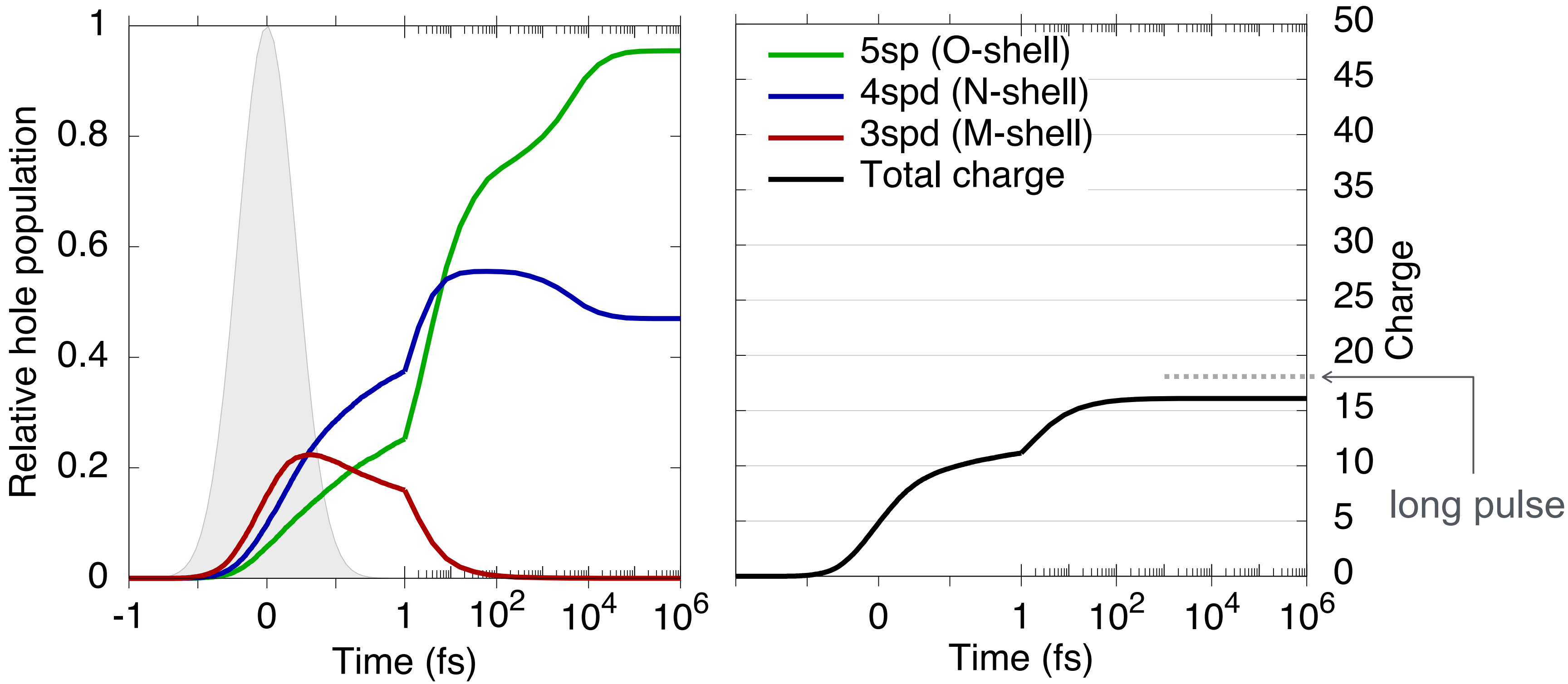
❶ *Low* fluence, *long* pulse: 5×10^{10} ph/μm^2, 500 fs



- X-ray ionization dominated by M-shell ionization (*coring*)
- No hollow-atom (multiple-core-hole) formation is observed

