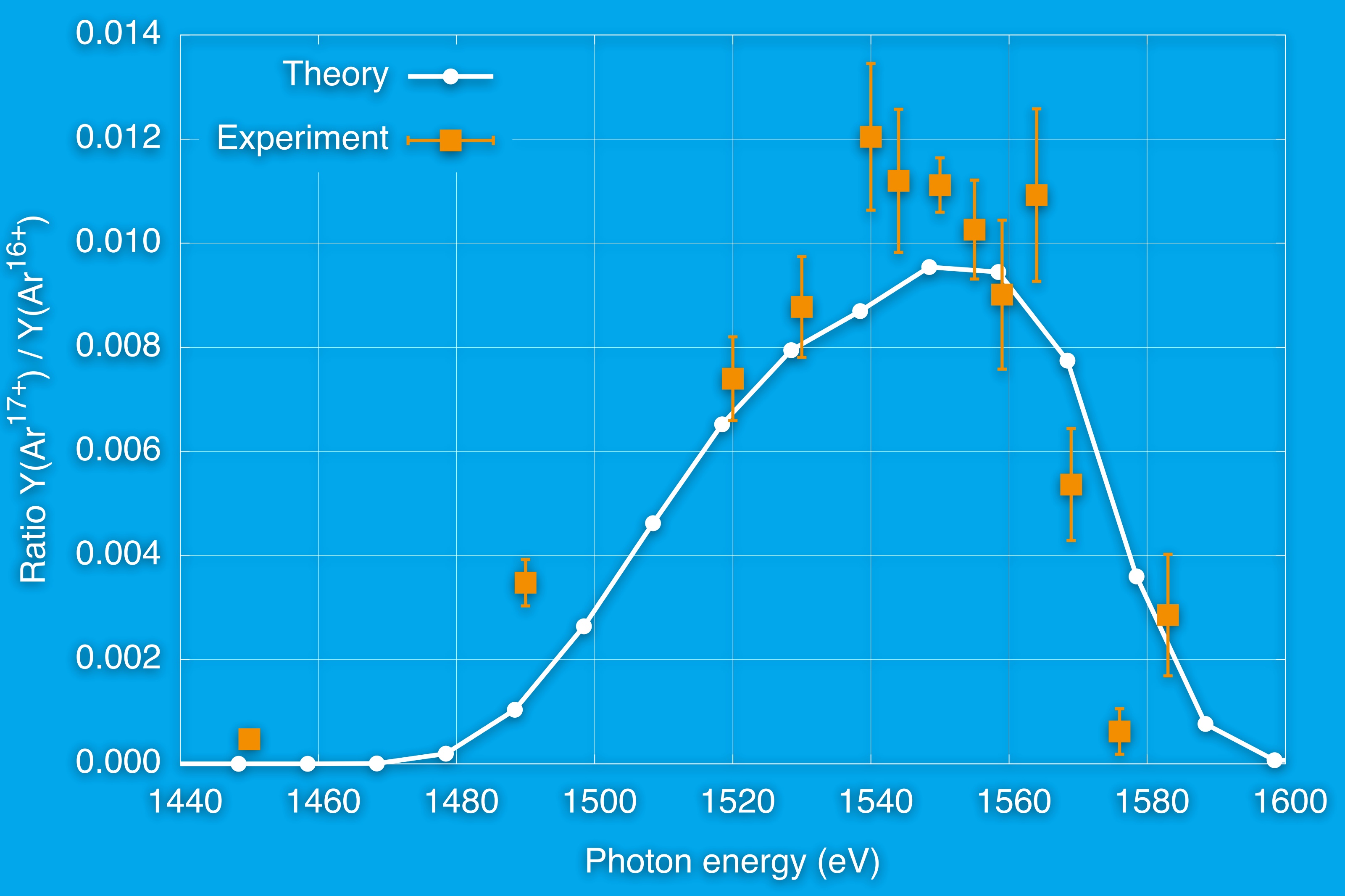
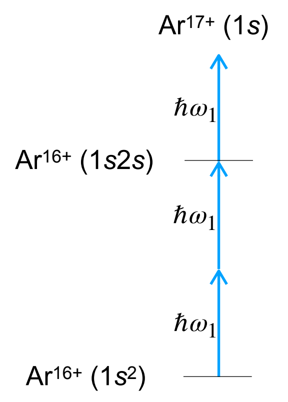
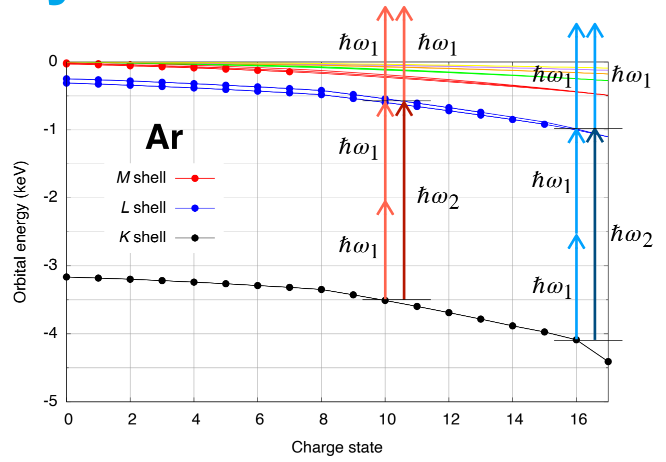
We first observed resonance-enhanced multiphoton ionization in the x-ray regime (XREMPI) using European XFEL.



