Unusual phenomena in x-ray multiphoton ionization of atoms.

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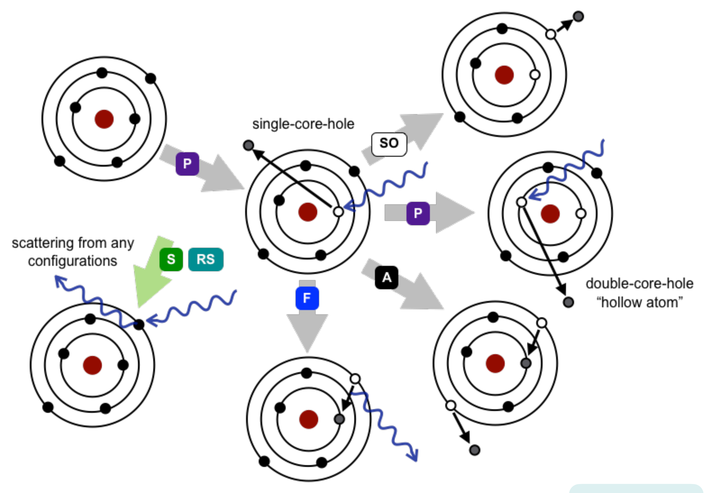
Abstract

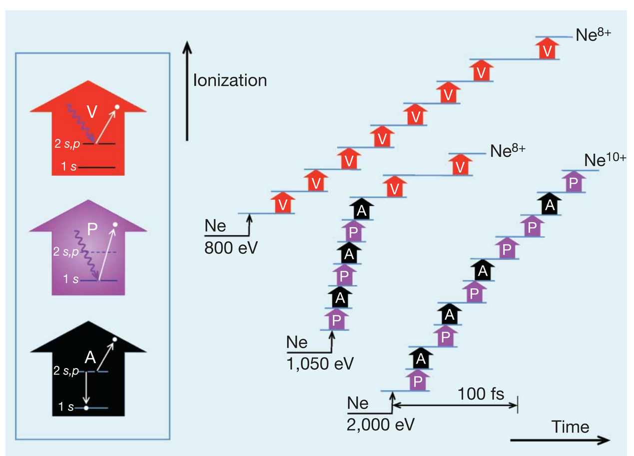
Unprecedentedly intense x-ray pulses, provided by x-ray free-electron laser (XFEL) facilities, can create unusual, highly excited states of matter, which have not been conceivable with conventional light sources. Interaction of atoms with intense XFEL pulses is characterized by x-ray multiphoton ionization, where complex ionization dynamics are involved with a sequence of photoionization events and accompanying relaxation processes. We have developed an integrated toolkit to describe x-ray-induced atomic physics, XATOM, which has been a key development for interpreting and designing XFEL experiments and advancing XFEL science. In this contribution, we will present an overview of XATOM, highlighting two recent results: the breakdown of frustrated absorption [10] and x-ray resonance-enhanced multiphoton ionization [11].



Diagrams of X-ray–atom interaction Ref. [7]

X-ray multiphoton ionization

Interaction with *ultraintense* x-ray pulses

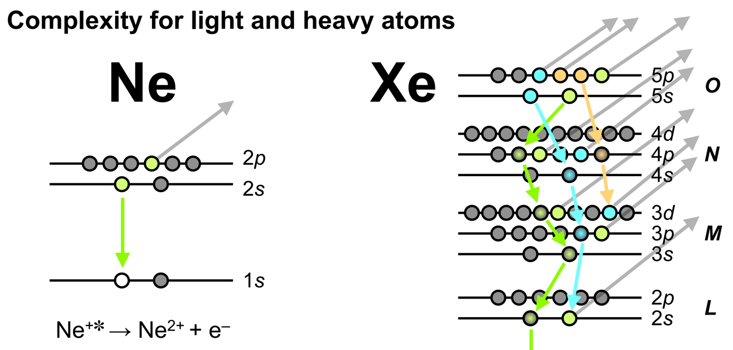


Young *et al.*, *Nature* **466**, 56 (2010).