Hole population dynamics: *low F, short τ*

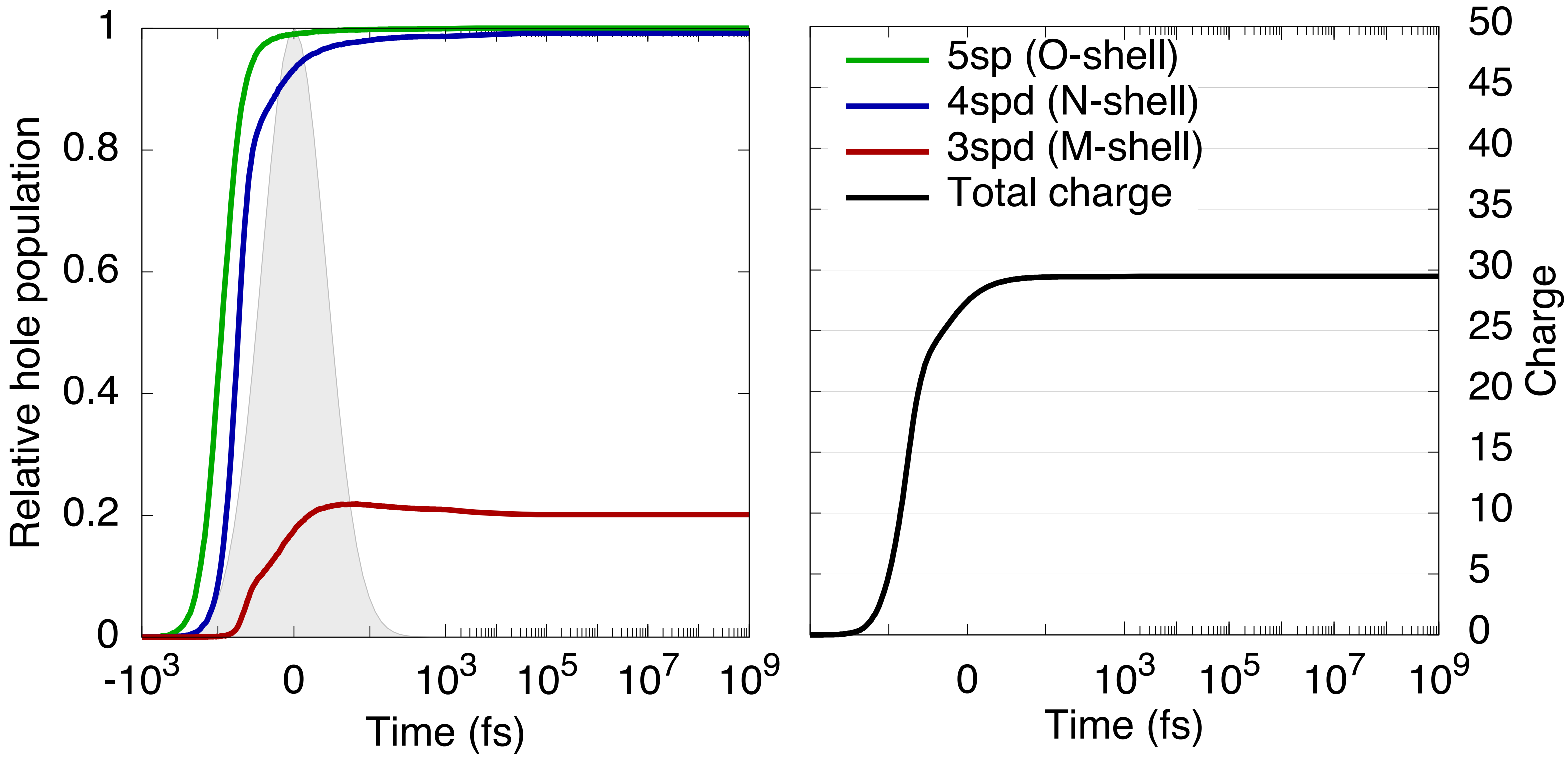
❷ *Low* fluence, *short* pulse: 5×10^{10} ph/μm², 0.5 fs



- Shorter pulse facilitates multiple-core-hole formation
- Reduction of cross section & Auger decays ➔ mean charge decreases

Hole population dynamics: *high F, long τ*

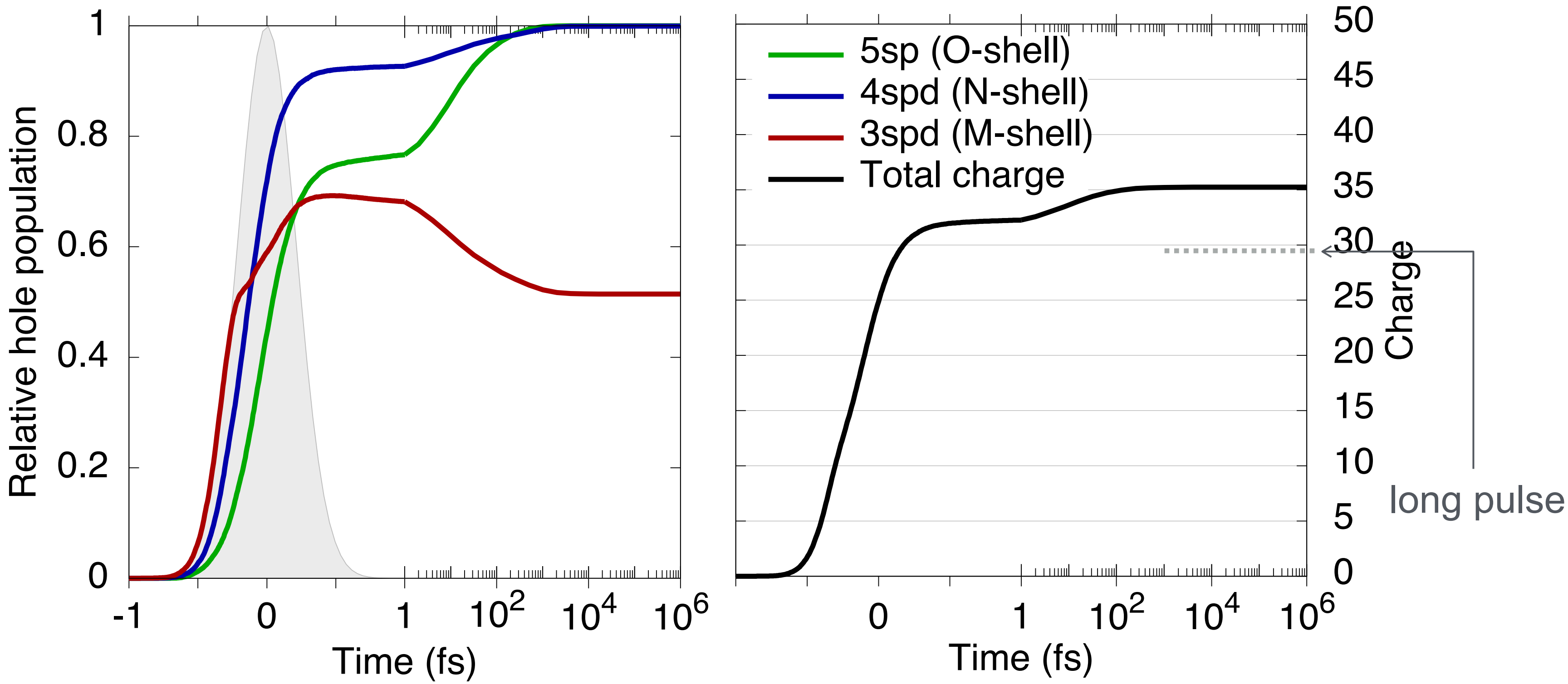
❸ *High* fluence, *long* pulse: $1{\times}10^{12}$ ph/μm², 500 fs