Ionization pathways

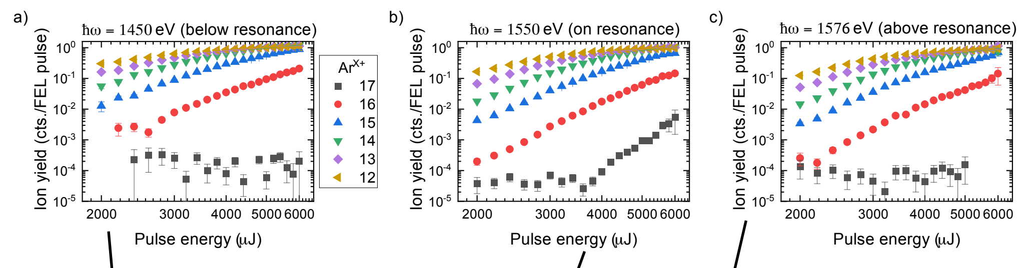


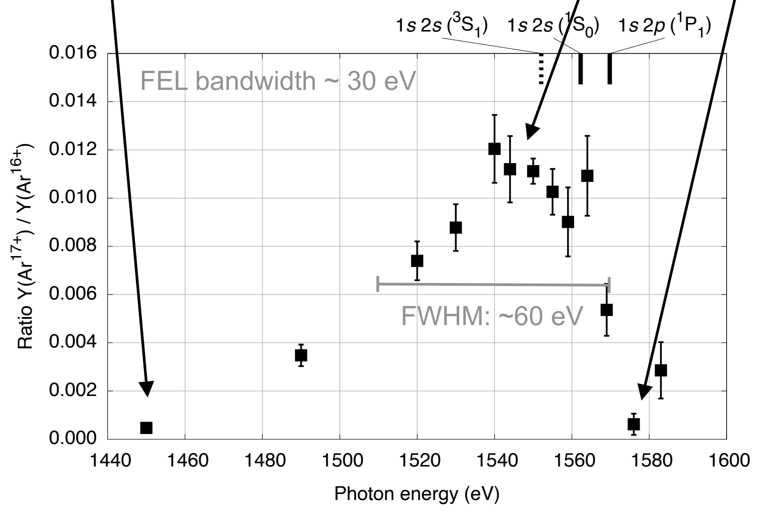


- IP of Ar^{16+} = 4130 eV
- Photon energy ~ 1550 eV
- Detection of Ar^{17+} ➔ (2+1)-REMPI?
- Any other ionization pathways?

- Various ionization channels besides (2+1)-REMPI
- 2nd harmonic (0.2% contrib.) ➔ (1'+1)-REMPI
- At low charges ➔ (2+*n*)-REMPI or (1'+*n*)-REMPI

