Stabilization and entanglement of XUV-dressed atoms

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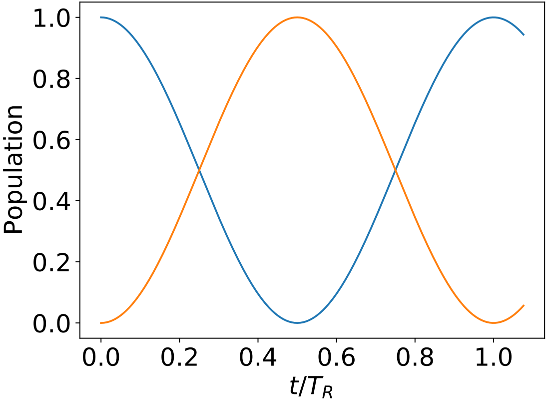
Rabi oscillations

- Two-level system driven close to resonance by external field: $\Delta\omega$
- Populations oscillate with the generalized Rabi frequency: $W = \sqrt{\Omega^2 + \Delta\omega^2}$
- Rabi frequency: $\Omega = eE_0 z_{ba}/\hbar$

$$a(t) = \left[\cos\frac{Wt}{2} - i\frac{\Delta\omega}{W}\sin\frac{Wt}{2}\right] e^{i\Delta\omega t/2}$$

$$b(t) = -i\frac{\Omega}{W}\sin\frac{Wt}{2}\, e^{-i\Delta\omega t/2}$$



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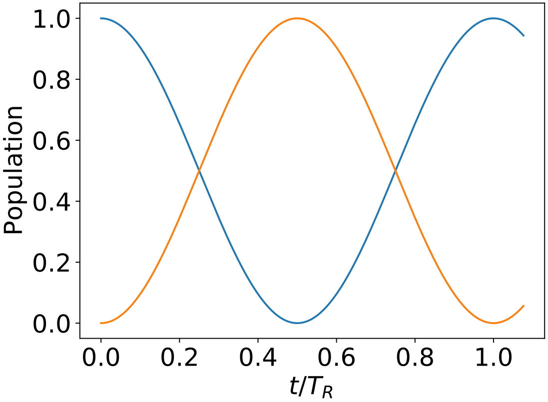
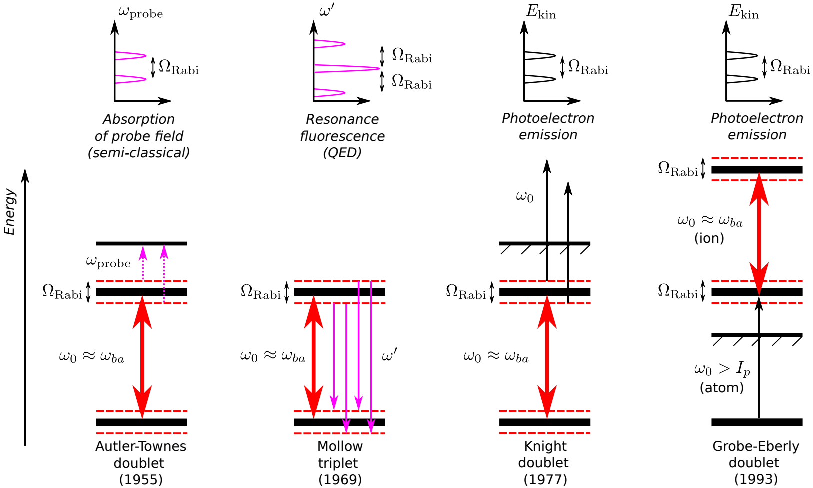
$$b(t) = -\frac{\Omega}{2W}\left({\color{red}e^{iWt/2}} - {\color{blue}e^{-iWt/2}} \right) e^{-i\Delta\omega t/2}$$