- At high fluence, M-shells are ionized after losing N- & O-shell electrons
- Direct M-shell ionization stops at +19; resonance makes higher charges

Hole population dynamics: *high F, short τ*

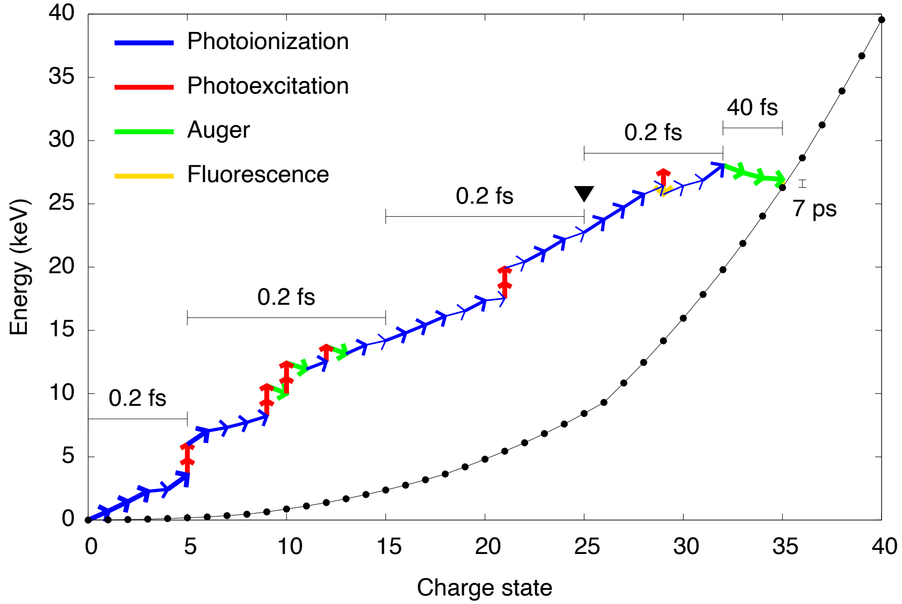
❹ *High* fluence, *short* pulse: 1×10^{12} ph/μm², 0.5 fs



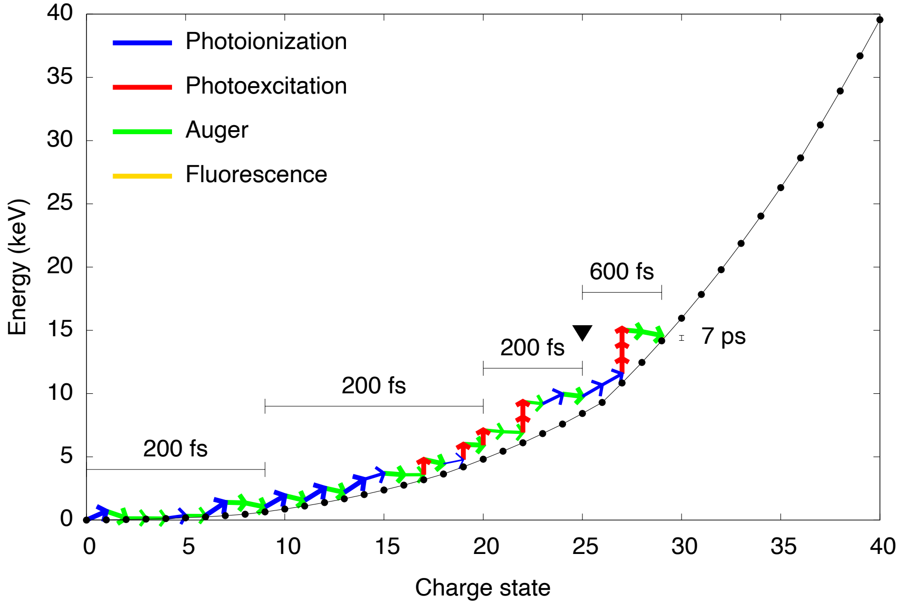
- *Massively hollow atom* (MHA): ~75% of M-shell electrons are ionized
- Early formation of MHA enables to go beyond the ionization limit

Ionization mechanism: short vs. long pulses

❹ 1×10¹² ph/μm², 0.5 fs (FWHM)



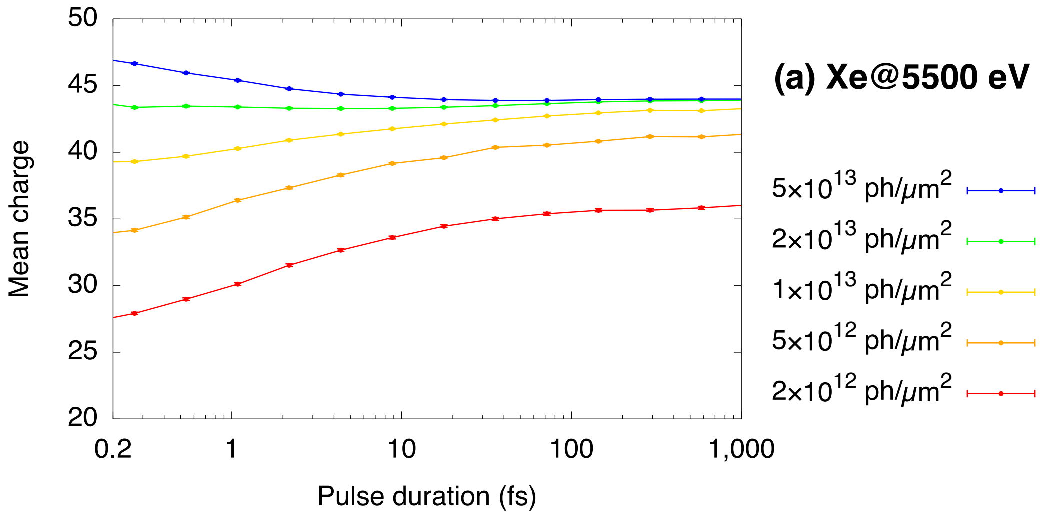
❸ 1×10¹² ph/μm², 500 fs (FWHM)