Interaction of matter with intense XFEL pulses is characterized by sequential multiphoton multiple ionization dynamics.
> First experiment of Ne: fundamental atomic physics in XFEL
> Sequence of K-shell ionization (P), Auger decay (A), and fluorescence (F)
> Extremely complicated ionization dynamics
> Highly excited electronic structure involved
> No standard quantum chemistry code available

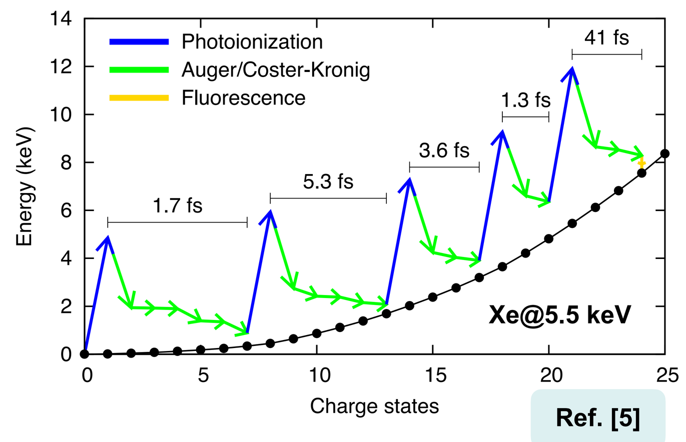
We implement an integrated toolkit, XATOM, to treat x-ray multiphoton ionization dynamics, based on rate-equation approach, within a consistent theoretical framework of nonrelativistic quantum electrodynamics, perturbation theory, and the Hartree–Fock–Slater model.

Complexity for light and heavy atoms



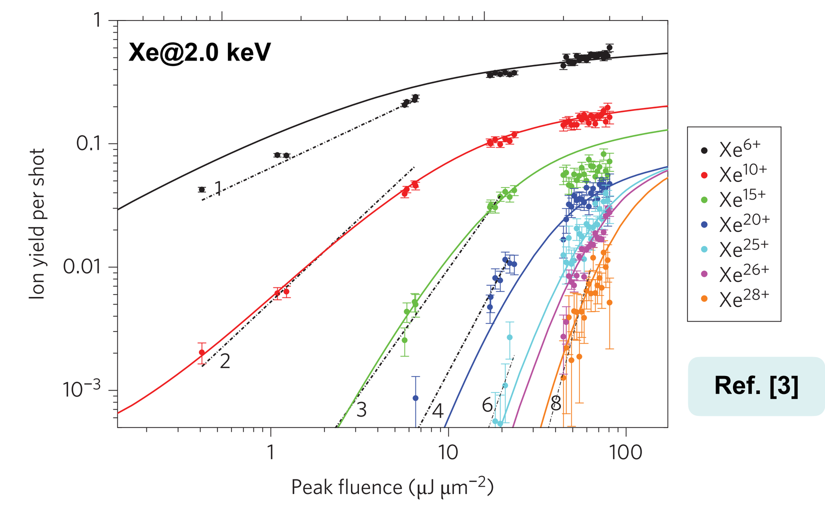
How many coupled rate equations?
Ne: 63
Xe: > 70 millions
Xe + relativity: > 15 billions
Xe + relativity + excitation: infinity
Ref. [8]

Multiphoton multiple ionization mechanism



Ref. [5]

Quantitative comparison between theory and experiment



Ref. [3]