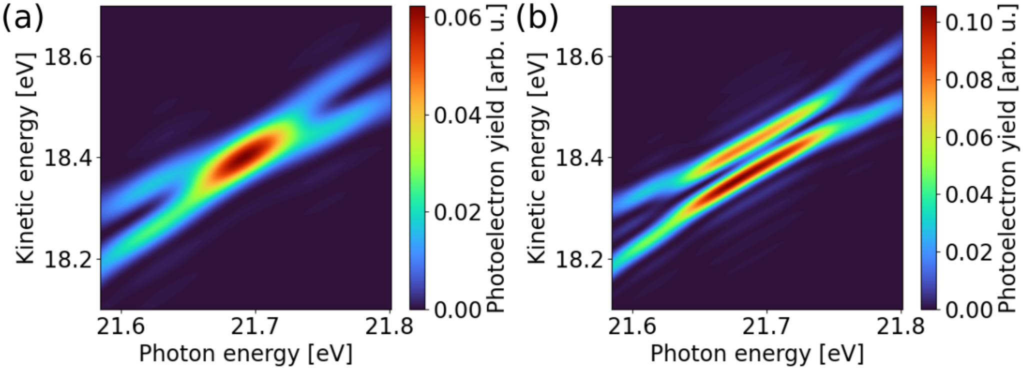


Physical phenomena from Rabi cycling atoms



Ultrafast Knight doublet

Low-intensity Rabi-cycling regime → symmetric (AT-like) doublet



E Olofsson and J M Dahlström *Phys. Rev. Research* **5**, 043017 (2023)

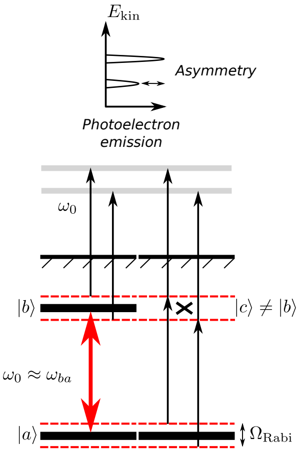
Earlier work with smooth evelopes: Roguś and Lewenstein *J. Phys. B: At. Mol. Phys.* **19** 3051 (1986)

Experiments using Free-Electron Laser

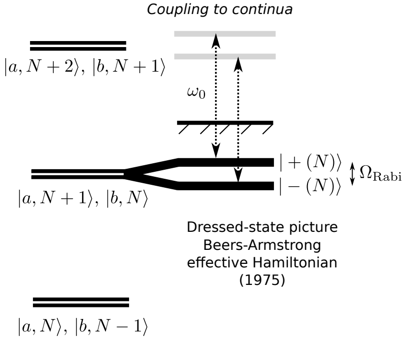
Low Density Matter beamline at FERMI

- Seeded Free-Electron Laser
- Ultra-short coherent pulses
- High intensity ($\sim 10^{13}\,\mathrm{W/cm^2}$)
- Short wavelength (XUV)
- Tuned to resonance between He $1s^2$ and $1s4p$ (23.7 eV)
- Experimentalist S. Nandi, CNRS researcher in Lyon.

More intense photoionization from Rabi cycling atoms



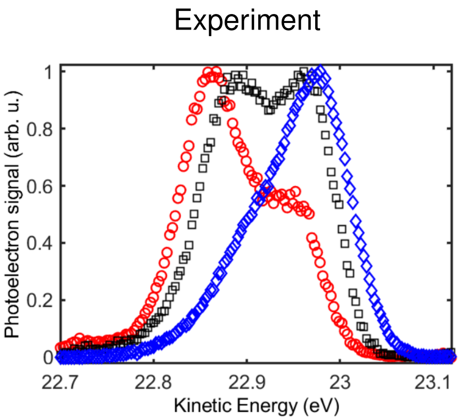
Asymmertric doublet
TDSE: LaGattuta (1993)
Seeded XUV-FEL experiment:
Nandi *et al.* (2022)