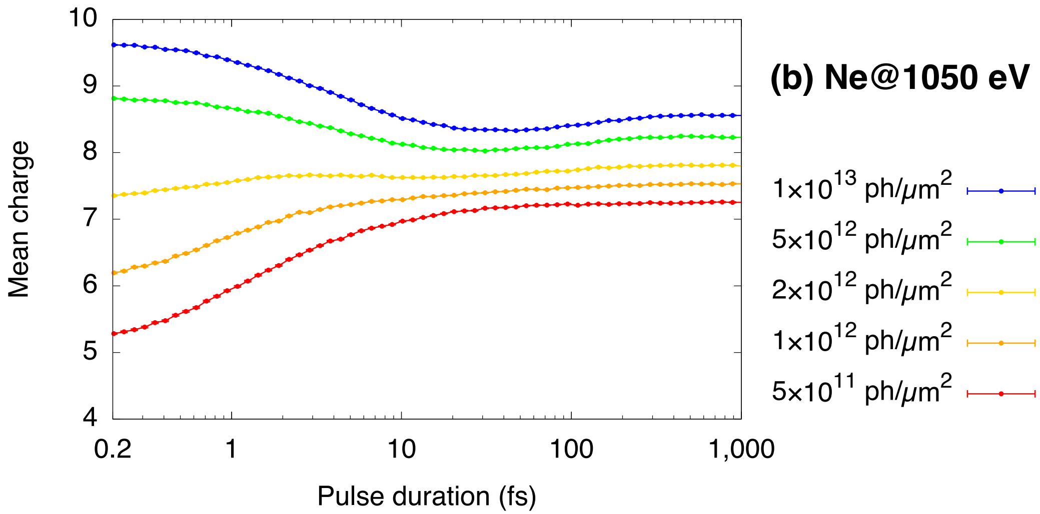


- Get a higher final charge via highly excited (~10 keV) states
- *Quasi-nonsequential absorption* in the X-ray regime

- Close to the ionization pathway via ground states

Pulse-duration dependence of Xe and Ne





- Heavy atoms at hard X-ray
  - FA at low fluences
  - anti-FA at high fluences

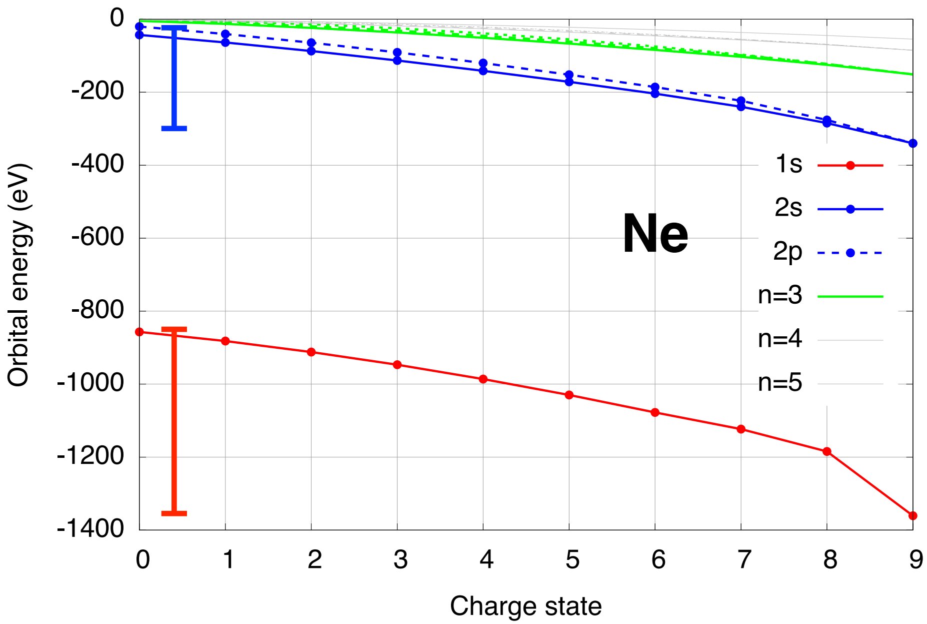
c.f.) Xe experiment at LCLS CXI: peak fluence ~2.5×10^{12} ph/μm^2

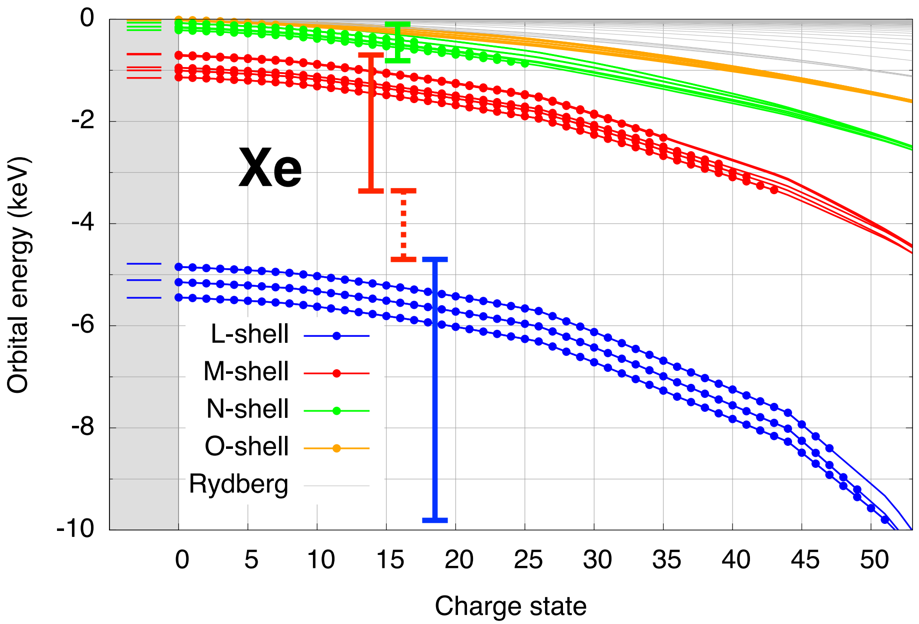
- Light atoms at soft X-ray
  - FA at low fluences
  - anti-FA at high fluences

