High-order harmonic generation from Rydberg states

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Motivation and previous work

- Rydberg atoms provide an alternative venue for studying strong field phenomena without the need for **high intensities** and *short* **pulse durations**.
- Keldysh parameter γ<1 implies **tunneling ionization**. When I_p scales as $\sim 1/n^2$, scaling F by $\sim 1/n^4$ and ω by $\sim 1/n^3$ leaves γ **unchanged**.
- Enhancement for HHG yields has been proposed utilizing **superposition of excited states and the ground state as the initial state**, as well as **two-colored driving lasers**. All these processes include excitation to an excited state, which assists the ionization stage of HHG.

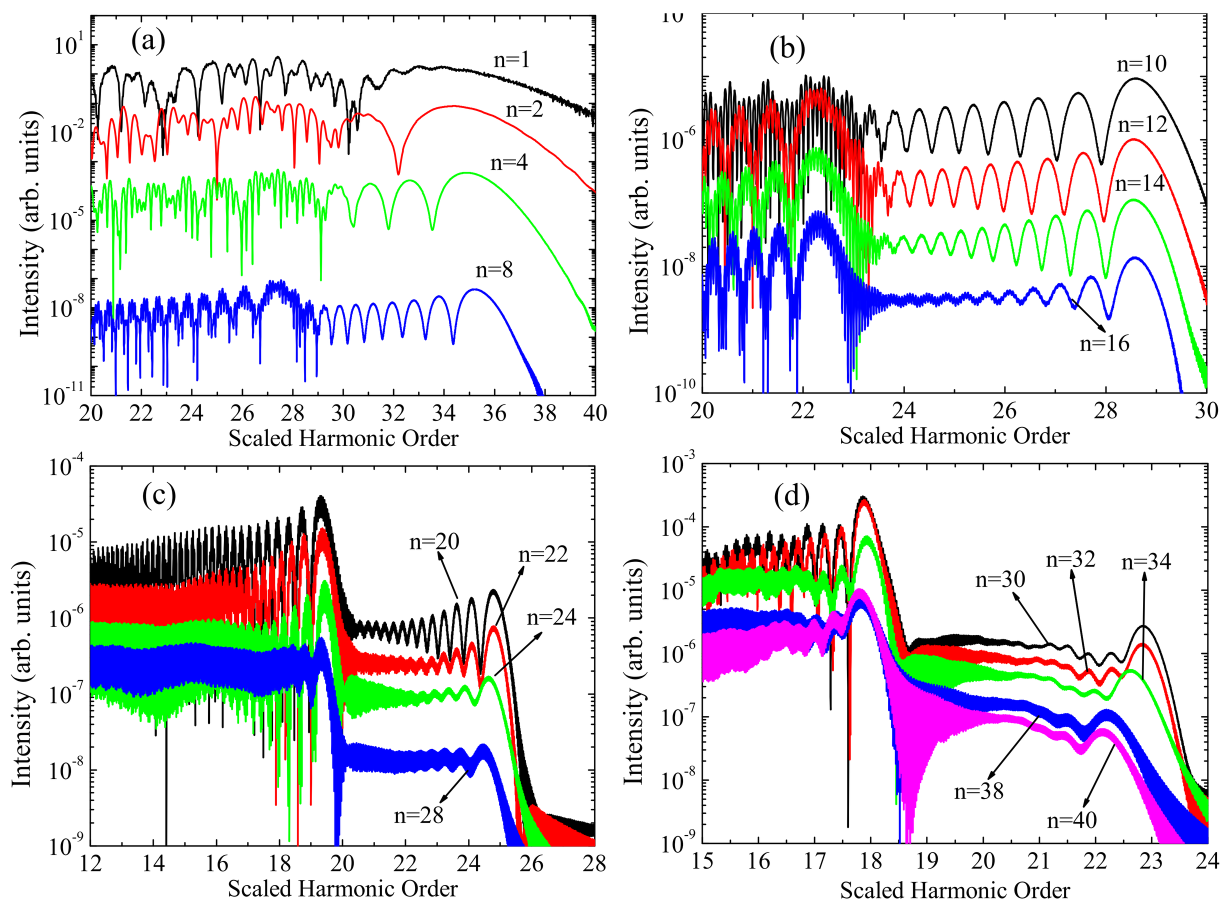
[Z. Zhai *et. al*, Phys. Rev. A **83** 043409 (2011)]

One-dimensional Calculations

We solve the time-dependent Schrödinger equation within a one-dimensional s-wave model using a linearly polarized 4-cycle laser pulse for H atom.

$$i\frac{\partial\psi(r,t)}{\partial t} = \left[-\frac{1}{2}\frac{d^2}{dr^2} - \frac{1}{r} + rF(t)\right]\psi(r,t).$$

The selected parameters correspond to $\gamma = 0.75$ for all cases, which implies **tunneling** regime. The cut-off frequency for a given state n scales as $\omega_c^{(n)} = \omega_c^{(1)}/n^2$. The emergent double plateau structure is similar to that seen for two-color driving.