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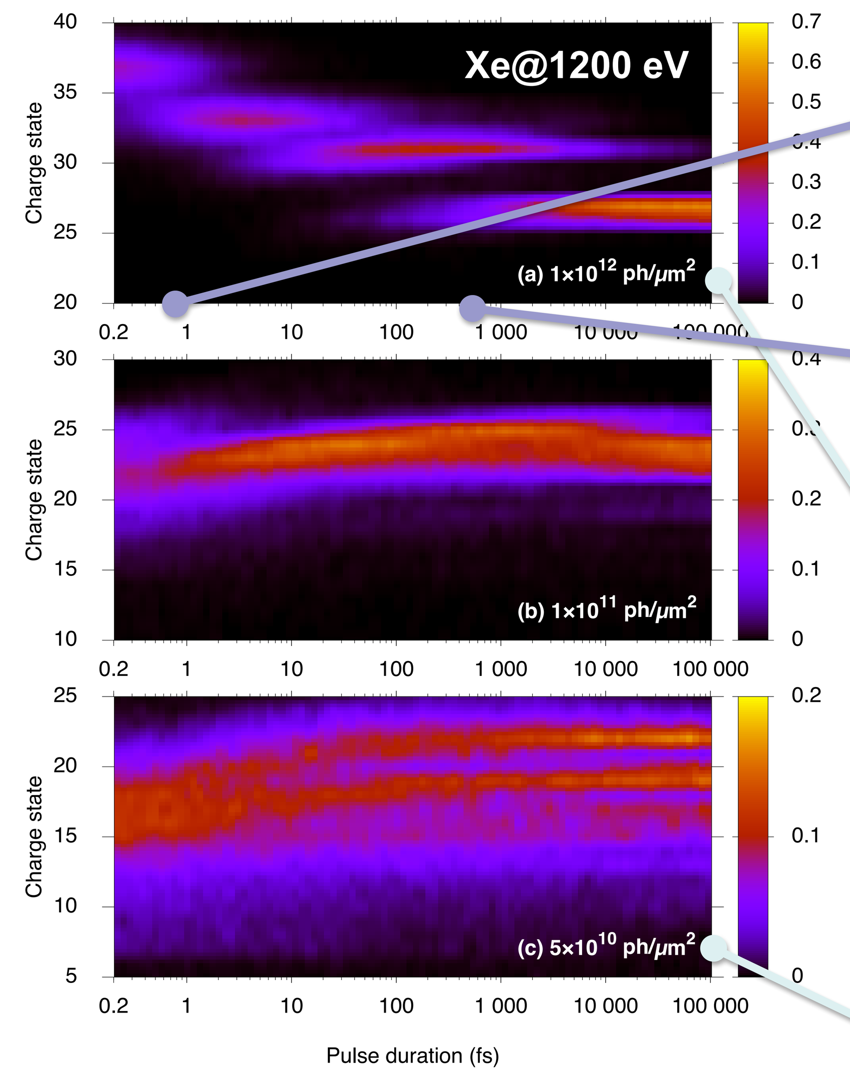
Breakdown of frustrated absorption

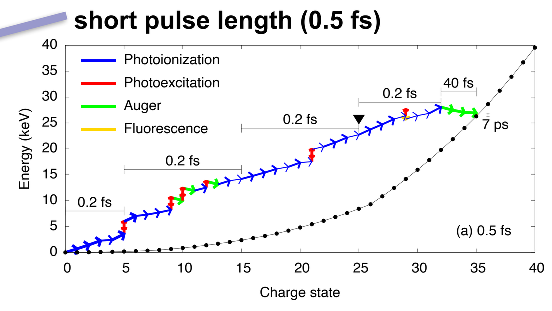
Ref. [10]

Charge-state distribution of Xe at 1200 eV

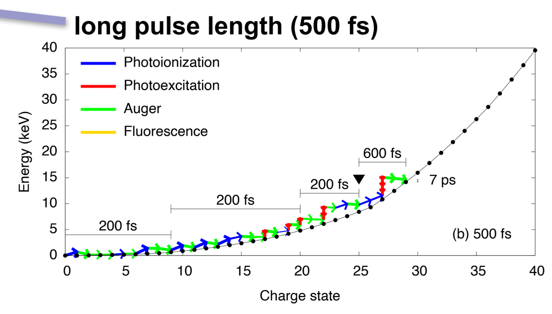


Ionization pathway at different pulse lengths
> A short pulse facilitates multiple-core-hole formation
> Detour of ionization barrier makes a higher charge state

short pulse length (0.5 fs)

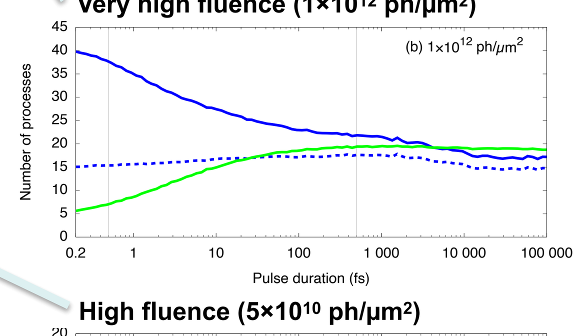


long pulse length (500 fs)