- Light atoms at hard X-ray?

When anti-frustrated absorption occurs?

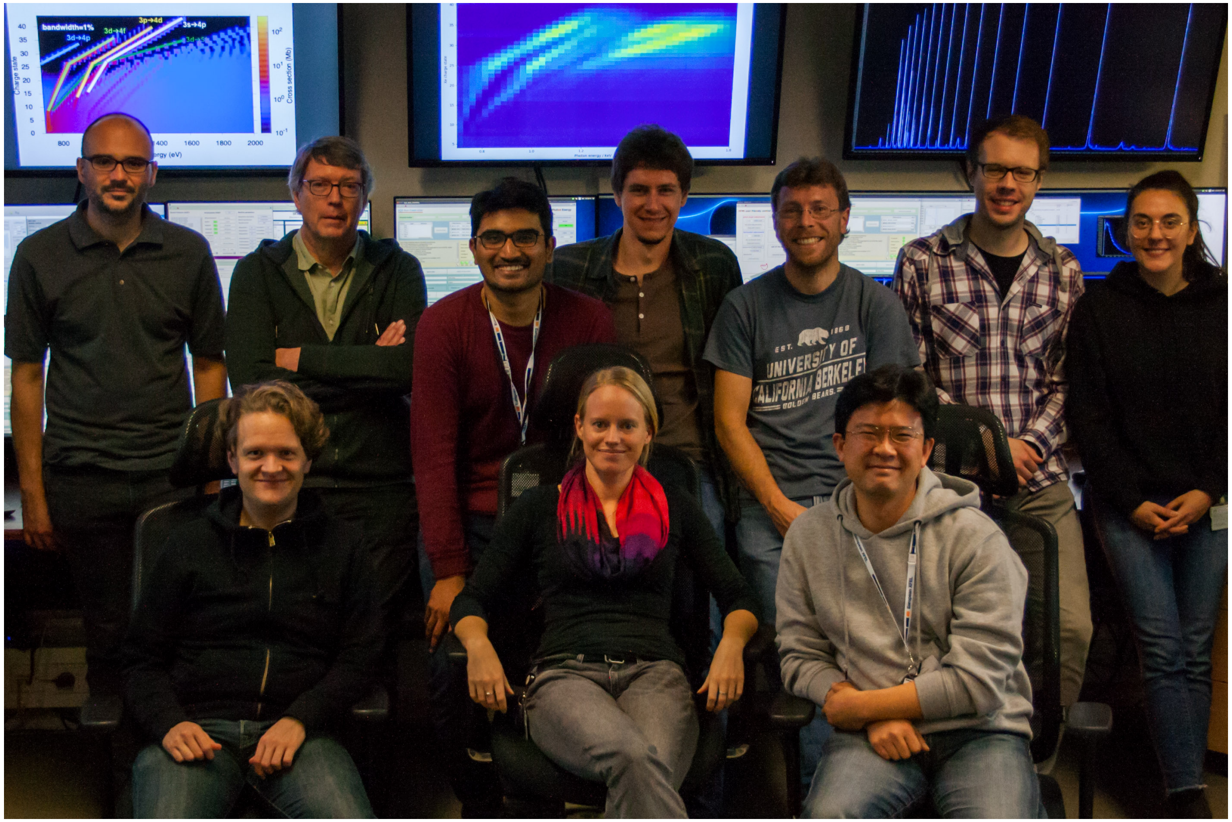
- As pulse duration becomes shorter, X-ray multiphoton ionization can be suppressed or enhanced, depending on fluence and photon energy
- Breakdown of frustrated absorption when fluence is sufficiently high
- No anti-frustrated absorption when photon *E* exceeds max ionization *E*





Wavelength-dependence of soft x-ray ionization (preliminary results)