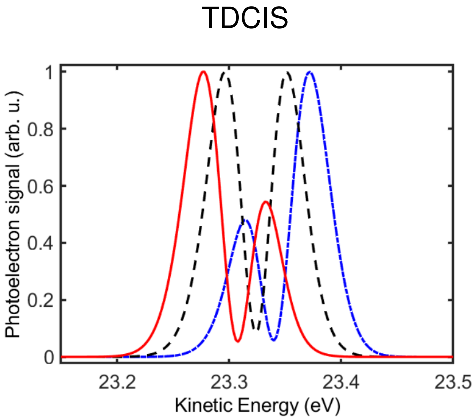


Dressed-state picture
Beers-Armstrong
effective Hamiltonian
(1975)

Quantum optics picture
Jaynes-Cummings (RWA)
(1963)

Experimental and *ab initio* results

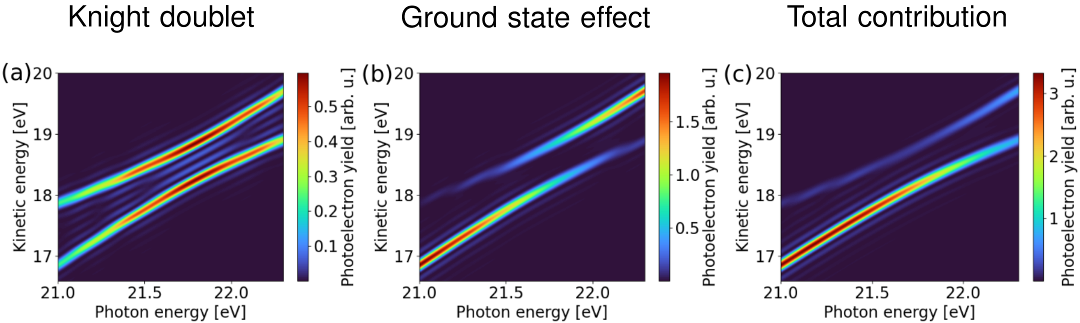




$I \approx 2 \times 10^{13}$ W/cm^2, FWHM $\approx$ 56 fs (Gaussian)

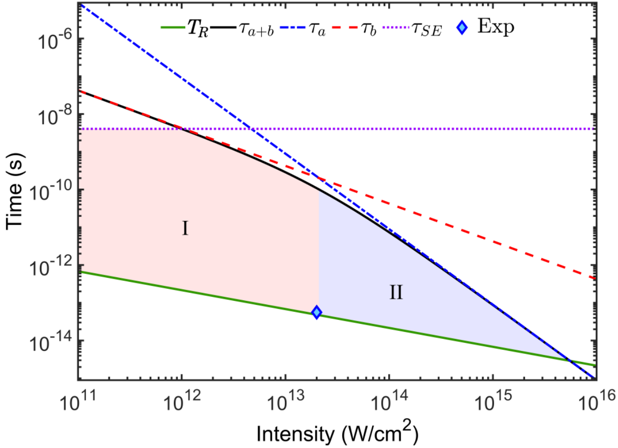
S Nandi, E Olofsson, M Bertolino, ... J M Dahlström *Nature* **608**, 488 (2022)

Quantum path interference (He: $1s^2 - 1s2p$)



E Olofsson and J M Dahlström *Phys. Rev. Research* **5**, 043017 (2023)

Photoionization domains: He $1s^2 - 1s4p$



S Nandi, E Olofsson, M Bertolino, ... J M Dahlström *Nature* **608**, 488 (2022)

Time scales:

- $T_R \propto 1/\sqrt{I}$
- $\tau_b \propto 1/I$
- $\tau_a \propto 1/I^2$

Experiment:

- $\tau_a \approx \tau_b$
- Ionization: $P \leq 0.1\%$

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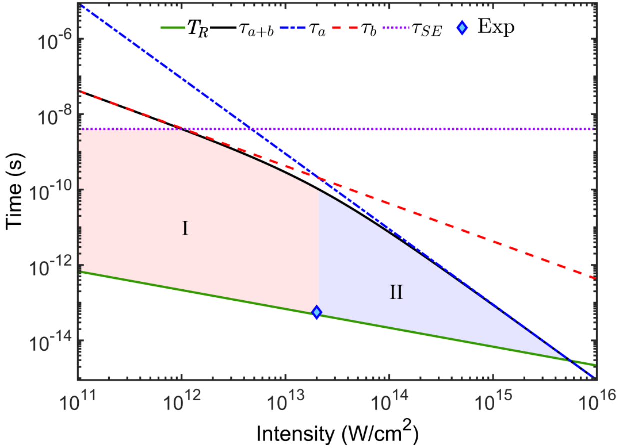

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What happens for non-perturbative photoionization?

