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

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Synopsis

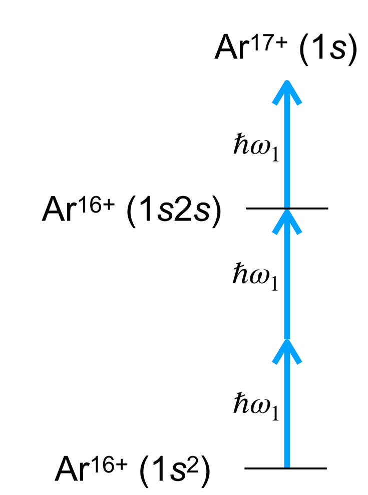
Here, we report on resonance-enhanced multiphoton ionization (REMPI) of argon atoms in the short wavelength regime using ultraintense x rays from the European XFEL. We demonstrate and discuss the differences between X-ray and conventional REMPI.

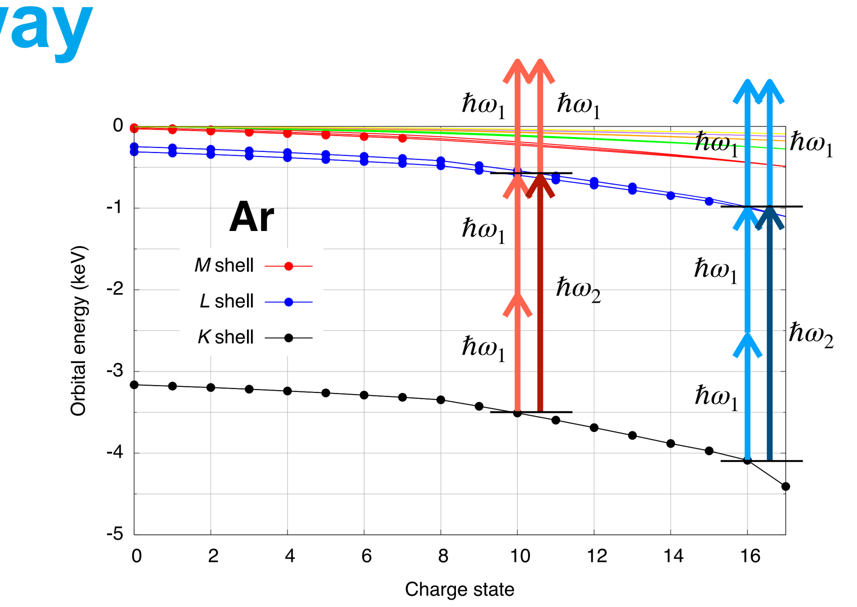
Introduction

Multiphoton ionization is one of the fundamental nonlinear processes when matter interacts with intense laser fields. In particular, REMPI has been a widely-used spectroscopic technique due to high sensitivity and selectivity. X-ray free-electron lasers have offered new avenues for studying x-ray multiphoton ionization. Extending REMPI to the x-ray regime, however, requires entirely different physical processes and interpretation. Conventional REMPI at long wavelengths relies on the resonant excitation of a valence electron where the only relaxation pathway is radiative decay. On the other hand, a core-excited state after x-ray resonant excitation is subject to Auger decay, which is orders of magnitude faster than radiative decay. Thus, the complex interplay between Auger processes and REMPI renders this process challenging to fully resolve in the x-ray regime.

We present a first observation of REMPI in the x-ray regime. We observe nonlinear ionization to create Ar^{17+}, where photon energies are insufficient to directly ionize a 1*s* electron. With the aid of state-of-the-art theoretical modeling, we attribute the ionization to a two-color REMPI-like process where the second harmonic creates a $1s \rightarrow 2p$ transition and the fundamental pulse subsequently ionizes the system. The measured resonance profile of x-ray REMPI shows a broad, asymmetric, red-shifted distribution, which is a clear distinction from the conventional REMPI case. Moreover, theoretical results demonstrate a strong pulse-length dependence of the resonance profile. Our analysis shows that the REMPI process occurs not only for Ar^{16+} but also for lower charge states, where multiple ionization competes with Auger lifetimes. We find the observed broadband nature and pulse-length dependence of the resonance profile to be due to overlapping resonances with lower Ar charge states.