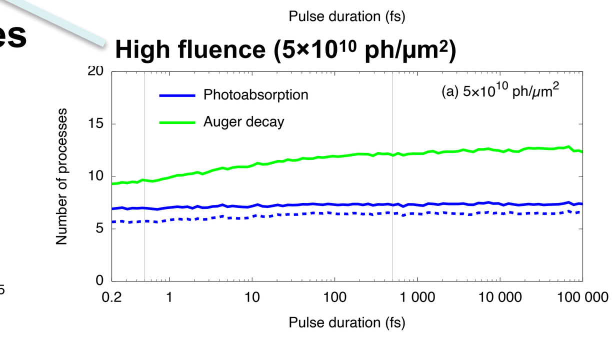


Frustrated absorption or suppression of ionization
> At intermediate or high fluence ➔ frustrated absorption
> At extremely high fluence ➔ anti-frustrated absorption

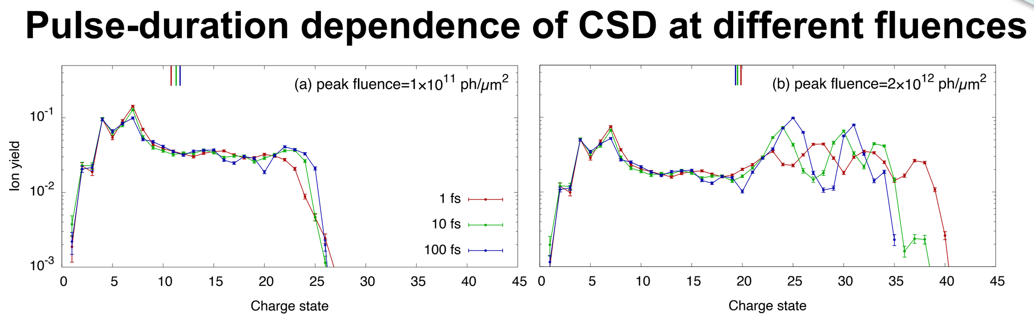
Very high fluence (1×10^{12} ph/μm²)



High fluence (5×10^{10} ph/μm²)