PHYSICAL REVIEW A VOLUME 12, NUMBER 6 DECEMBER 1975

Exact solution of a realistic model for two-photon ionization*

Brian Lee Beers and Lloyd Armstrong, Jr.
Department of Physics, The Johns Hopkins University, Baltimore, Maryland 21218
(Received 30 June 1975)

A realistic model for resonant two-photon ionization of an atom is developed which can be solved exactly to give the ionization probability as a function of time, photon intensity, and a few atomic parameters. The exact solutions are compared to various commonly used perturbation results in order to ascertain the regions of validity of the perturbation calculations. It is shown that there are combinations of the parameters for which ionization rates cannot accurately describe the correct results, and that one commonly used perturbation development is not generally applicable to resonant two-photon ionization. Extension of the model to $(n+m)$-photon ionization having an m-photon resonance is discussed. The model is applied to two- and three-photon ionization in Cs.

- Main points:
  - Effective (non-hermitian) two-level Hamiltonian
  - Photoionization: $P(t) = 1 - |U_{aa}(t)|^2 - |U_{ba}(t)|^2$
  - Different domains of photoionization: T_R, τ_a and τ_b
  - Stabilization regime: $P(t \to \infty) = 50\%$ for $\tau_a = \tau_b$

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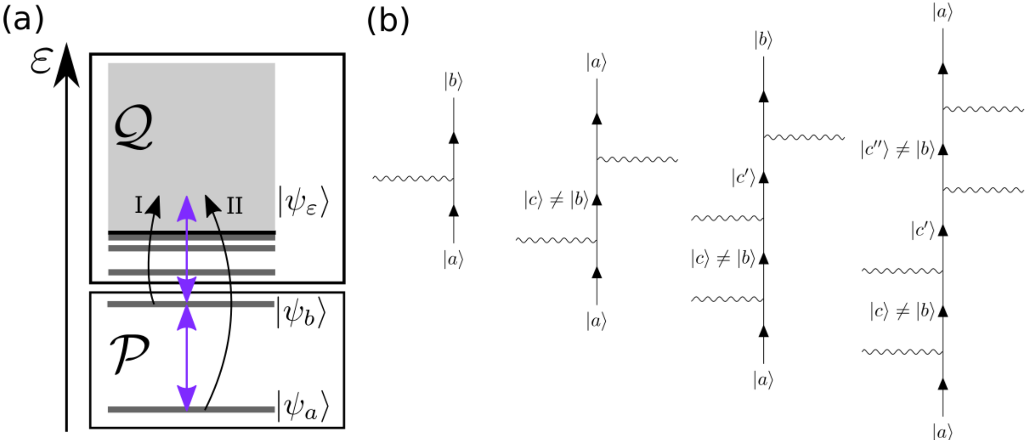
Beyond perturbation theory: *Resolvent operator technique*



Analytical theory with field effects up to fourth order.

E Olofsson and J M Dahlström *Phys. Rev. Research* **5**, 043017 (2023)

Analytical expressions for photoionization of Rabi cycling atom

Contribution from the two dressed-states, $|\pm\rangle$:

$$U^{(1)}_{\epsilon a\pm}(t) = \mp i\mathcal{E}_0 z_{\epsilon b}\frac{h_{ab}}{W}\exp[-i\frac{t}{2}(E_\epsilon+\lambda_\pm)]\frac{\sin[\frac{t}{2}(E_\epsilon-\lambda_\pm)]}{(E_\epsilon-\lambda_\pm)},$$