European XFEL SQS beamtime

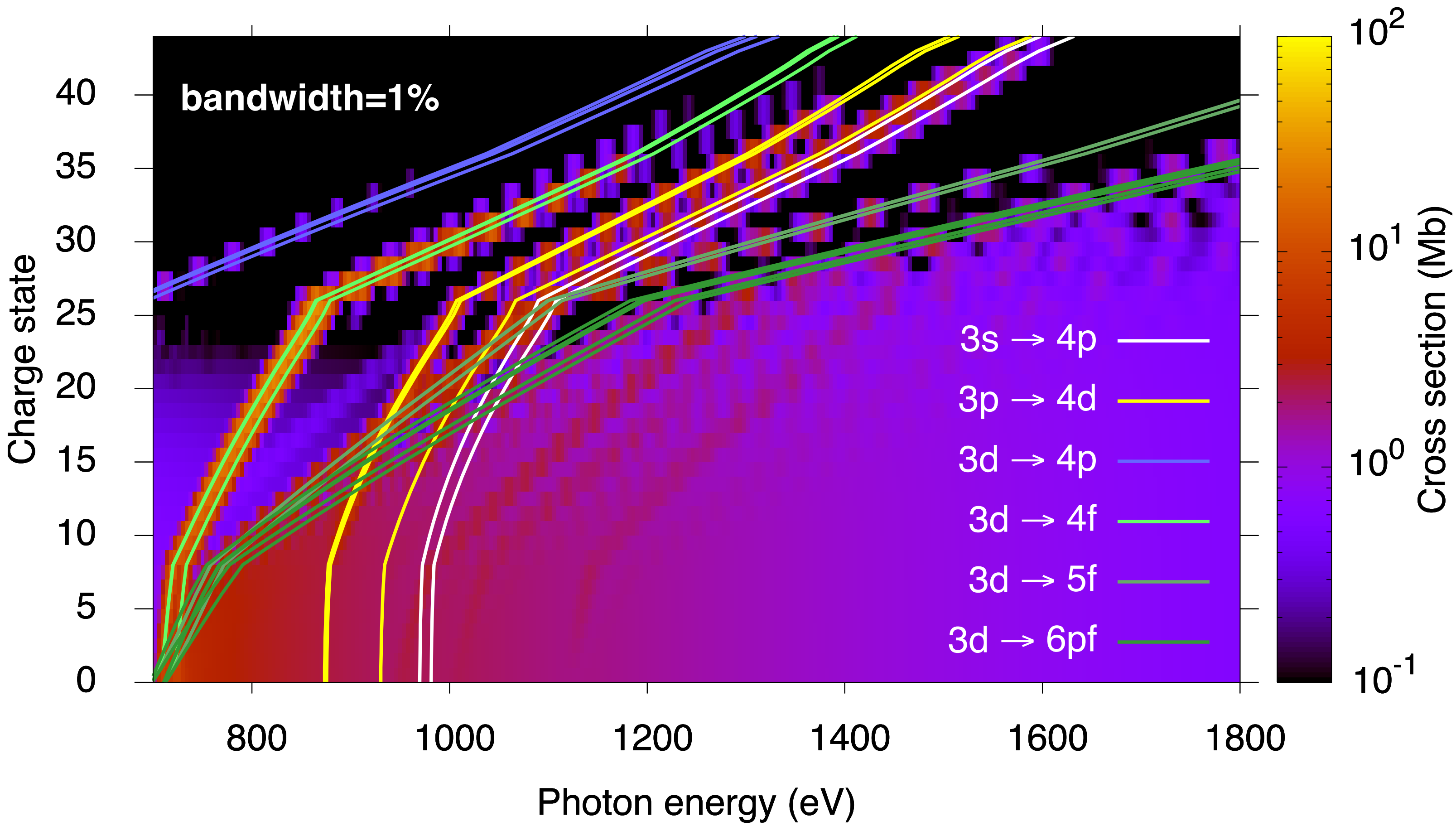


Proposal 2310:
October 16–23, 2019

Rebecca Boll, Thomas Baumann, Alberto De Fanis, Valerija Music, Daniel Rivas, Aljoscha Rörig, Philipp Schmidt, Sergey Usenko, Michael Meyer (EuXFEL SQS), Joakim Laksman (EuXFEL XPD), Svitozar Serkez (EuXFEL SPF), Benjamin Erk (DESY), Shashank Pathak, Daniel Rolles (KSU) — to be completed

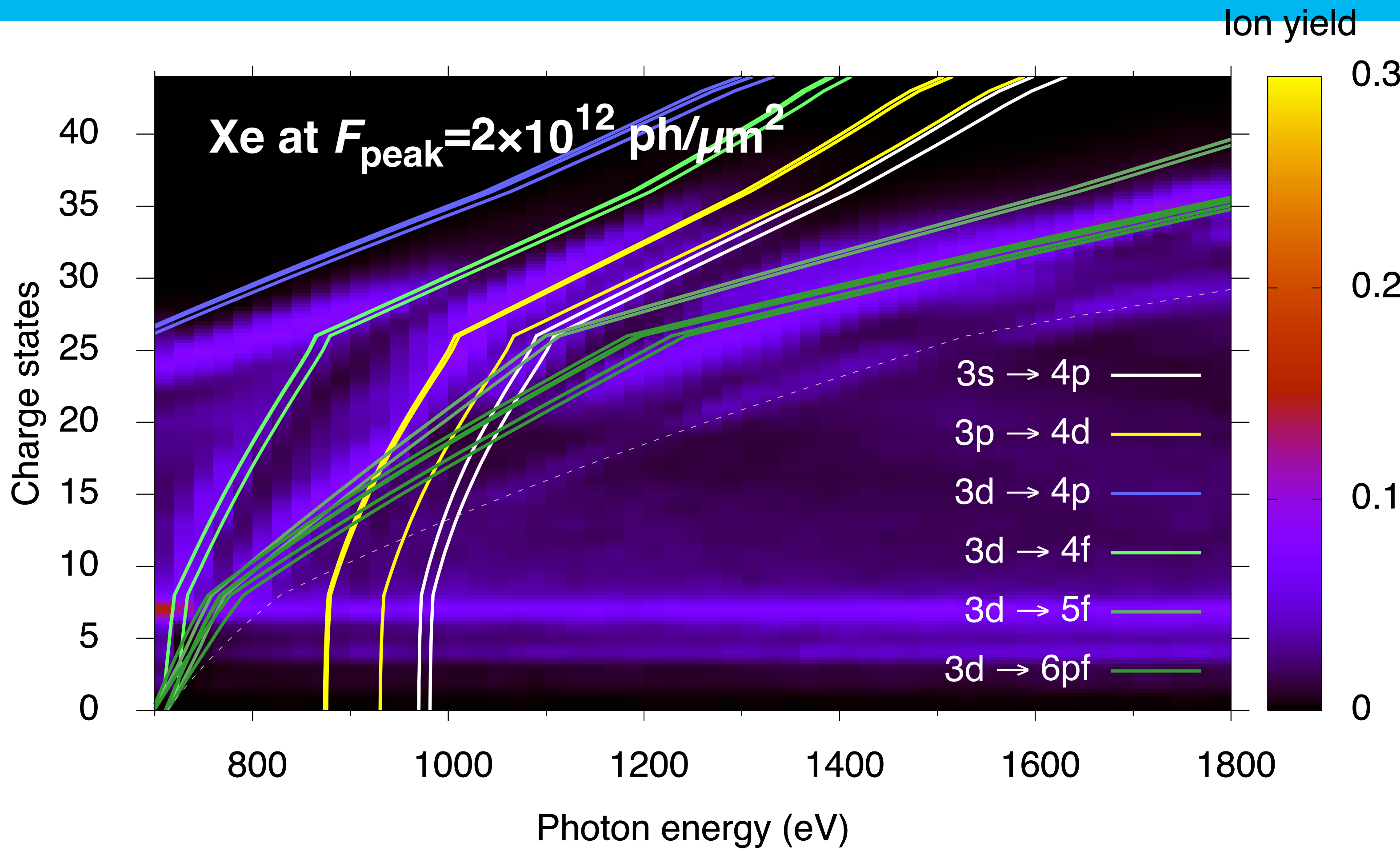
- Photon energy scanned from 700 eV to 1800 eV
- Highest pulse energy (~6–7 mJ) ➔ estimated peak $F\sim2\times10^{12}$ ph/μm^2

