# Multiple-core-hole resonance spectroscopy with ultraintense x-ray pulses

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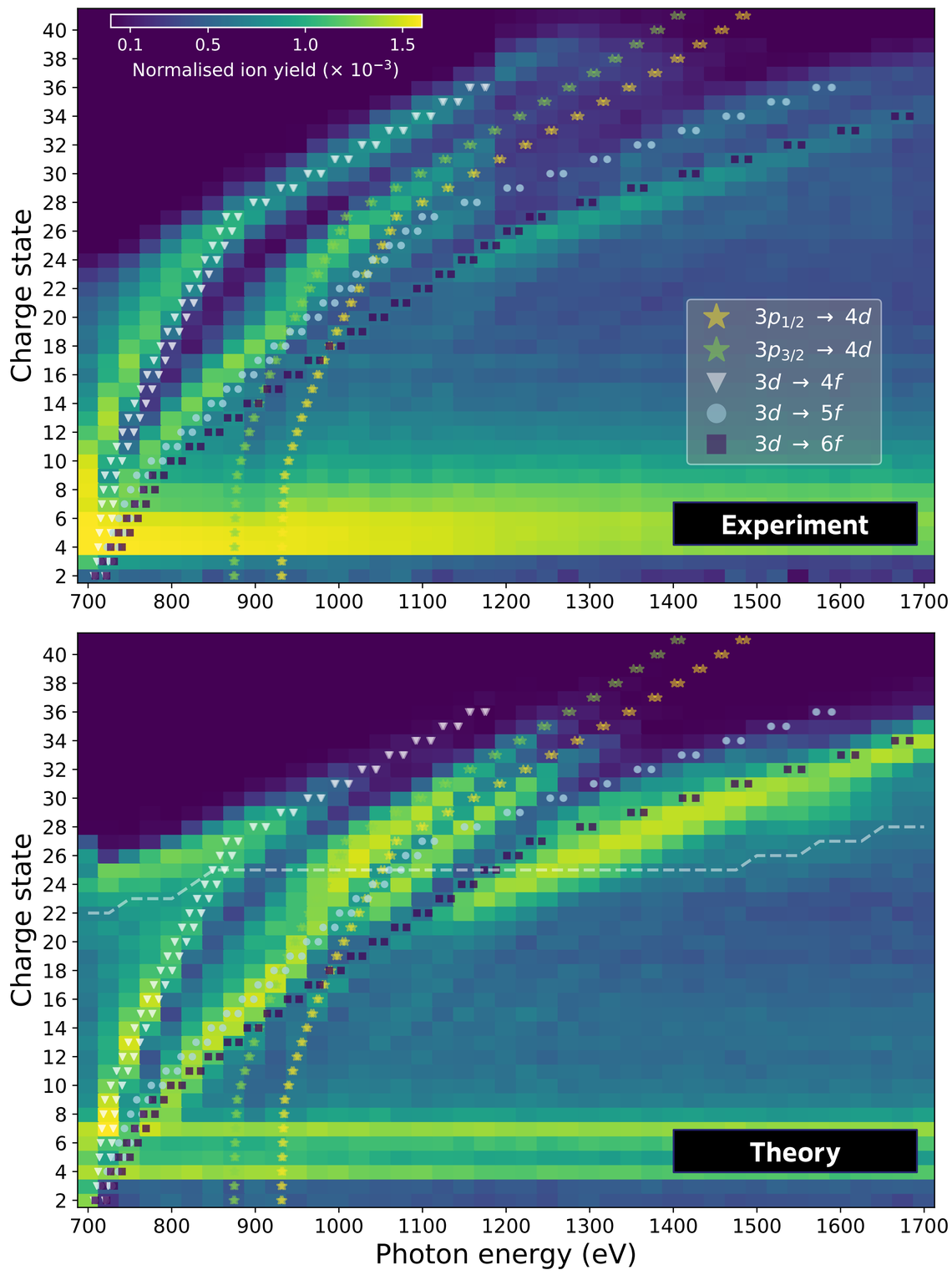
## Abstract

Understanding the interaction of **ultraintense and ultrafast x-ray pulses** with heavy atoms is crucial for gaining insights into the structure and dynamics of matter. One key aspect of nonlinear light-matter interaction is its dependence on the photon energy, but there has been no systematic study of **x-ray free-electron lasers** (XFELs) so far. We present a joint theoretical and experimental study of highly charged xenon ions after interaction with XFEL pulses scanning the photon energy over a wide range, which enables us to map out the transient resonances occurring during the complex charge-up pathways. **Massively hollow atoms** featuring up to six simultaneous core holes determine the spectra at specific photon energies and charge states. The extraction of **resonance spectra** is facilitated by the fact that the ion yields become independent of the peak fluence beyond a saturation point. Our study lays the groundwork for novel spectroscopy of transient atomic species in exotic, **multiple-core-hole states** that have not been explored.

