Quantum-state-resolved ionization dynamics induced by x-ray free-electron laser pulses

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Abstract

Intense **x-ray free-electron laser (XFEL)** pulses can induce multiple sequences of inner-shell ionization events and accompanying decay processes in atoms, producing highly-charged atomic ions. In general, x-ray multiphoton ionization dynamics have been described in terms of time-dependent populations of the electronic configurations visited during the ionization dynamics, neglecting individual state-to-state transition rates and energies. Combining a state-resolved electronic-structure method based on first-order many-body perturbation theory [1] with a Monte Carlo rate-equation method [2] enables us to study state-resolved dynamics based on time-dependent quantum-state populations. Here we present a theoretical study of **state-resolved x-ray multiphoton ionization dynamics** of neon atoms. Our results demonstrate that configuration-based and state-resolved calculations provide similar charge-state distributions, but differences are visible when resonant excitations are involved. Calculated **time-resolved spectra of electrons and photons** allow us to investigate ultrafast dynamics of x-ray multiphoton ionization in detail. In addition, we will present a comparison with a recent experiment on Ne [3] and discuss how to handle the extremely large number of atomic parameters involved in state-resolved dynamics calculations via machine-learning techniques [4].

X-ray multiphoton ionization

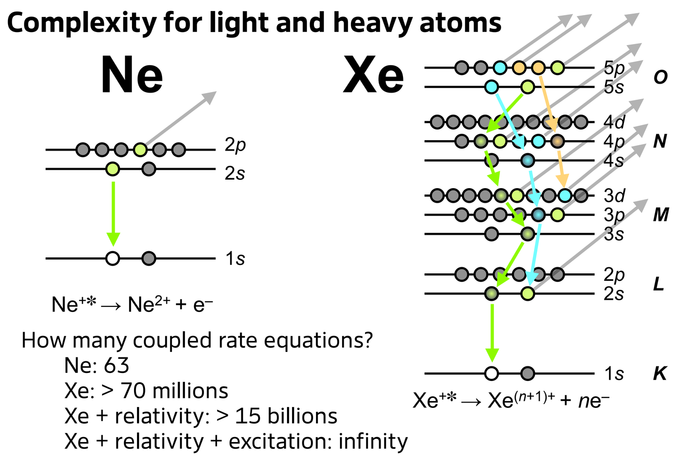
Interaction of matter with intense XFEL pulses is characterized by sequential multiphoton multiple ionization dynamics.

- 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

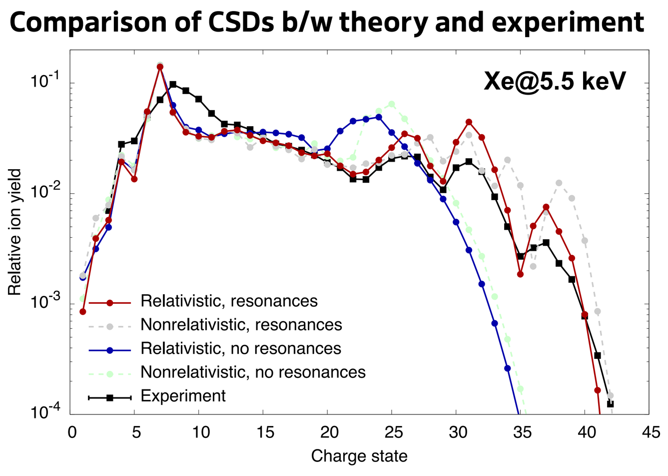
We implement an integrated toolkit, **XATOM** [5], to treat x-ray multiphoton ionization dynamics, based on rate-equation approach, within a consistent theoretical framework of nonrelativistic quantum electrodynamics, perturbation theory, and the Hartree–Fock–Slater model.