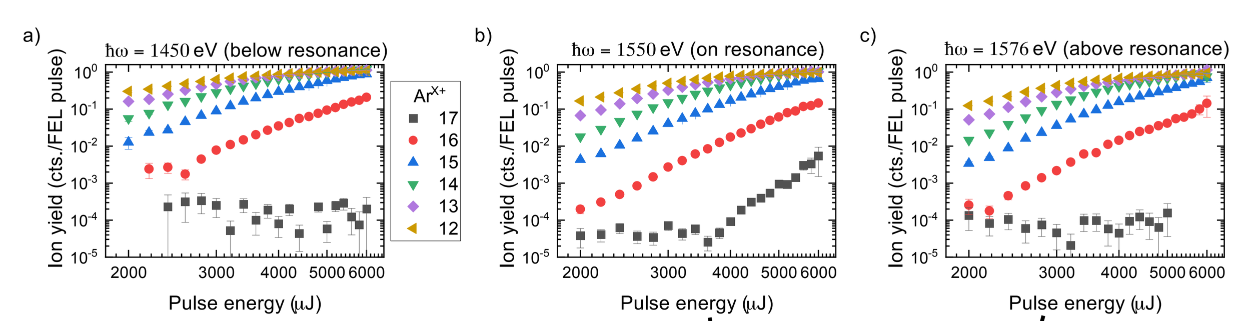
Ionization pathway

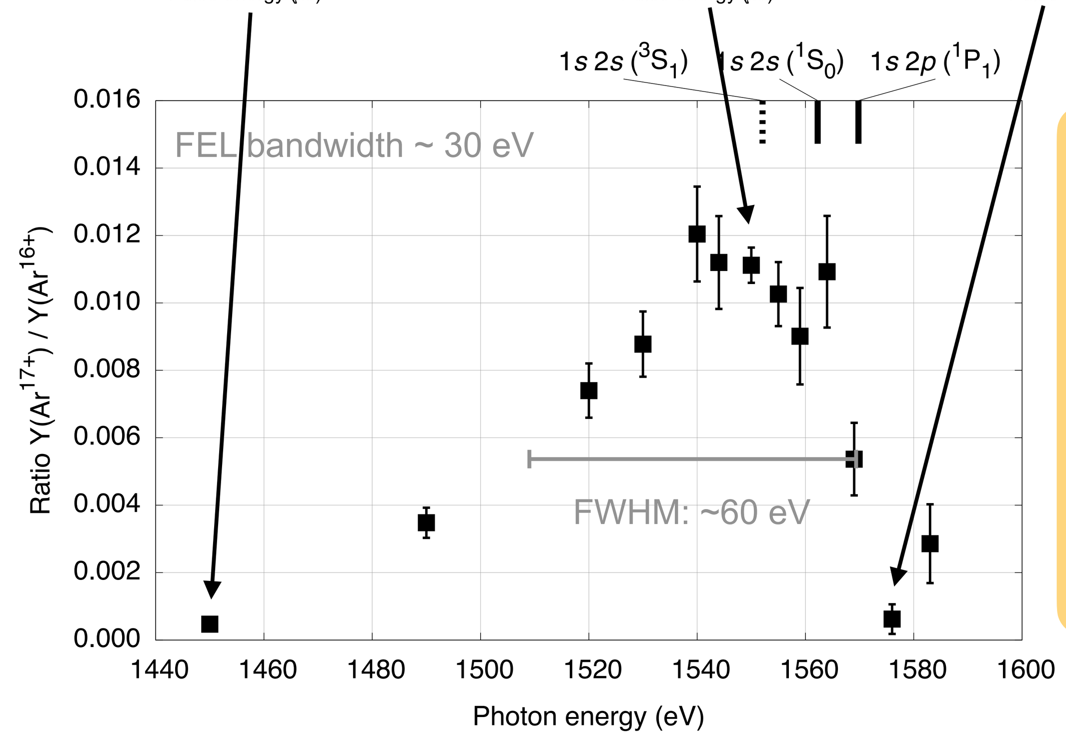




- IP of Ar^{16+} = 4130 eV
- Photon energy ~ 1550 eV
- Detection of Ar^{17+} ➔ (2+1)-REMPI?
- Any other ionization pathways?
- Various ionization channels besides (2+1)-REMPI
- 2nd harmonic (0.2% contrib.) ➔ (1'+1)-REMPI
- At low charges ➔ (2+*n*)-REMPI or (1'+*n*)-REMPI