## Photon-energy dependence of charge-state distributions of Xe



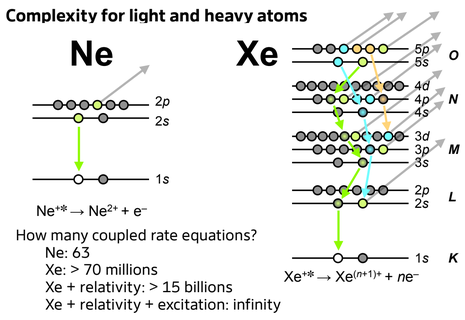

## X-ray multiphoton ionization

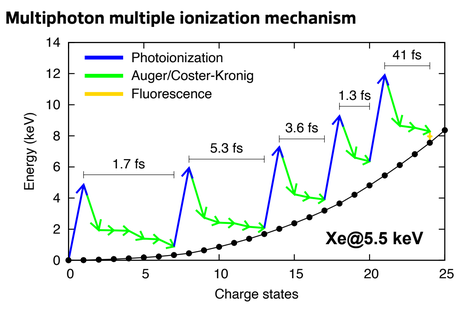
Interaction of matter with intense XFEL pulses is characterized by sequential multiphoton multiple ionization dynamics.

- Sequence of K-shell ionization (P), Auger-Meitner decay (A), and fluorescence (F)
- Extremely complicated ionization dynamics
- Highly excited electronic structure involved
- No standard quantum chemistry code available

We implement an integrated toolkit, **XATOM**, to treat x-ray multiphoton ionization dynamics, based on rate-equation approach, within a consistent theoretical framework of nonrelativistic quantum electrodynamics, perturbation theory, and the Hartree–Fock–Slater model.

**XRAYPAC:**
a software package for modeling x-ray-induced dynamics of matter,
https://www.desy.de/~xraypac/



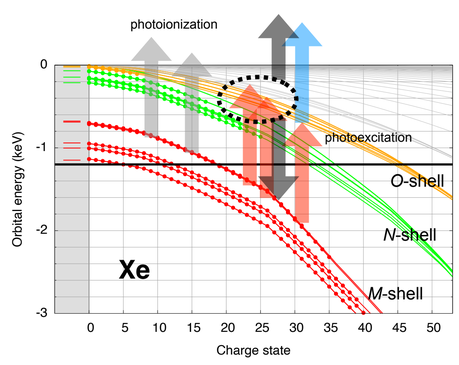

## Resonance effects: XREMPI & REXMI

**XREMPI: x-ray resonance-enhanced multiphoton ionization**

- single resonant excitation and another photoionization
- A. LaForge *et al.*, *Phys. Rev. Lett.* **127**, 213202 (2021).

**REXMI: resonance-enabled x-ray multiple ionization**

- multiple resonant excitations and Auger-Meitner-type decay: further ionization beyond the sequential one-photon ionization limit
- B. Rudek *et al.*, *Nat. Photon.* **6**, 858 (2012).
  B. Rudek *et al.*, *Nat. Commun.* **9**, 4200 (2018).