Photoabsorption cross section



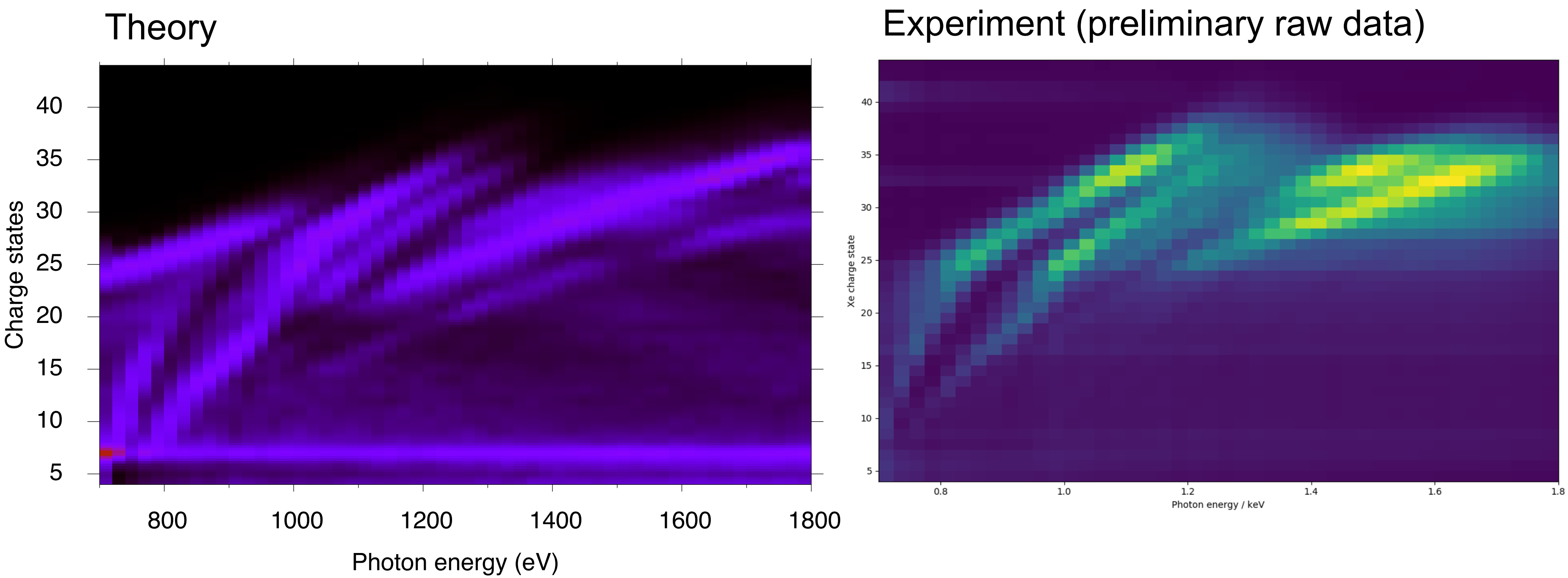
- One-to-one relation between bound–bound transition *E* and the peak structure of photoabsorption cross section

