Strong-Field Ionization of Rare Gases in Intense Bicircular Laser Pulses

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Abstract

The single and double ionization of rare gas atoms by intense bicircular laser fields is studied utilizing a classical ensemble computational model. Fundamental similarities emerge across all of the atomic species, indicating the underlying universality of the rescattering process. But distinct differences emerge as well, showing how the laser-atom interaction can be tuned by varying the bicircular field parameters and the atomic species. We will present the latest results from our "computational experiments" and consider avenues for future studies.

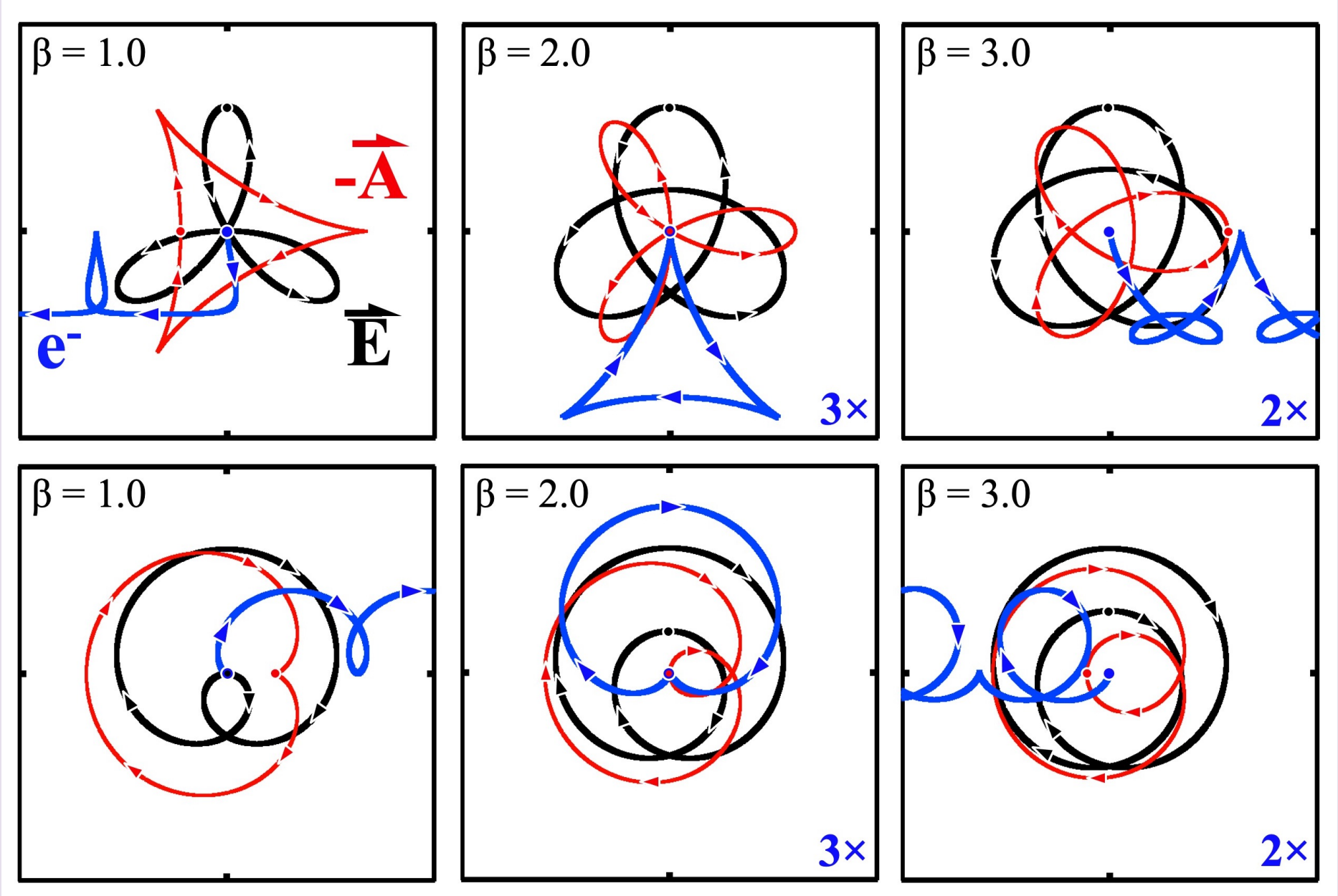
Classical Ensemble

In this study, a completely classical model atom [1-4] is used, and established softening parameters for the atomic potential are employed for the various atomic species [5]. A realistic, three-dimensional laser pulse (τ=10 fs, ω_0=10 μm) [6] interacts with the model atoms for large ensemble sizes to generate ionization yield curves, electron momentum distributions, and rescattering electron trajectories. Throughout this work, "normalized intensity" is used, where the peak electric field amplitude is fixed, allowing for direct comparison between the different field conditions [3-4].