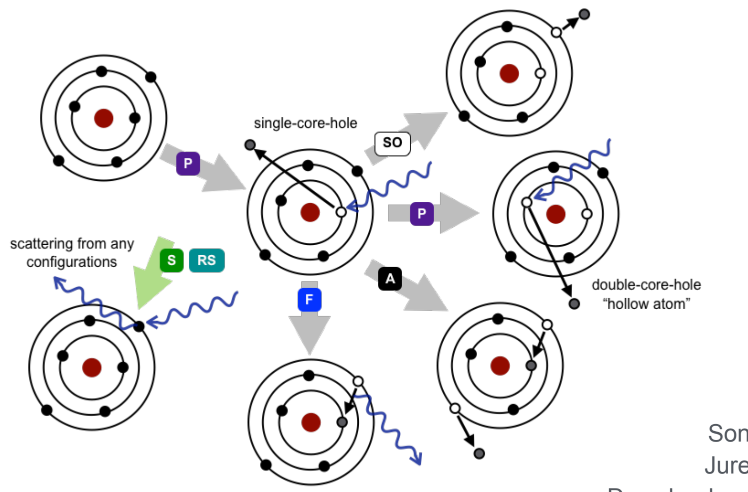
Experiment: European XFEL





- Small Quantum System (SQS) scientific instrument at the European XFEL
- Pulse length: 25 fs FWHM (nominal) / focal size: approx. 1.5×1.5 μm^2 (FWHM) / pulse energy: 2~6 mJ
- Photon energy: 1450~1583 eV / energy bandwidth: approx. 1% (FWHM) / second harmonic contrib.: est. 0.2~0.6%
- **Broad, red-shifted, asymmetric** resonance profile in stark contrast to the conventional REMPI

Theory: XATOM

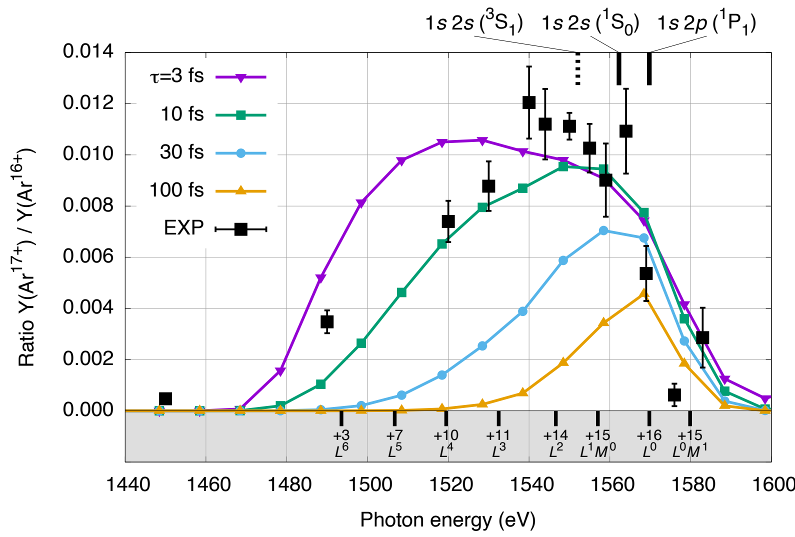


- X-ray-induced atomic processes calculated for any given element and configuration
- Electronic structure based on the Hartree–Fock–Slater model
- Ionization dynamics solved by a rate-equation approach
- Sequential ionization model has been tested by a series of atomic XFEL experiments

Son *et al.*, *Phys. Rev. A* **83**, 033402 (2011).
Jurek *et al.*, *J. Appl. Cryst.* **49**, 1048 (2016).

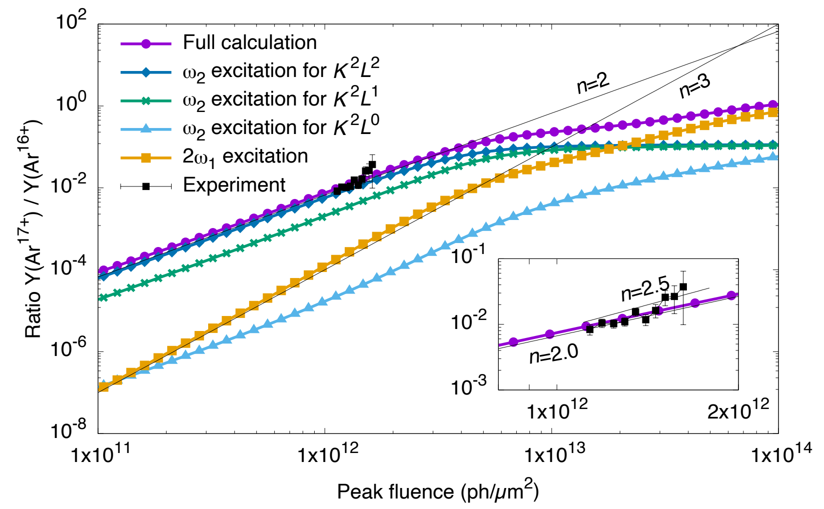
Download executables: http://www.desy.de/~xraypac