Experiment: European XFEL

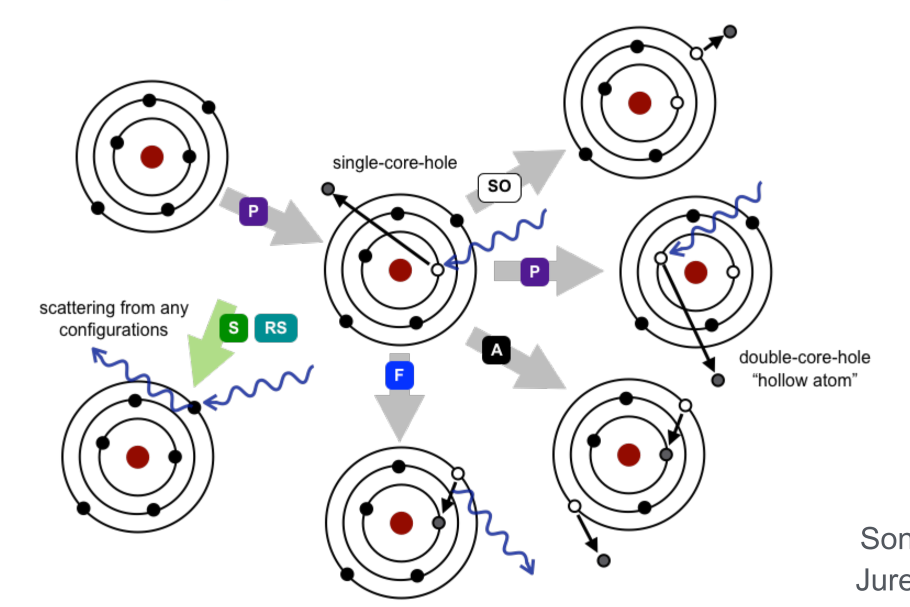




New observation: broad, red-shifted, asymmetric resonance profile

- Small Quantum System (SQS) scientific instrument at the European XFEL
- Pulse duration: 25 fs FWHM (nominal)
- Focal size: approx. 1.5×1.5 μm² (FWHM)
- Photon energy: 1450 eV to 1583 eV
- Energy bandwidth: approx. 1% (FWHM)
- Pulse energy: 2 mJ to 6 mJ
- Second harmonic contrib.: est. 0.2~0.6%