[1] Panfili R, Haan S L and Eberly J H 2002 Phys. Rev. Lett. 89 113001
[2] Ho P, Panfili R, Haan S L and Eberly J H 2005 Phys. Rev. Lett. 94 093002
[3] Chaloupka J L and Hickstein D D 2016 Phys. Rev. Lett. 116 143005
[4] Chaloupka J L 2020 J. Phys. B: Atom. Mol. and Opt. Phys. 53 18 185601
[5] Wang X 2013 PhD Thesis University of Rochester
[6] Paquette J P and Chaloupka J L 2009 Phys. Rev. A 79 043410
[7] Eichmann H, Egbert A, Nolte S, Momma C, Wellegehausen B, Becker W, Long S and McIver J K 1995 Phys. Rev. A 51 R3414
[8] Milosevic D B, Becker W and Kopold R 2000 Phys. Rev. A 61 063403
[9] Fleischer A, Kfir O, Diskin T, Sidorenko P and Cohen O 2014 Nat. Photon. 8 543
[10] Kfir O et al 2015 Nat. Photon. 9 99
[11] Mancuso C A et al 2016 Phys. Rev. A 93 053406
[12] Linden v d, Heuvell H B v and Muller H G 1988 Multi-Photon Processes Studies in Modern Optics vol 8 (Cambridge) p 25
[13] Gillen G D, Walker M A and Van Woerkom L D 2001 Phys. Rev. A 64 043413

Bicircular Fields

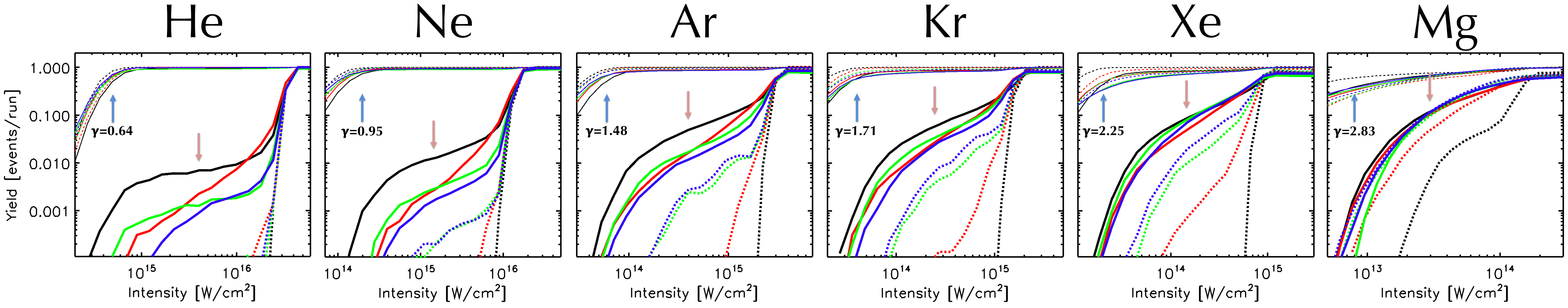
Intense bicircular fields [7-11] are generated through the linear combination of two laser pulses of circularly polarized light, typically at the fundamental and second harmonic frequencies. In this study, 400nm:800nm fields are used in both counter-rotating and corotating superpositions, and the 2ω:ω amplitude ratio (β) is varied to generate different field shapes. Electric field patterns are shown for counter-rotating (upper panels) and corotating (lower panels) fields. The negative vector potential shows the electron momentum distributions expected from the "simpleman" model [12], where the electron is released with zero initial velocity and the effect of the Coulomb potential is ignored. A simple electron trajectory is shown for release at the peak of the field (counter-rotating, upper panels), showing a perfect returning trajectory for a 2:1 amplitude ratio. For corotating fields (lower panels), the electron must be released at the null of the field in order to be driven back towards the parent ion. As a result, rescattering is much more effective with counter-rotating fields than with corotating pulses. However, the effect of the Coulomb potential can help improve corotating rescattering by 1) effectively delaying the release of the electron into the laser field and 2) significantly distorting electron trajectories.



Keldysh Parameter

The Keldysh parameter $\gamma=(I_p/2U_p)^{1/2}$ is typically used to describe tunneling vs multiphoton behavior (small vs large γ). Here we also use it to indicate the relative strength of the Coulomb potential. For small γ, we can expect Coulomb effects to be small, giving rise to behavior closest to that predicted by the "simpleman" model, while for large γ, the effect of the Coulomb potential will become more significant. But at the same time, the two active electrons in heavier atoms are more weakly bound compared to their mutual repulsion. These effects act together to enhance the efficiency of double ionization for large γ, even reproducing the NSDI of Mg with circular polarization [13].

