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

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Abstract

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)
	- > 1*s* 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 STATE-RESOLVED IONIZATION DYNAMICS OF A NEON … PHYSICAL REVIEW A **107**, 013102 (2023)
- In time-resolved fluorescence spectra, SCH and DCH are well separated

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 rateequation 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-rayinduced dynamics of matter, <https://www.desy.de/~xraypac/>

> Feedforward neural networks and random forest regressors are employed to predict atomic data [4] considered for the 1-fs result. Let's use machine-learning models for predicting ward neural netwo IS die employed to predict

State-resolved XATOM otale reported Articles

- First-order many-body perturbation theory to improve HFS calculations [1] \bullet Eirst-order many-body perturbation ringt of activiting the set of th
The set of the set of
- Electronic configuration (1*s*n12*s*n22*p*n3…) + quantum number (*L*, *S*, *ML*, *κ*) III. **19 II. VALIDATION**
- X-ray ionization dynamics following quantum-state populations, rather than electronic configuration populations ➔ *N* of rate equations explodes → Monte Carlo on-the-fly approach State-resolved approach $\frac{1}{\sqrt{1-\frac{1}{1-\$ \sim λ -ray ionization dynamics following \rightarrow N of rate equations explodes different theoretical strategies for describing physical pro-
- Almost no difference in charge-state distributions (CSDs), but dramatic improvement on photon and electron spectra $\sqrt{126}$ 126; $\frac{1}{2}$ tion in the order side, in the first-order strategy, in the first-order strategy, $\frac{1}{2}$

initial configuration on $\frac{1}{1000}$ (000) $\frac{1}{1000}$ (200) $\frac{1}{1000}$

for both the initial and final configurations avoids is uncertainty with $\mathcal{L}_{\mathcal{A}}$

- XATOM: enabling tool for studying x-ray multiphoton ionization dynamics $VATOM = \epsilon$ λ Al OM. Chabing tool for staaying λ fay
- XATOM has been extended to study quantum-state-resolved ionization dynamics and peak (iii), can also be observed mainly for short pulse $\mathcal{O}(\mathcal{O})$ α quantum-state-resolved ionization
- First-order many-body perturbation theory improves accuracies of transition energies, which are critical for electron and photon spectra generated long after the pulse on time scales up to ∼10 ns. $\frac{1}{2}$ finally, we turn to the fluorescence spectra for inner-shell $\frac{1}{2}$ for $\frac{1}{2}$ relation via 2*p triansition* via 2*p* transition as shown in Fig. 10. We do the s
- Calculated time-resolved spectra demonstrate how frustrated absorption manifests itself during intense x-ray pulse prioton speetra
Folculated time received cnectra Calculated time-resolved spectra, even though to the lower intence with a unit comparison with a maniform $\frac{1}{2}$ riamics is relation of military charged in the military consequence takes the interest of the set of the inter
- ML-based state-resolved MC implementation helps to reduce computational cost hole spectra for Ne ions are well separated and ordered by computational cost the charge state state state, the photon state state state state \sim

Intense **x-ray free-electron laser (XFEL)** pulses can induce multiple sequences of innershell ionization events and accompanying decay processes in atoms, producing highlycharged 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 stateresolved 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].

> L. Budewig, S.-K. Son & R. Santra, *Phys. Rev. A* **107**, 013102 (2023). or ≈40% [case (iii)] of initial states all possible all possible all possible all possible all possible all po
Initial states all possible all atomic transition parameters are calculated in the training and T oill T illustrate this point, in F ill T

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Machine-learning XATOM durie-rediting AATOM thing loopning VATAM 1012 photons*/µ*m2 is used.

Publication

the 1*s* shell and, thus, further inner-shell photoabsorption, **ionization buddewig et al. Physical Review Research atoms, like argonization dynamics framework to heavier atoms, like argonization and the second contract to heavier atoms, like argument to the second contract of the sec IIIT ML performance on test data for Ar@5 keV**

Conclusions as shown in Figs. 8(c) and 8(e), which is consistent with the involved initial configurations. A fluence of 1012 photons*/µ*m2 is used.

References effect was observed in $\mathcal{L}_{\mathcal{F}}$ experiments on warm dense $\mathcal{E}_{\mathcal{F}}$ aluminum [67].) The 1*s*–2*p* fluorescence energy is given by

[1] L. Budewig, S.-K. Son, and R. Santra, *Phys. Rev. A* **105**, 033111 (2022). [2] S.-K. Son and R. Santra, *Phys. Rev. A* **85**, 063415 (2012). [3] S.-K. Son, T. Baumann, J.-E. Rubensson, *et al.*, in preparation. [4] L. Budewig, S.-K. Son, Z. Jurek, M. M. Abdullah, M. Tropmann-Frick, and R. Santra, *Phys. Rev. Res.* **6**, 013265 (2024). [5] Z. Jurek, S.-K. Son, B. Ziaja, and R. Santra, *J. Appl. Cryst.* **49**, 1048 (2016). [6] M. Hoener *et al.*, *Phys. Rev. Lett.* **104**, 253002 (2010); L. Young *et al.*, *Nature* **466**, 56 (2010). **I** double 2011 **A doe** 077111 (2022) rilys. Rev. A **109**, 033111 (2022).

Figure 3 shows (a) photoelectron and (b) fluorescence

are explained in Tables III in the Appendix. The Appendix is the Appendix. The Appendix. The Appendix. The Appendix.

$9970M$ and state vecessed VA Comparison between config-based XATOM and state-resolved XATOM: As a next example, we examine photoionization of a

 $(0\frac{1}{2}0)$

(110) (11 *±* 1)

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e CSDs [left panels] and spectra [right panels] of photoelectron (P), Auger-Meitner electron (AM), and fluoresce ectron (AM), and fluorescence (F) $\,$ configurations are given by the Aufbau principle. For the resonant Ne CSDs [left panels] and spectra [right panels] of photoelectron (P), Auger-Meitner electron (AM), and fluorescence (F) STATE-RESOLVED IONIZATION DYNAMICS OF A NEON … PHYSICAL REVIEW A **107**, 013102 (2023) the original version of the correction of the first-order of the first-order of the first-order of \sim neutral argon atom (1*s*22*s*22*p*63*s*23*p*6) in the region of the fon (P) , Auger-Meither electron (AM), and huorescence (P)

10 20 30 40 50 Z

state for all *j* (no

⎦*.* (10)

excitations up to *n*max = 7 and *l*max = 2 are considered and an energy bandwidth of 1% is assumed. The error bar indicates the statistical control indicates the statistical control of error. between experiment and theory still remaining for the firstorder calculation might be attributed to the use of the same set of initial and final orbitals, the neglect of higher-order terms, and relativistic effects. We remark that no value is shown for

 \mathbf{S} . Similar to \mathbf{S}

 $\frac{1}{2}$ and 9(b) $\frac{1}{2}$ and 9(b)], Table VIII $\frac{1}{2}$ and 9(d)], Table VIII $\frac{1}{2}$

(iii)

(viii)

Other parameters are the same as used in Fig. 2. The peak labels are

 $10⁰$

Auger-Meitner electron and the x-ray fluorescence spectra

and Table IX [for Figs. 9(e) and 9(f)].

The error bars indicate the statistical error bars indicate the σ

energies predicted by the neural network [case (i)], *E*pred

Energy (eV)

B. Photoionization cross sections for argon