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

Complexity for light and heavy atoms



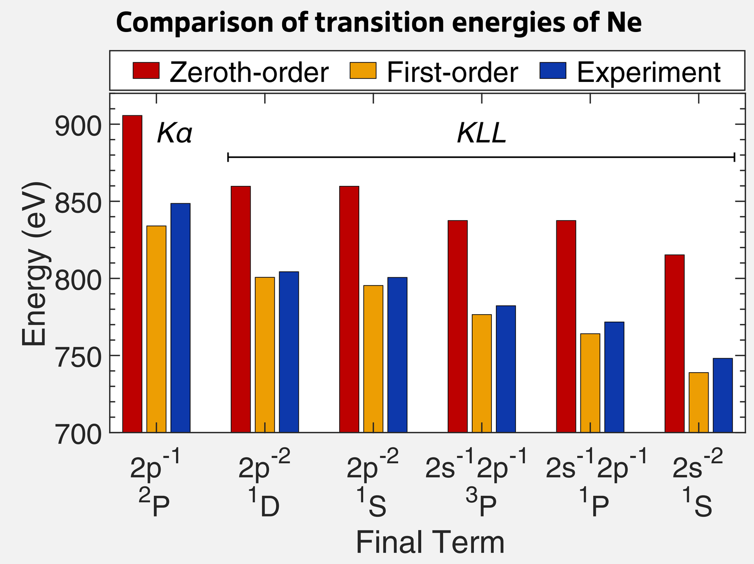
Comparison of CSDs b/w theory and experiment