$$U^{(2)}_{\epsilon a\pm}(t) = \mp i\mathcal{E}_0^2 z_{\epsilon\neq b}\frac{(\lambda_\pm-h_{bb})}{2W}\exp[-i\frac{t}{2}(E_\epsilon+\lambda_\pm)]\frac{\sin[\frac{t}{2}(E_\epsilon-\lambda_\pm)]}{(E_\epsilon-\lambda_\pm)},$$

where total first- and second-order amplitudes are: $U^{(n)}_{\epsilon a}(t) = U^{(n)}_{\epsilon a+}(t) + U^{(n)}_{\epsilon a-}(t)$, short-hand energy: $E_\epsilon - \lambda_\pm = \epsilon - 2\omega - \lambda_\pm$ and transition elements:

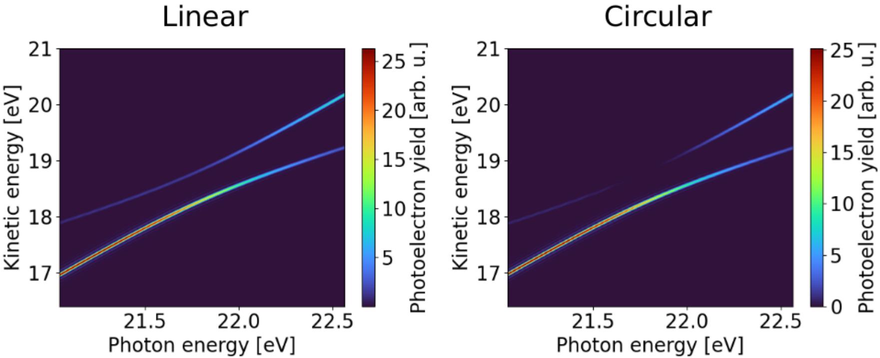
$$z_{\epsilon b} = \langle\psi_\epsilon|\, z\,|\psi_b\rangle$$

$$z_{\epsilon\neq b} = \sum_{c\neq b}\frac{\langle\psi_\epsilon|\, z\,|\psi_c\rangle\langle\psi_c|\, z\,|\psi_a\rangle}{E_a-E_c}.$$

E Olofsson and J M Dahlström *Phys. Rev. Research* **5**, 043017 (2023)

Photoelectron signature of stablization (He $1s^2 - 1s2p$)

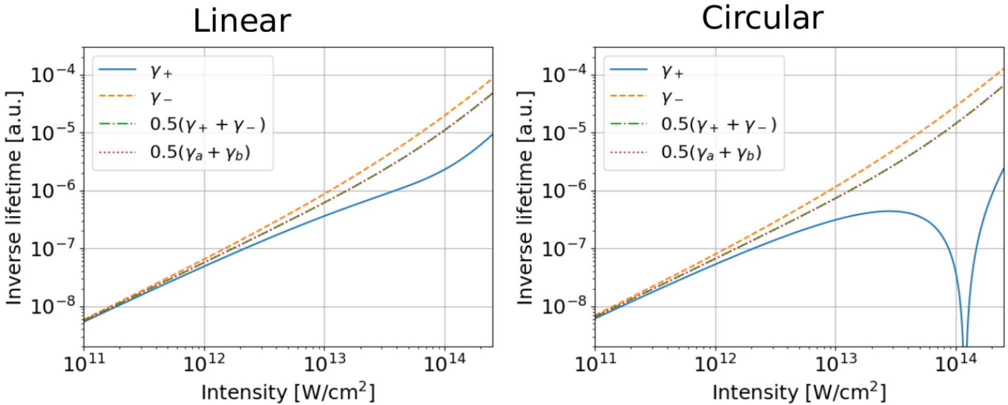
10 cycles



Prediction: Single photoelectron peak for circular polarization ($\tau_a = \tau_b$).

E Olofsson and J M Dahlström *Phys. Rev. Research* **5**, 043017 (2023)

Dressed-atom inverse lifetimes: γ_+ and γ_-



Conclusion: Infinite lifetime of dressed state $(+)$ for circular polarization.