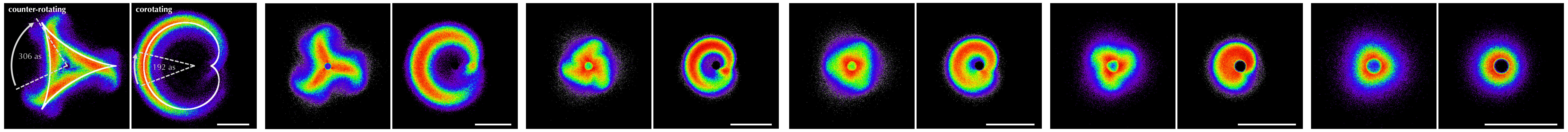
ionization yields



- 800nm linear
- 1:1 counter
- 2:1 counter
- 3:1 counter
- 800nm circular
- 1:1 co
- 2:1 co
- 3:1 co

each graph is the result of
1.6×10^7 simulation runs
(8 curves, 20 intensities,
10^5 runs per intensity)

SI electron momenta



transverse electron momenta from single ionization

counter-rotating (left panel)
corotating (right panel)

1:1 400nm:800nm
intensity at blue arrow
one atomic unit scale indicated

image ensemble size of 10^6

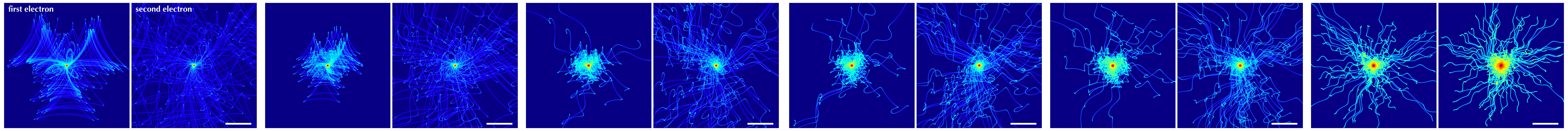
The dynamics of the single and double ionization of **helium and neon** are well described within the strong-field tunneling framework. Non-sequential double ionization (NSDI) is most effective with linear polarization and counter-rotating fields [3], and the electron momentum distributions (above) from helium single ionization match the "simpleman" prediction remarkably well. The peak of the momentum distributions is shifted from the ionization time demonstrating a fully classical manifestation of an "attoclock" [4]. The trajectories of electrons involved in double ionization (below) exhibit beautiful rescattering behavior. Trajectories from the first (left) and second (right) released electron are displayed. For each trajectory, two optical cycles before each electron's release are shown, and (for the first electron) the next two optical cycles are shown, or up to the rescattering time, whichever comes first (for the second electron, the next two optical cycles are shown). For both helium and neon, every double ionization occurrence is due to a returning event, either a short, sub-cycle looping path, a single-cycle triangular path, or multiple triangular paths.

In the middle ground between the tunneling and multiphoton regimes, the ionization of **argon and krypton** begins to exhibit significant effects from interactions with the Coulomb potential. NSDI becomes more enhanced even with corotating fields, and the electron momentum distributions are substantially distorted. Returning electron trajectories still account for the majority of the double ionization events in both argon and krypton, but double ionization now occurs even in some cases where the first electron never makes a close return to the parent ion. A larger Keldysh parameter signifies an effectively stronger Coulomb potential *relative to the laser field*, but at the same time, the weaker Coulomb potential *in absolute terms* means the electron-electron repulsion has a bigger influence. These effects, combined with the smaller electron excursions into the laser field, mean that double ionization can occur in unusual pathways.

Finally, at the weakest laser fields, the ionization of **xenon and magnesium** is barely recognizable from what was seen in helium. NSDI is still more effective with counter-rotating fields in xenon, but some corotating fields outperform their counterparts in magnesium, and even a significant enhancement in double ionization is seen with circular polarization (as famously observed [13]). The electron momentum distributions from the wildly different counter-rotating and corotating fields look nearly identical (in stark contrast to what was seen in helium). And while some returning trajectories contribute to double ionization in xenon, they are essentially absent in magnesium.

Future work: continue to explore the transition in the dynamics of non-sequential double ionization using the classical ensemble approach with a comprehensive analysis of electron momentum, energy, timing, and trajectories.

DI electron trajectories



electron trajectories from double ionization

first electron (left panel)
second electron (right panel)

2:1 400nm:800nm
counter-rotating
intensity at red arrow

10Å scale indicated
100 trajectories per image