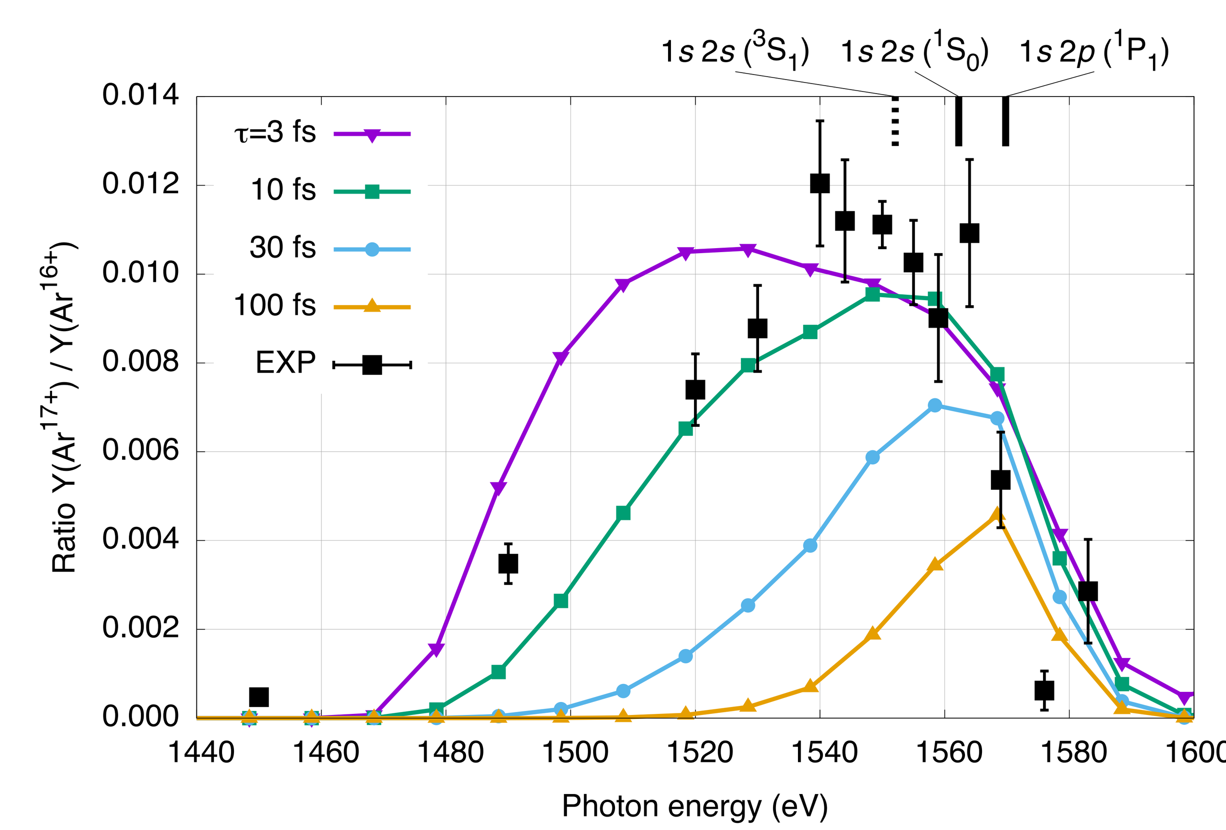
Theory: XATOM



- 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

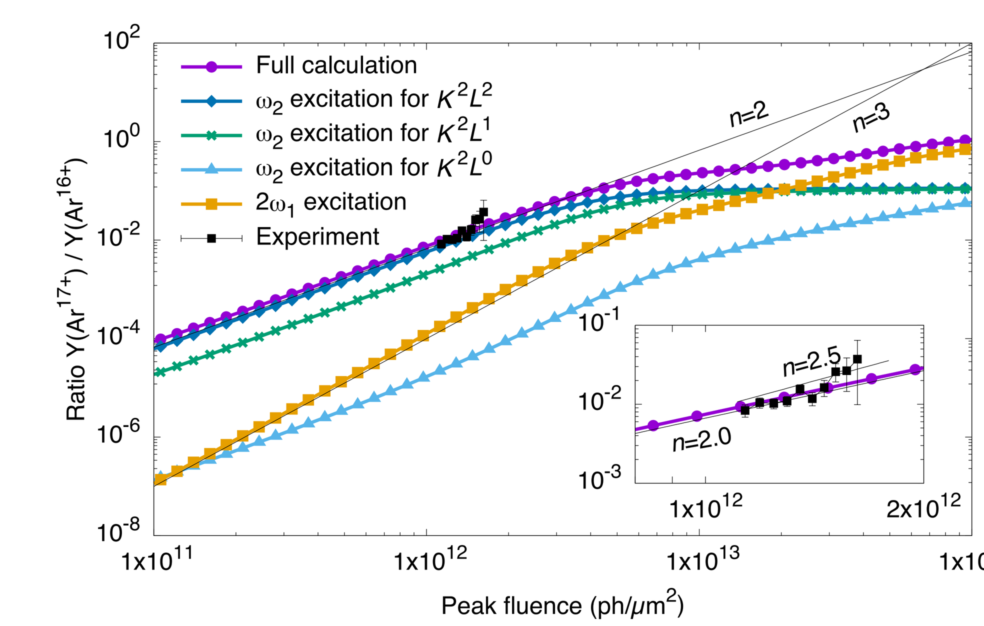
Pulse-length dependence



- Calculation with 10-fs matches well with experimental data.
- In experiment, the resonance profile looks all different from conventional REMPI.
- In theory, the predicted pulse-length dependence cannot be explained by ordinary REMPI, because of the same bandwidth applied and negligible AC Stark shift in the x-ray regime.
- It can be explained by the various ionization pathways and associated decay lifetimes, rather than the bandwidth ➔ potentially applicable to characterize FEL beam parameters.