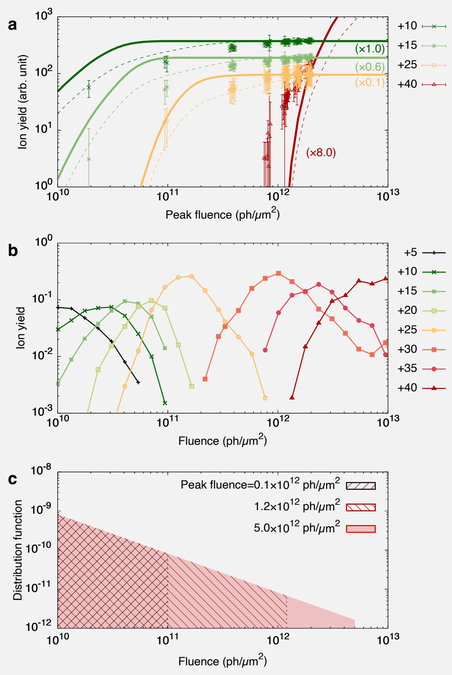

## Saturation effects at high fluences

**Volume integration**

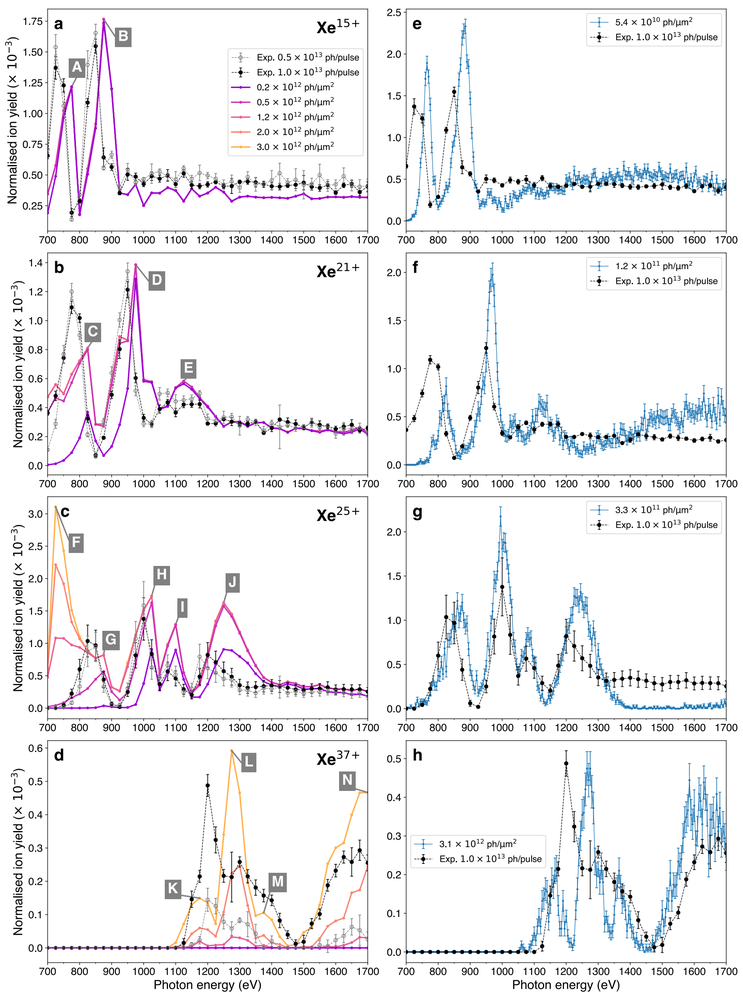
- Taking into account different fluence points in the interaction volume
- Critical for quantitative comparison between theory and experiment

**Peak-fluence insensitivity**

- Before volume integration, a specific charge state is generated with a narrow fluence range
- After volume integration, ion yields become independent of the peak fluence beyond a saturation

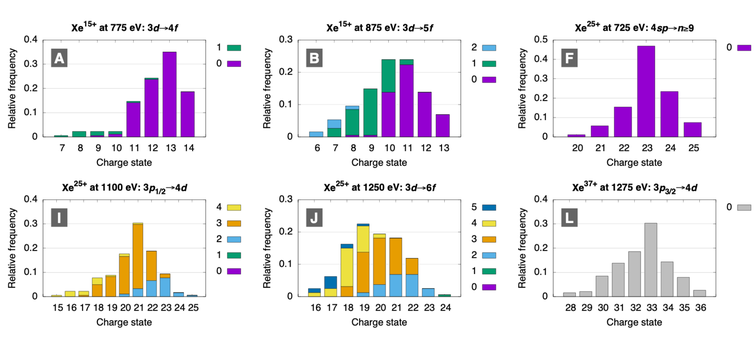


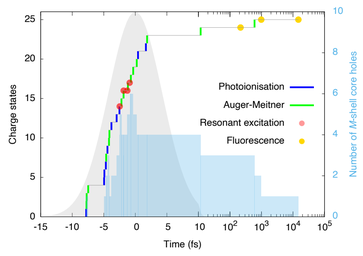

## Resonance ion spectra of Xe




**"Each charge state selects its own local fluence."**

## Multiple-core-hole analysis




**Massively hollow atom formation**
one exemplary ionization pathway, involving six simultaneous core holes

- 6-core-hole: lifetime < 1 fs
- 5-core-hole: lifetime ~ 1 fs
- 4-core-hole: until 12 fs
- 3-core-hole: until 600 fs

## Conclusions

- We present a new type of resonance ion spectroscopy using ultraintense femtosecond x-ray radiation
  - > Wide tunability of photon energy at European XFEL
  - > Exploit saturation of ion yields at very high fluences
- Transient multiple-core-hole states (up to six simultaneous core holes) are found to be crucial for explaining the peaks in the resonance spectra
- Extremely short-lived, as well as unusually long-lived, highly charged ions in exotic electronic configurations can be created and probed with intense x-ray pulses

## Publication

A. Rörig, S.-K. Son, R. Boll, *et al.*, *Nat. Commun.* **14**, 5738 (2023).

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