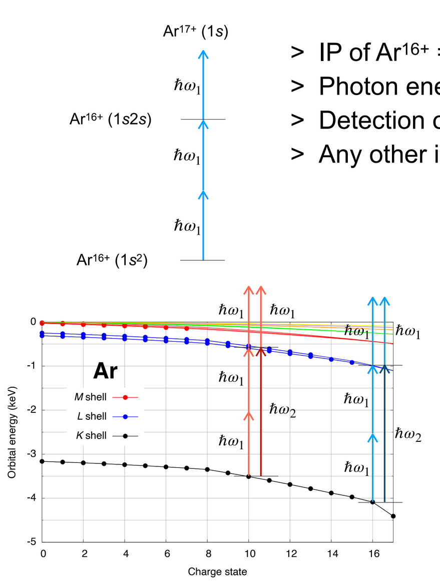


Pulse-duration dependence of CSD at different fluences



X-ray resonance-enhanced multiphoton ionization

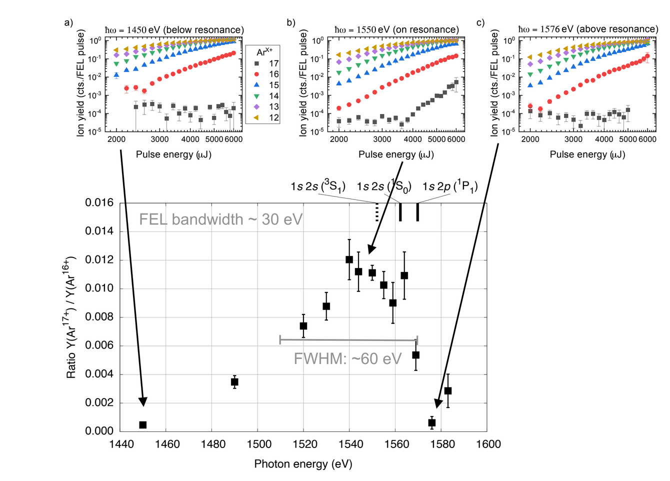
Ionization pathways of Ar



> 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

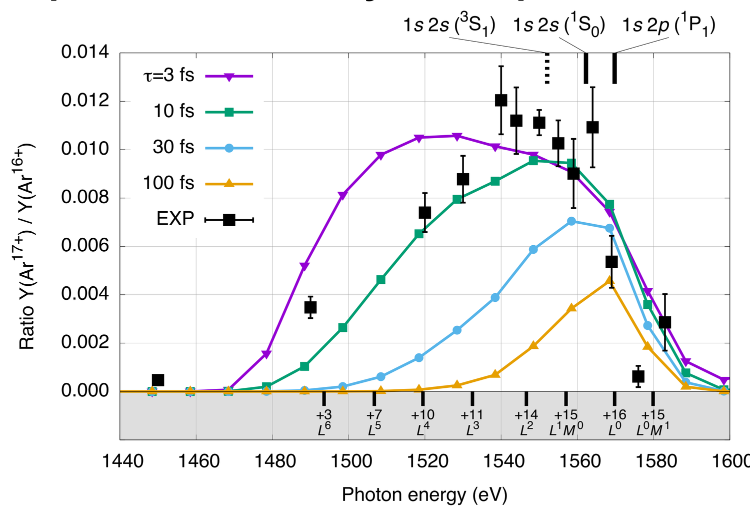
Experiment: European XFEL



> Small Quantum System (SQS) scientific instrument
> Pulse length: 25 fs FWHM (nominal) / focal size: approx. 1.5×1.5 μm² (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

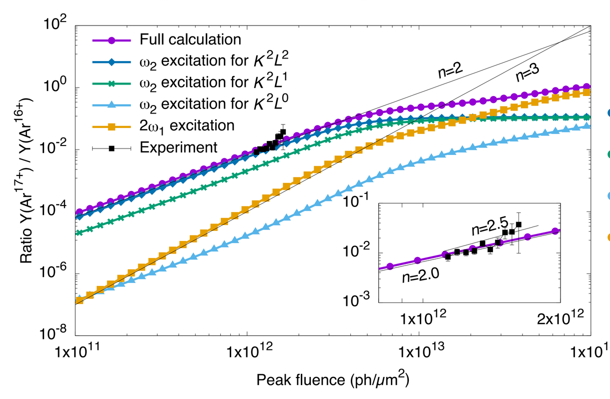
Comparison b/w theory and experiment

Ref. [11]



> In theory, the profile is not only broadened but also shifted to lower energies as the pulse length gets shorter.
> Calculation with 10-fs matches well with experimental data.
> 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.

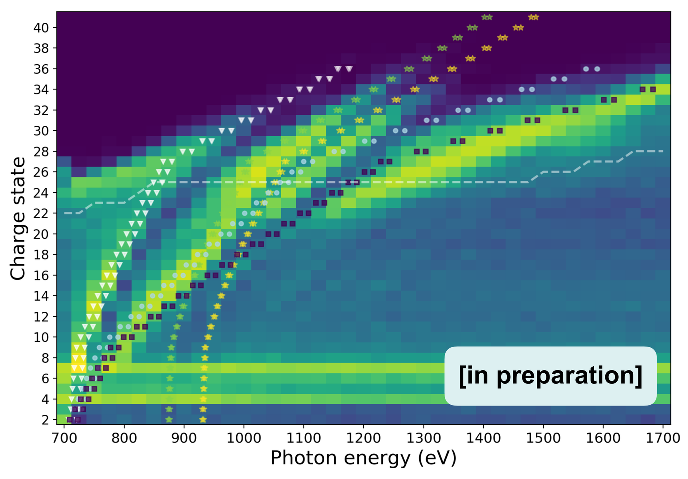
Analyzing resonant processes



• (1′+3)-REMPI at Ar^{14+}
• (1′+2)-REMPI at Ar^{15+}
• (1′+1)-REMPI at Ar^{16+}
• (2+*n*)-REMPI at all *Q*

> Add and subtract processes in calculations
> Dominant process: resonant excitation by 2nd harmonic at Ar^{14+} (more precisely, $K^2L^2M^m$ for $0\le m\le 8$)