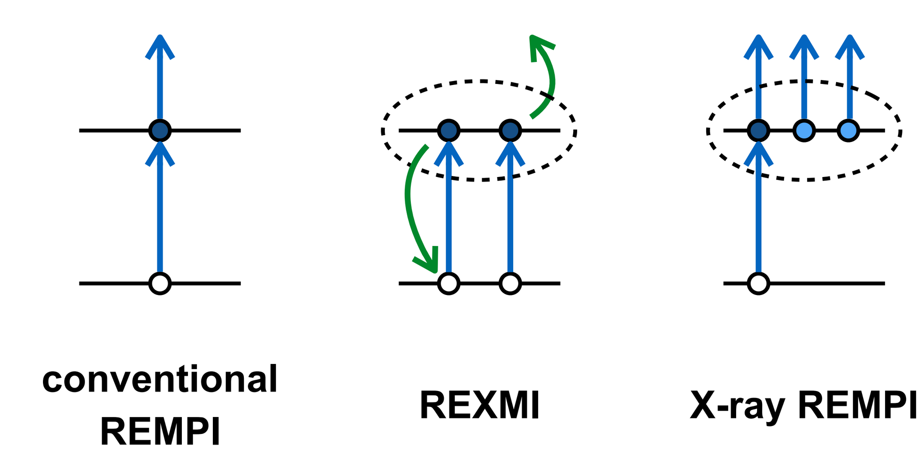
New prediction: the profile is broadened and shifted to lower energies as the pulse length gets shorter

Analyzing resonant processes



- Add and subtract processes in calculation
- (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 $0 \leq m \leq 8$)

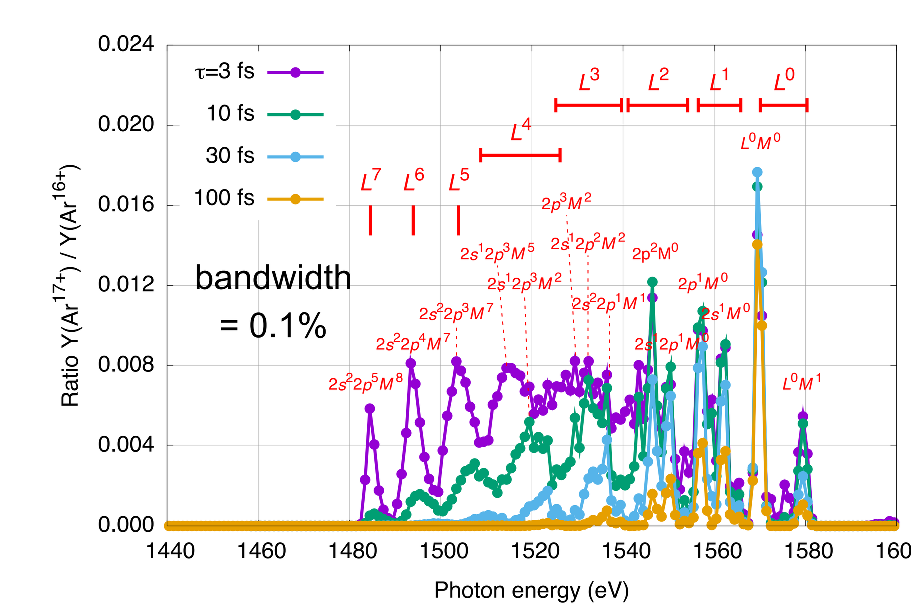
REMPI vs. REXMI vs. X-ray REMPI



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

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

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

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