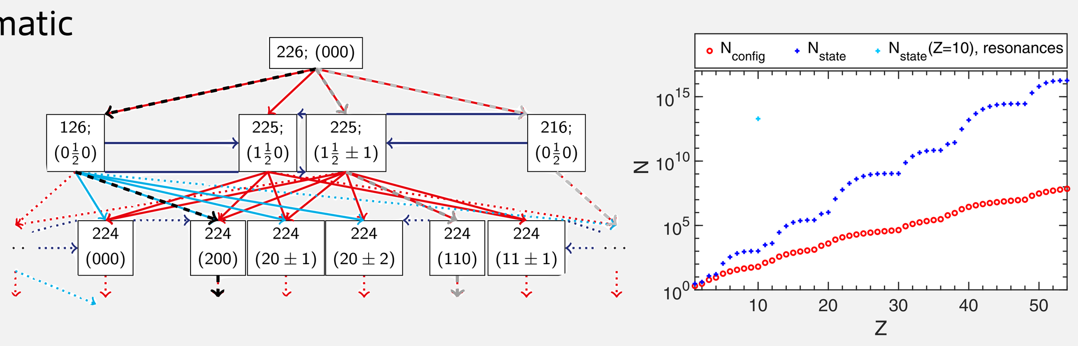


State-resolved XATOM

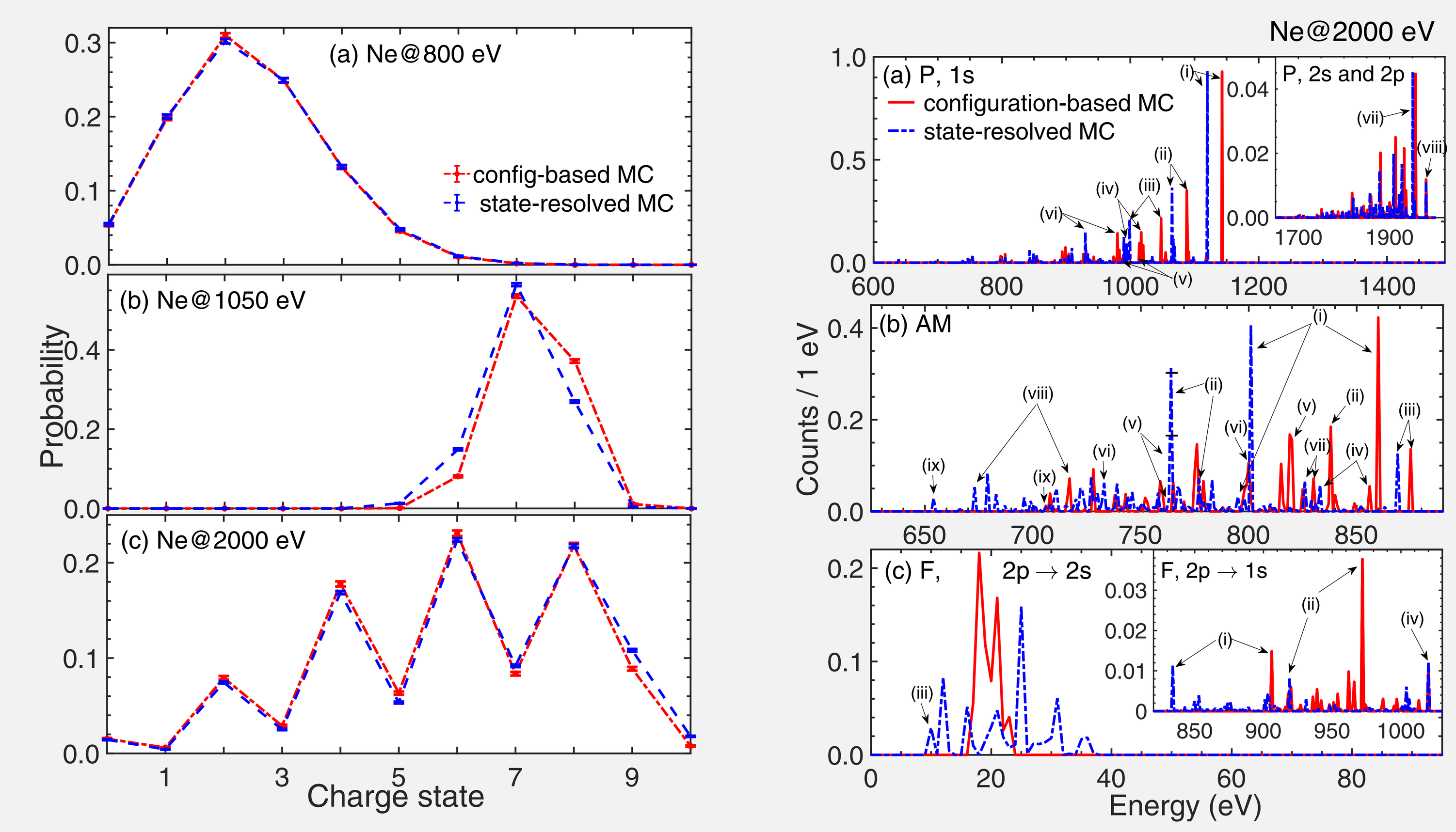
- First-order many-body perturbation theory to improve HFS calculations [1]
- Electronic configuration ($1s^{n_1}2s^{n_2}2p^{n_3}$...) + quantum number (L, S, M_L, κ)
- X-ray ionization dynamics following quantum-state populations, rather than electronic configuration populations
  ➔ N of rate equations explodes
  ➔ Monte Carlo on-the-fly approach
- Almost no difference in charge-state distributions (CSDs), but dramatic improvement on photon and electron spectra