E Olofsson and J M Dahlström *Phys. Rev. Research* **5**, 043017 (2023)

Related topic: Adiabatic passage to the continuum: Controlling ionization with chirped laser pulses
U. Saalmann, S. K. Giri, and J. M. Rost, *Phys. Rev. Lett.* **121**, 153203 (2018).

Grobe-Eberly doublet

PHYSICAL REVIEW A VOLUME 48, NUMBER 1 JULY 1993

Observation of coherence transfer by electron-electron correlation

R. Grobe and J. H. Eberly
Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627
(Received 14 December 1992)

We present the results of exact wave-function calculations that are equivalent to numerical experiments on a two-electron quantum system. We show that Rabi oscillations of the core electron can modify the photoelectron spectrum of the outer electron by efficient coherence transfer via *e-e* correlation, even in the absence of an autoionizing level or other continuum structure. We compare a simplified analytically soluble two-electron theory with our numerical results.

PACS number(s): 32.80.Rm, 32.80.Fb, 32.80.Wr

- Main points:
  - Different time-order compared to Knight doublet processes
  - 1. Photoelectron propagates freely: $|g\rangle \to |E^{\mathrm{kin}}\rangle$
  - 2. Photoion is dressed by laser field: $|a\rangle \leftrightarrow |b\rangle$
  - 3. A "doublet" in the photoelectron distribution appears.

Prior experiment: Observation of continuum-continuum Autler-Townes Splitting.
Walker, Kaluza, Sheehy, Agostini, and DiMauro *Phys. Rev. Lett.* **75**, 633 (1995)

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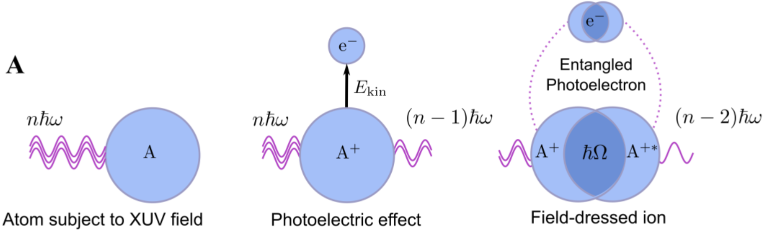
Schematic for Grobe-Eberly doublet formation



Interpretation: Time-dependent Von Neumann entropy of entanglement:

$$S_{\mathrm{vN}}(t) = -\,\mathrm{Tr}[\rho_{\mathcal{P}}(t)\log_2 \rho_{\mathcal{P}}(t)] = -\,\mathrm{Tr}[\rho_{\mathcal{I}}(t)\log_2 \rho_{\mathcal{I}}(t)]$$

Generation of entanglement using a short-wavelength seeded free-electron laser
Saikat Nandi, Axel Stenquist, Asimina Papoulia, ... J M Dahlström arXiv:2312.04442 [quant-ph] (2023)

Textbook on entanglement: Exploring the Quantum: Atoms, Cavities, and Photons
Serge Haroche and Jean-Michel Raimond (Oxford Graduate Texts) 1st Edition (2006)

Relation to attosecond chemistry: Quantum coherence in molecular photoionization
Ruberti, Patchkovskii and Averbukh Phys. *Chem. Chem. Phys.* **24**, 19673 (2022)

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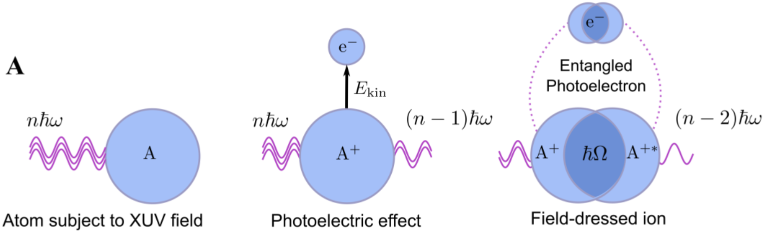

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Theory for the Grobe-Eberly formation

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$$|\Psi(t)\rangle = g(t)e^{-\mathrm{i}\tilde{\epsilon}_g t}\,|g, N\rangle + \frac{\Omega^{ag}}{2}\int_0^t dt' \int dE^{\mathrm{kin}}\, e^{-\mathrm{i}\tilde{\epsilon}_a(t-t')}g(t')e^{-\mathrm{i}\tilde{\epsilon}_g t'}$$

$$\times\,|a, N-1\rangle \otimes |E^{\mathrm{kin}}\rangle.$$

The first term describes the state of the He atom, which is ionized by the FEL field.
The second term describes transition to (and dynamics in) the bipartite system.
For flat-top fields, a and b are Rabi amplitudes, and g is exponential damping (one time index).