XATOM

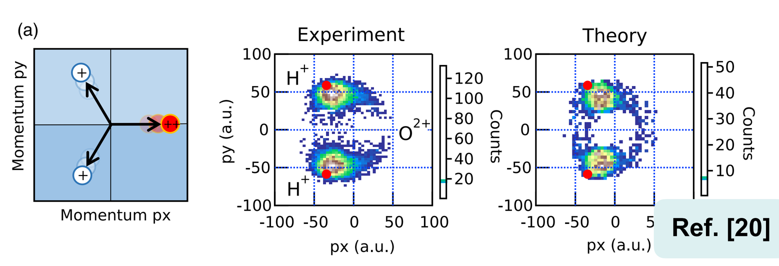


Photon-energy-dependent Xe charge-state distribution

XATOM has been extended to include the resonance and relativistic effects. Recently it has been employed for a joint theoretical and experimental study of multiple-core-hole resonance spectroscopy of Xe atoms irradiated by ultraintense soft-x-ray pulses. With unprecedented wide tunability offered by the variable-gap undulators available at the European XFEL, the photon energy is scanned over a wide range of 1 keV, while maintaining a constant 10^{13} photons on target. The ion yields as a function of photon energy show rich structures. XATOM reveals that they originated from resonance excitations of a broad range of precursor charge states and formation of multiple-core-hole states.

Poster 27: Multiple-core-hole resonance spectroscopy with ultraintense X-ray pulses, presented by **Aljoscha Rörig**

XMOLECULE



Ref. [20]

Dissociation dynamics of H_2O imaged in coincident ion momenta

We develop an x-ray molecular physics toolkit, XMOLECULE. The molecular electronic structure is solved within the HFS model with core-hole-adapted basis functions calculated with XATOM. To describe ionization dynamics, coupled rate equations are solved with a Monte Carlo approach. To describe fragmentation dynamics, the nuclear motions are propagated classically with molecular forces, rates, and cross-sections calculated on the fly.

References
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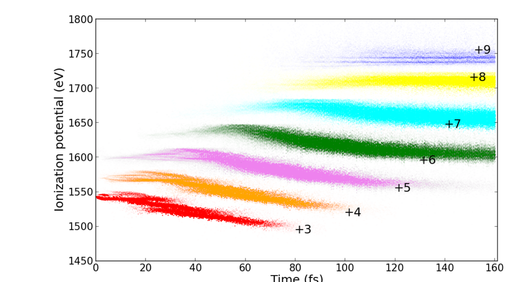
Poster 7: Structural Dynamics in Molecules with X-ray Spectroscopy and Simulations, presented by **Ludger Inhester**

XMDYN+XPOT



Transient ionization potentials for individual charged ions in Al plasmas created by XFEL pulses

Ref. [26]

XMDYN is a computational tool to simulate dynamics of matter exposed to high intensity x-rays (main developer: Zoltan Jurek). Atomic ions and free electrons are treated as classical particles by molecular dynamics, and electron configurations are tracked using a Monte Carlo algorithm. All atomic data are calculated with XATOM. It has been extended to simulate WDM in combination with the supercell approach and periodic boundary conditions, and the plasma environmental effects such as IPD have been incorporated via XMDYN+XPOT.

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Talk in Session 5 (Wed. at 11:20): Plasma environmental effects in the atomic structure for simulating XFEL-heated solid-density matter, presented by **Rui Jin**

Conclusions

> Enabling tools to investigate x-ray multiphoton physics of atoms, molecules, and complex systems exposed to intense XFEL pulses
> XATOM: the key player for x-ray-related tools of XMOLECULE and XMDYN (+XPOT)
> In contrast to frustrated absorption as expected for x-ray multiphoton ionization, the opposite trend is observed when the fluence is extremely high.
> XREMPI shows a broader, red-shifted, asymmetric resonance profile, in contrast to conventional REMPI.
> Theory provides the predictive power for x-ray multiphoton ionization.
> New phenomena to be taken into account for future XFEL applications

Collaboration for XREMPI

Experimental team

Univ. of Conneticut Aaron C. LaForge, Debadarshini Mishra, Stephen Duncanson, Nora Berrah

European XFEL Markus Ilchen, Rebecca Boll, Alberto De Fanis, Michael Meyer, Yevheniy Ovcharenko, Daniel E. Rivas, Philipp Schmidt, Sergey Usenko

Univ. Turku Eemeli Eronen, Edwin Kukk

LCLS, SLAC National Accelerator Lab. Peter Walter

Theory team

CFEL-DESY Theory Division Stanislaw Wirok-Stoletow, Daria Kolbasova, Robin Santra

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DESY – Max Planck Society – University of Hamburg