Comparison of transition energies of Ne



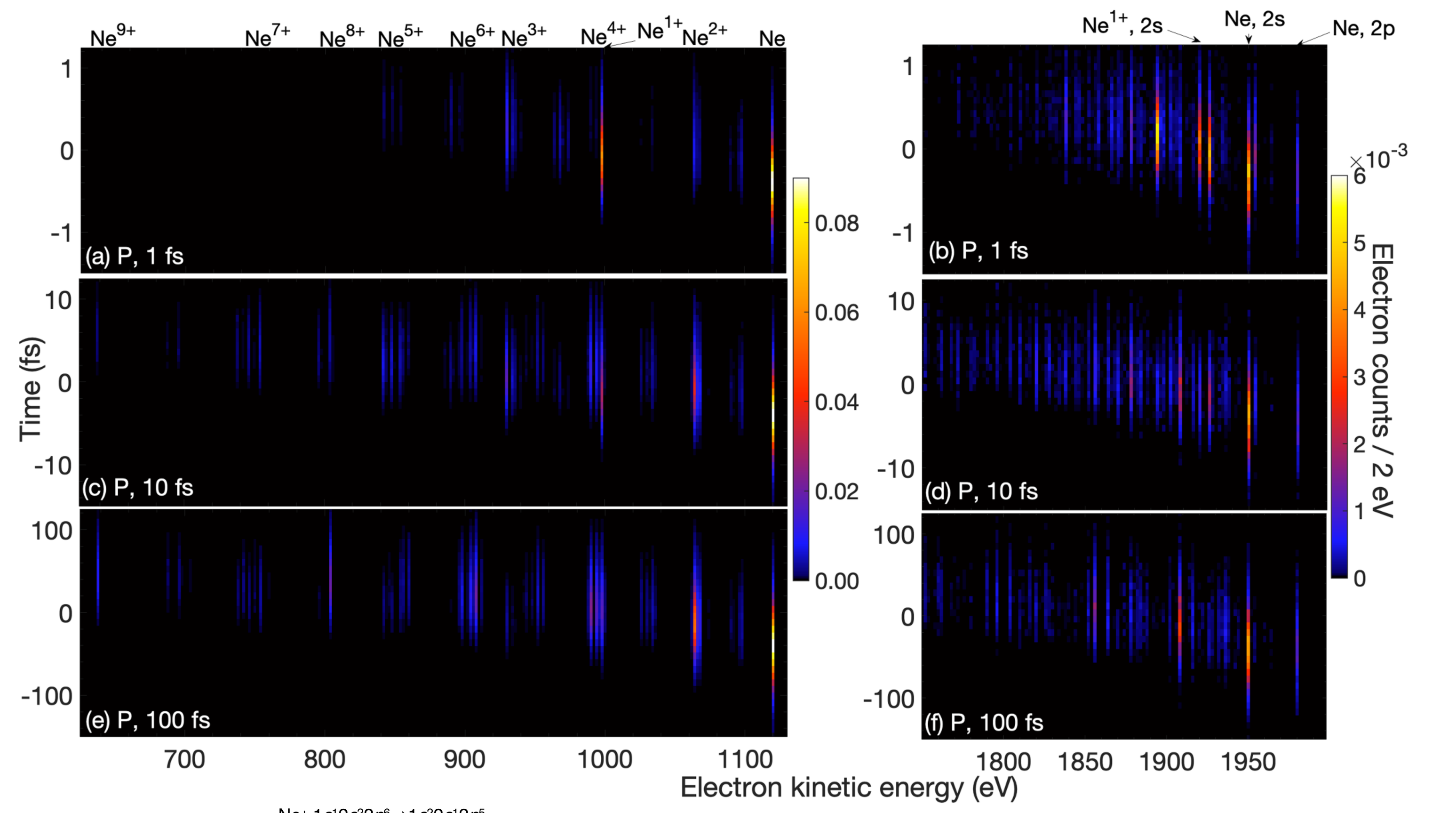


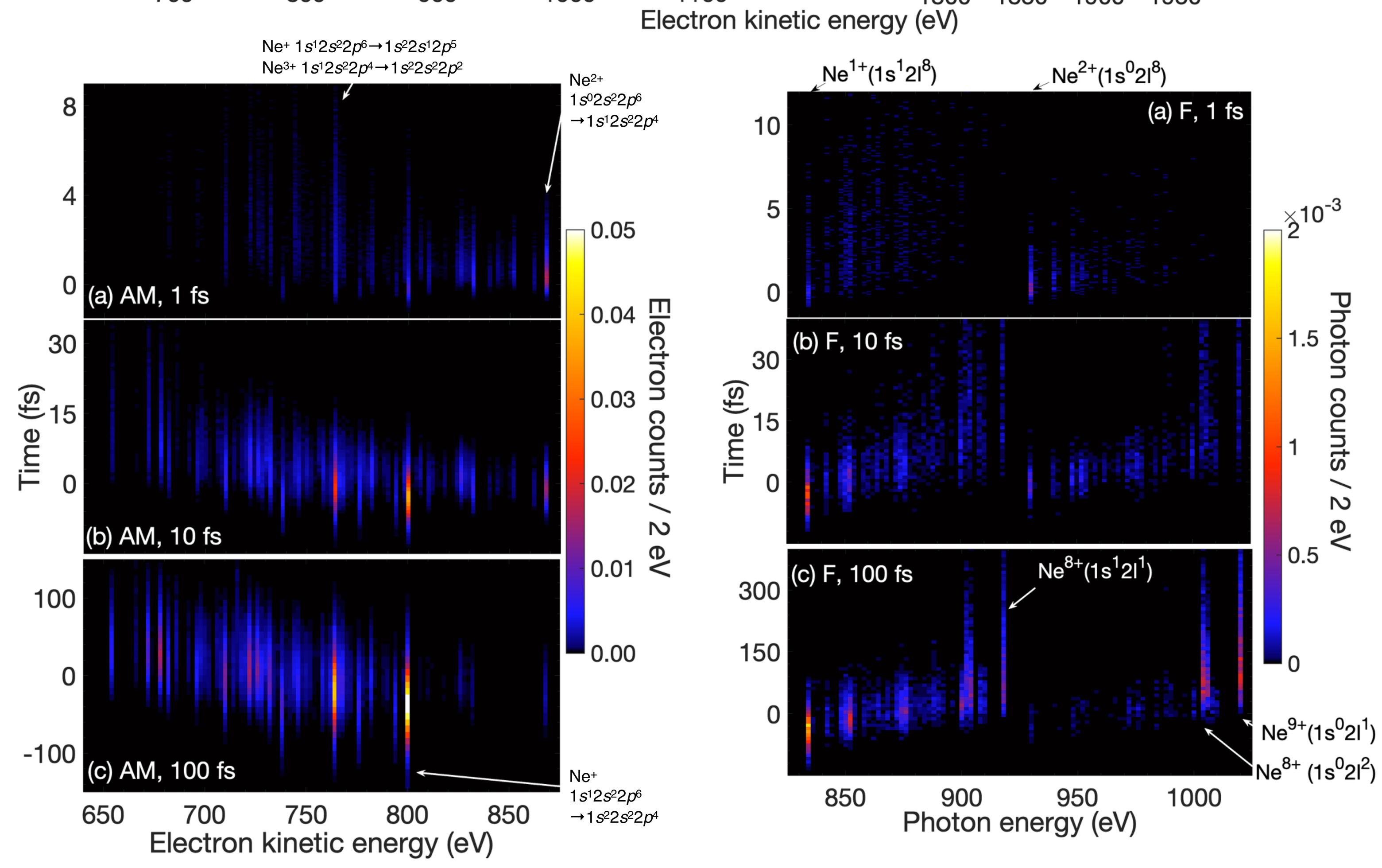
Comparison between config-based XATOM and state-resolved XATOM:
Ne CSDs [left panels] and spectra [right panels] of photoelectron (P), Auger-Meitner electron (AM), and fluorescence (F)



Time-resolved electron and photon spectra of Ne

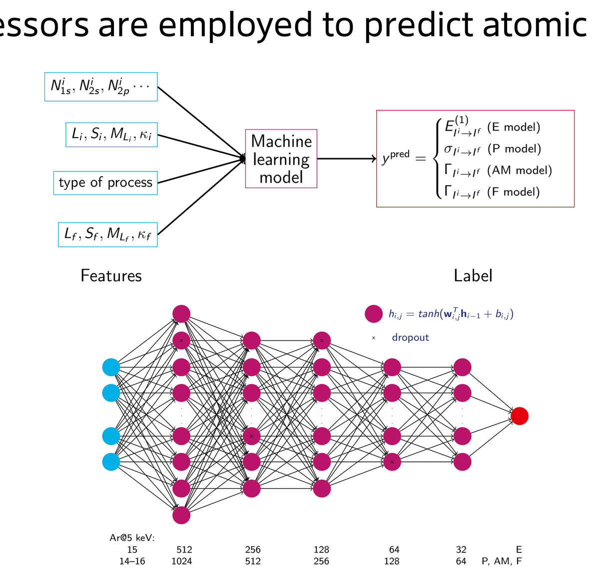
- Frustrated absorption or intensity-induced x-ray transparency [6]
  > the degree of ionization is reduced for shorter pulse duration (higher intensity)
  > 1s photoionization defeats Auger-Meitner (AM) decay as the intensity increases
- Time-resolved photoelectron spectra show more lines of highly charged ions for longer pulses
- In time-resolved AM spectra, AM lines become weaker and take place later for shorter pulses
- In time-resolved fluorescence spectra, SCH and DCH are well separated