Related theory works with envelope: Photoemission spectroscopy with high-intensity short-wavelength lasers, Zhang and Rohringer *PRA* **89**, 013407 (2014)
Core-resonant ionization of helium by intense XUV pulses: Analytical and numerical studies ...Yu and Madsen *PRA* **98**, 033404 (2018)

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$$\times \Big(a(t-t')\,|a, N-1\rangle + b(t-t')e^{-\mathrm{i}(\tilde{\epsilon}_b-\tilde{\epsilon}_a)(t-t')}\,|b, N-2\rangle\Big) \otimes |E^{\mathrm{kin}}\rangle.$$

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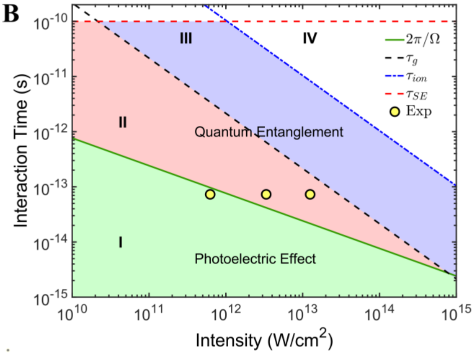
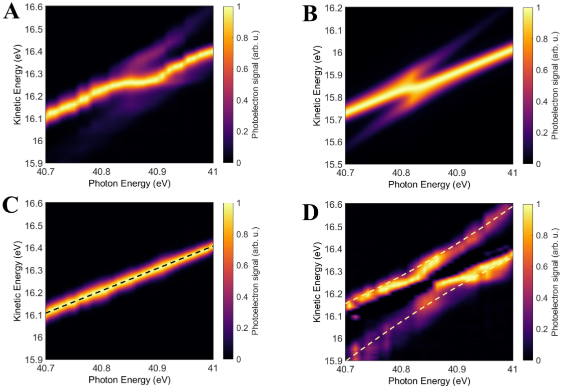
Parameter space for generation of quantum entanglement



Crossing the ionic-Rabi border: Seeded XUV-FEL experiments at FERMI.

Generation of entanglement using a short-wavelength seeded free-electron laser
Saikat Nandi, Axel Stenquist, Asimina Papoulia, ... J M Dahlström (2023) arXiv:2312.04442 [quant-ph]

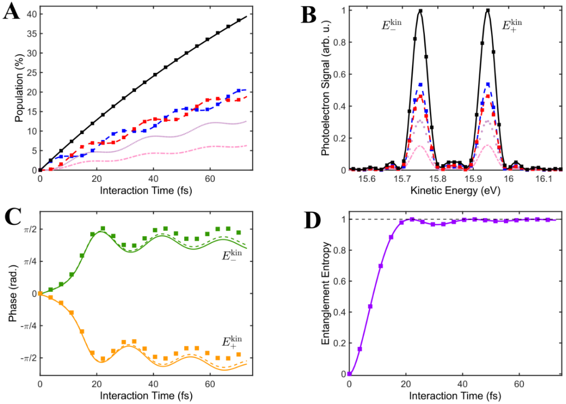
Experimental analysis



Interpretation: Entangled and non-entangled photoelectrons

A Deconvoluted experimental data (1.25×10^{13} W/cm^2). **B** Macroscopic average with entangled and non-entangled contributions (theory). **C** Non-entangled experimental contribution (Einstein's photoelectric effect). **D** Entangled experimental contribution (Grobe-Eberly doublet).