Volume-integrated CSD



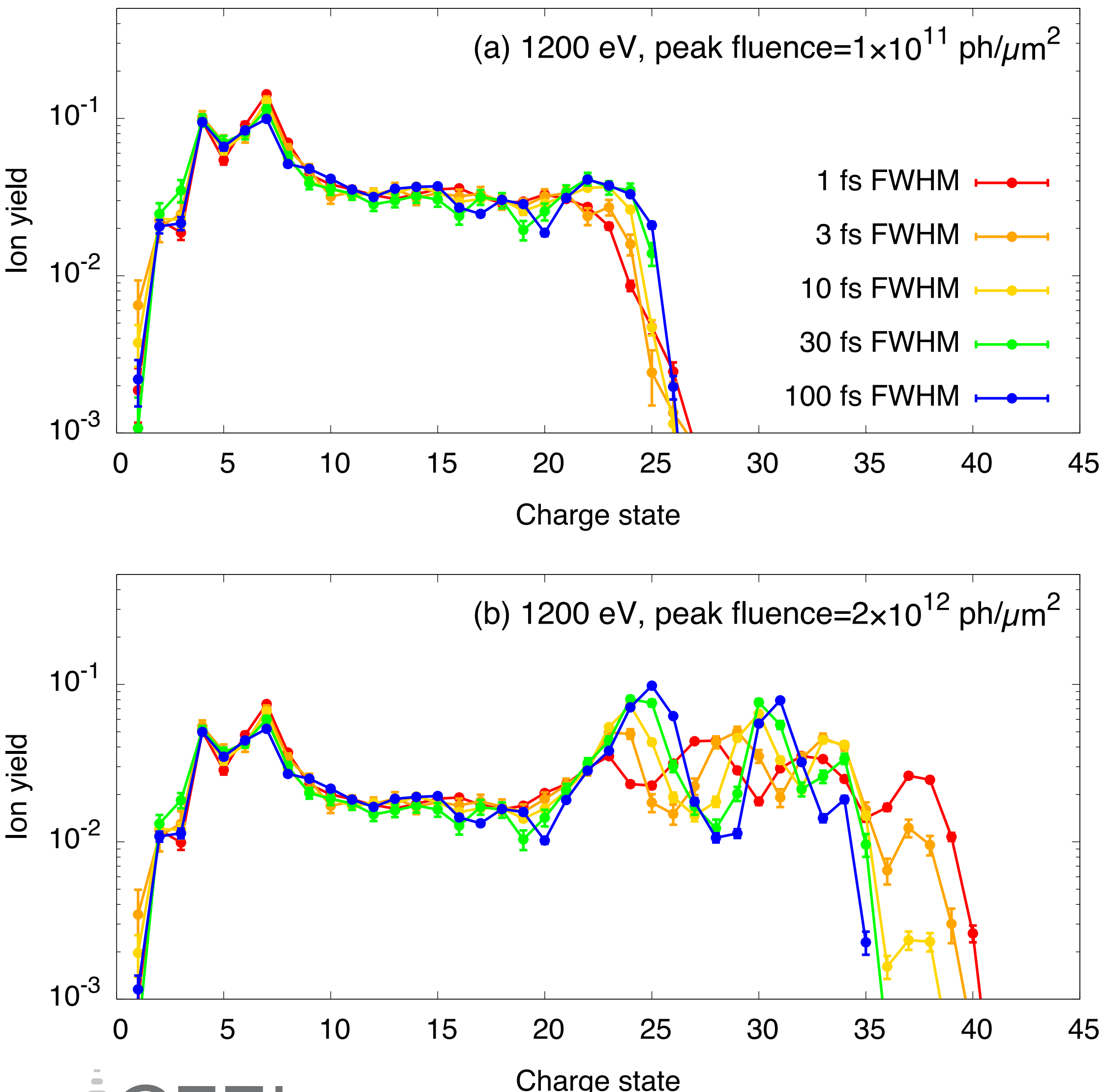
- Volume integration: single Gaussian, F_{peak}=2×10^{12} ph/μm^2 assumed
- Characteristic fringes explained by bound–bound transitions

Comparison with experimental results



- Some spurious signals in theory: n_{max} to be increased
- Volume integration in theory: fluence distrib. to be calibrated with Ar data
- Correction factor in experiment: differences in detector signal heights
- Overall, very good agreement between theory and experiment

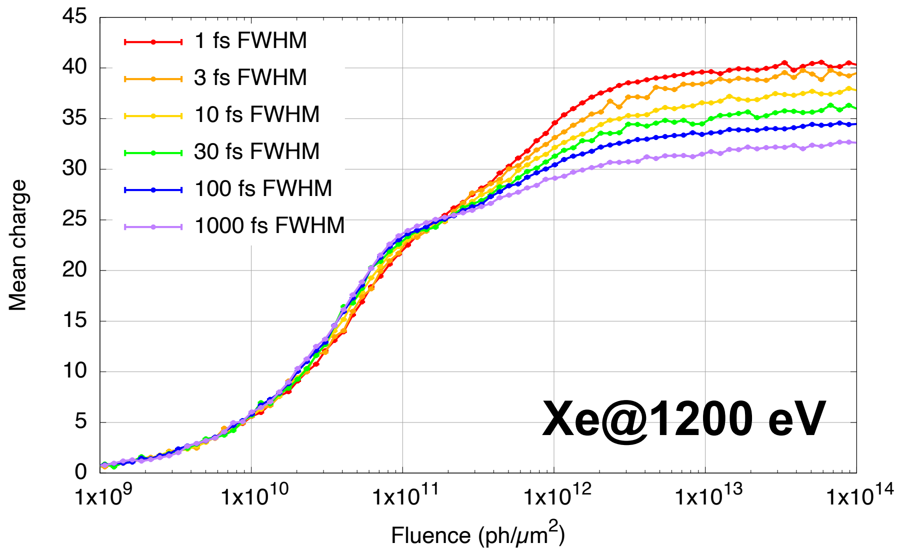
Pulse-duration dep. of volume-integ. CSD



- Effects of volume integration
  - Gaussian spatial profile contains a range of *F*
  - all contributions together
- Low peak fluence: shorter pulse, less ionization (FA)
- High peak fluence: shorter pulse, more ionization (anti-FA)
- Peak structure ➔ fingerprint of pulse duration?

Conclusions

- XATOM: enabling tool for studying X-ray multiphoton physics
- Frustrated absorption: one of the fundamental aspects in the XFEL–matter interaction
- At extremely high X-ray fluence, the paradigm of frustrated absorption can break down



- As an XFEL pulse gets shorter, X-ray multiphoton ionization can be suppressed or enhanced, depending on fluence and photon energy
  ➔ critical benchmark for terawatt-attosecond XFEL pulses
- European XFEL SQS can deliver extremely intense soft X-ray radiation, where we expect to see the breakdown of frustrated absorption