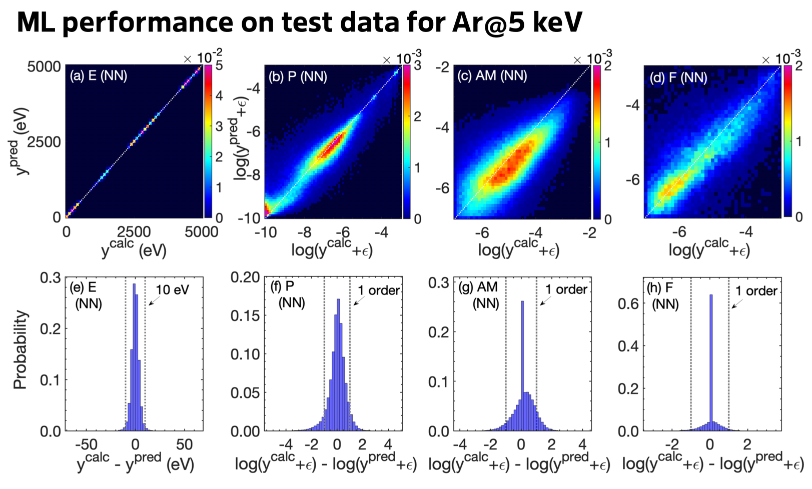


Machine-learning XATOM

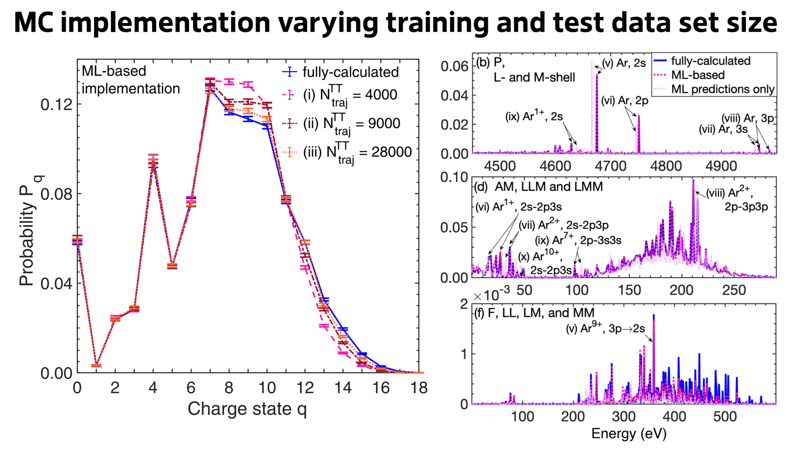
Feedforward neural networks and random forest regressors are employed to predict atomic data [4]



ML performance on test data for Ar@5 keV



MC implementation varying training and test data set size



Conclusions

- XATOM: enabling tool for studying x-ray multiphoton ionization dynamics
- XATOM has been extended to study quantum-state-resolved ionization dynamics
- First-order many-body perturbation theory improves accuracies of transition energies, which are critical for electron and photon spectra
- Calculated time-resolved spectra demonstrate how frustrated absorption manifests itself during intense x-ray pulse
- ML-based state-resolved MC implementation helps to reduce computational cost

References

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Publication

L. Budewig, S.-K. Son & R. Santra, *Phys. Rev. A* **107**, 013102 (2023).

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Center for Free-Electron Laser Science
CFEL is a scientific cooperation of the three organizations:
DESY – Max Planck Society – University of Hamburg

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