Interpretation: Generation of entanglement (single atom)



A Ionic populations over interaction time. **B** Ion-resolved photoelectron spectra.
C Relative phases of ionic channels. **D** Entropy of entanglement, $S_{\mathrm{vN}}(t) = -Tr[\rho_i \log_2(\rho_i)] \rightarrow 1,\, t > T_{\mathrm{R}}$.

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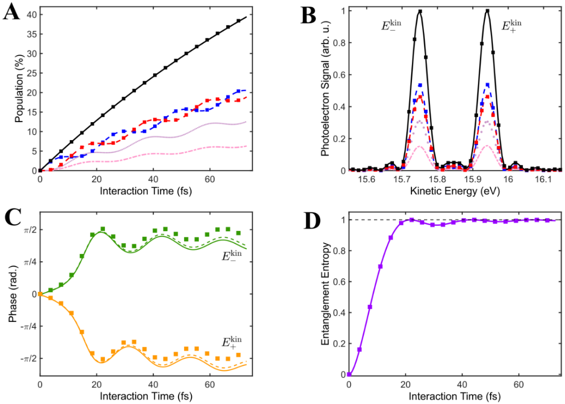

$$|\Psi^{(\mathcal{F})}\rangle \sim \frac{1}{2}[(|a, N-1\rangle + i|b, N-2\rangle) \otimes |E_-^{\rm kin}\rangle + (|a, N-1\rangle - i|b, N-2\rangle) \otimes |E_+^{\rm kin}\rangle]$$

Summary

- Photoionization mechanisms of Rabi cycling atoms driven by XUV-FEL fields.
- **Interpretation:** Asymmetry of doublet due to interference between I and II
  → blue shift of the symmetric (AT-like) doublet[1].
- **Prediction:** Stabilization of Rabi-cycling helium atoms with circular FEL pulses
  → a single photoelectron peak from Rabi cycling atom[2].
- **Interpretation:** Novel bipartite system: photoelectron and light-dressed ion
  → one Rabi period for full quantum entanglement (Grobe-Eberly doublet)[3].

[1] Nandi, E Olofsson, M Bertolino, ... J M Dahlström *Nature* **608**, 488 (2022)
[2] E Olofsson and J M Dahlström *Phys. Rev. Research* **5**, 043017 (2023)
[3] Saikat Nandi, Axel Stenquist, Asimina Papoulia, ... J M Dahlström (2023) arXiv:2312.04442 [quant-ph] → Sci. Adv. (2024)

Outlook

- **Complex systems:** from atoms to molecules[1]
- **Multiple pulses:** pump-probe with light-dressing from IR to XUV[2]
- **Structured light:** polarization control of ionization[3]
- **Coherent control:** envelope control of entanglement[4]
- **Non-classical light:** from quantum revivals to strong-field effects[5]

[1] Palacios, Bachau and Martín Phys. Rev. A **74**, 031402(R) (2006)
[2] Wollenhaupt *et al. Phys. Rev. A* **68**, 015401(R) (2003)
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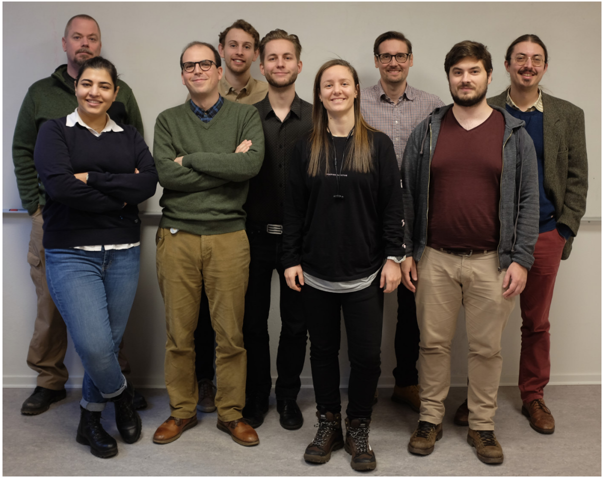
Team



Theoretical light-matter dynamics group

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Thank you for your attention!

Questions?

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