Comparison between theory and experiment



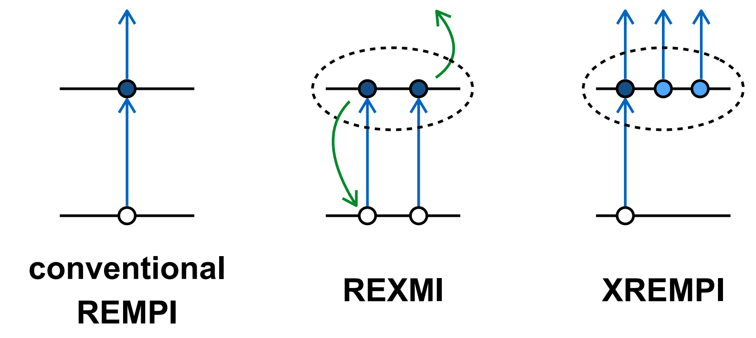
- Calculation with 10-fs matches well with experimental data.
- In experiment, the resonance profile looks all different from conventional REMPI.
- In theory, the profile is not only broadened but also shifted to lower energies as the pulse length gets shorter.
- The predicted pulse-length dependence cannot be explained by ordinary REMPI, because the same bandwidth is applied and AC Stark shift is negligible in the x-ray regime.
- It can be explained by the **various ionization pathways at lower charges** and associated decay lifetimes, rather than the bandwidth ➔ potentially applicable to characterize FEL beam parameters.

Analyzing resonant processes



- Add and subtract processes in calculations
- **(1'+3)-REMPI at Ar^{14+}**
- **(1'+2)-REMPI at Ar^{15+}**
- **(1'+1)-REMPI at Ar^{16+}**
- **(2+*n*)-REMPI at all *Q***
- Dominant process: resonant excitation by 2nd harmonic at Ar^{14+} (more precisely, $K^2L^2M^m$ for $0 \leq m \leq 8$)

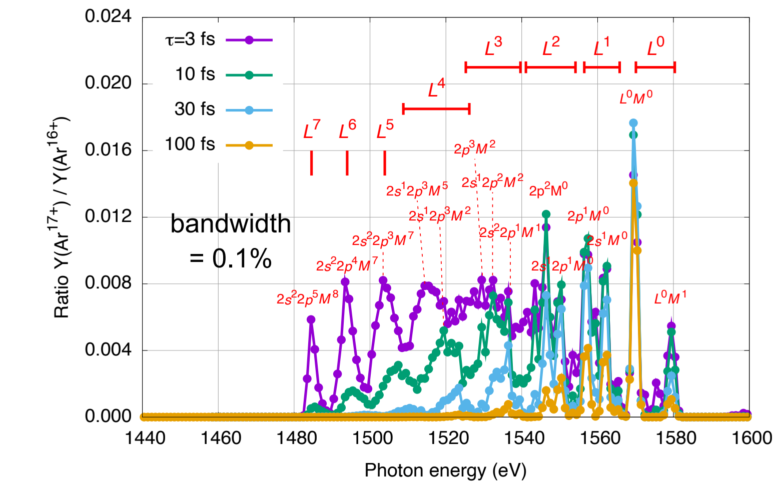
REMPI vs. REXMI vs. XREMPI



- **REXMI** (resonance-enabled or enhanced x-ray multiple ionization): multi-electron excitation involved; electron-correlation-driven relaxation; broad bandwidth favorable
- **XREMPI**: single-electron excitation; narrow bandwidth favorable; not necessarily single ionization; influenced by ultrafast decay processes

REXMI: Rudek *et al.*, *Nat. Photon.* **6**, 858 (2012);
Rudek *et al.*, *Nat. Commun.* **9**, 4200 (2018).

Narrower FEL bandwidth



- SASE FEL bandwidth given by the shortest pulse length of spiky pulses, typically ~1%
- Narrower bandwidth through the use of a monochromator or self-seeding techniques
- With narrower bandwidth, individual resonance structures of electron config. could be resolved ➔ potentially precision spectroscopy of highly charged ions of astrophysical relevance

DESY. CFEL SCIENCE

Resonance-enhanced multiphoton ionization in the x-ray regime.

Sang-Kil Son*, Aaron C. LaForge, Debadarshini Mishra, Markus Ilchen, Stephen Duncanson, Eemeli Eronen, Edwin Kukk, Stanislaw Wirok-Stoletow, Daria Kolbasova, Peter Walter, Rebecca Boll, Alberto De Fanis, Michael Meyer, Yevheniy Ovcharenko, Daniel E. Rivas, Philipp Schmidt, Sergey Usenko, Robin Santra, Nora Berrah

A. C. LaForge *et al.*, *Phys. Rev. Lett.* **127**, 213202 (2021).