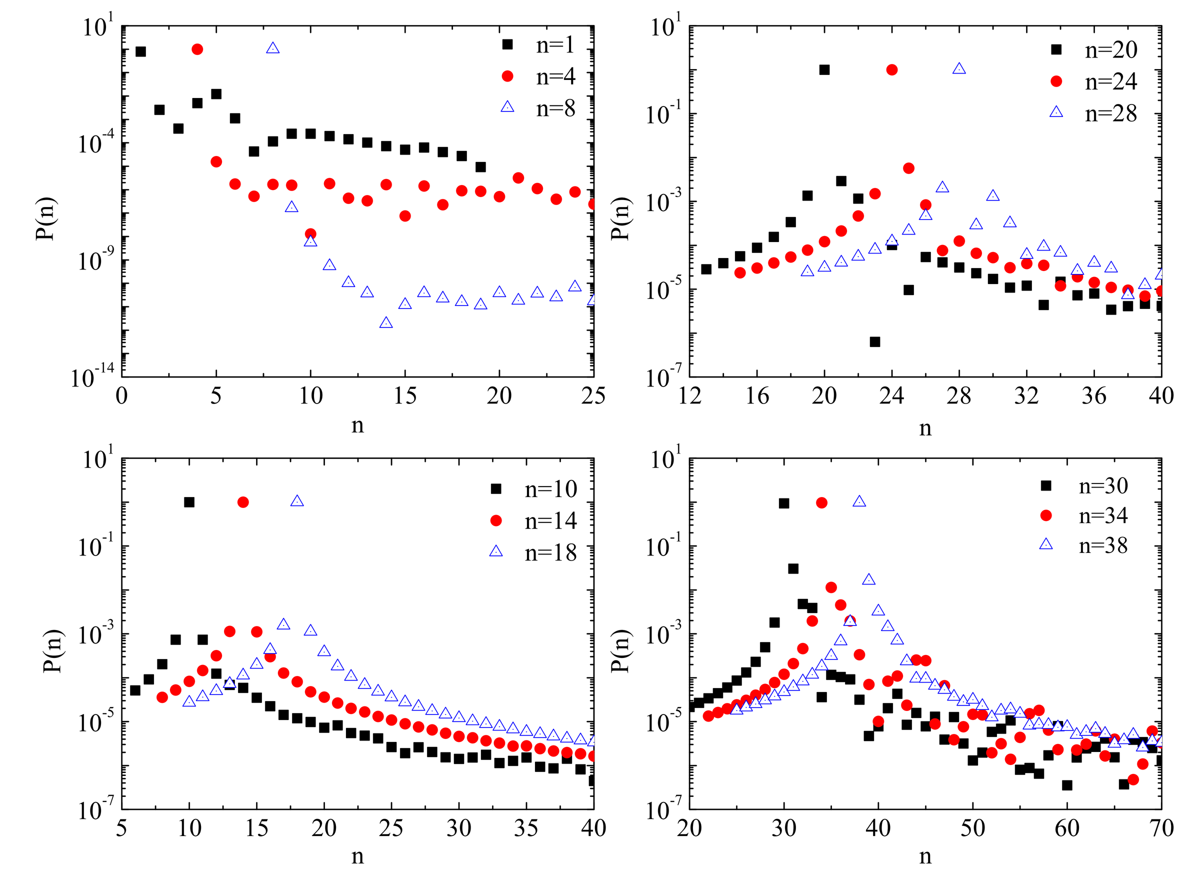
(a) 200/n^8 TW/cm^2 *and* 800n^3 nm, **(b)** 300/n^8 TW/cm^2 *and* 652n^3 nm, **(c)** 400/n^8 TW/cm^2 *and* 566n^3 nm, **(d)** 470/n^8 TW/cm^2 *and* 522n^3 nm.

The double plateau structure

The double plateau feature can be understood investigating scaled harmonic order of the cut-off as a function of n. **Consider the case of $n = 8$:**

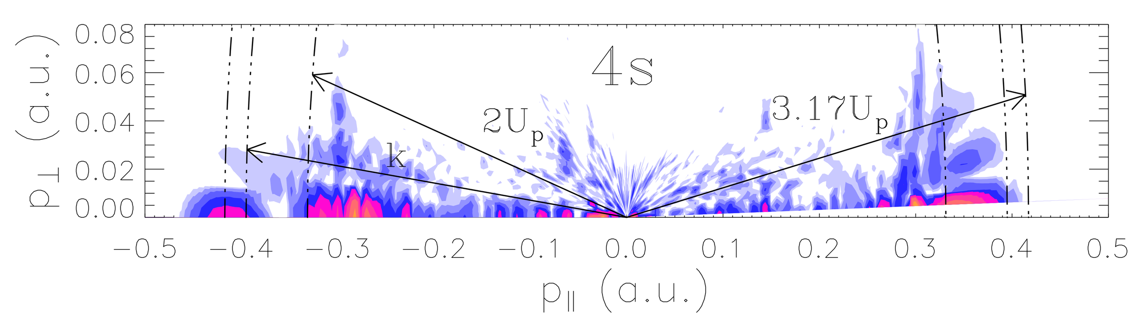
n	$(I_p + 3.17U_p)/(n\,\omega_0)$
8	~ 33
14	~ 27
∞	~ 24

The laser pulse excites the initial $n = 8$ state to states up to n~14, whose scaled cut-off is at ~27. This is the cut-off for the **first plateau of lower harmonics**, resulting from ionization and recombination from the excited states. The usual cut-off for $n = 8$ terminates the entire **HHG plateau at high** ω.



The final n-distributions taken after the laser pulse show population spreading to highly excited states. This is the process which forms the **secondary enhanced plateau** at the lower ω end of the spectrum.

Fourier transforming the ionized part of the **3d wave function** ψ, we can see its momentum distribution at the **end of the pulse**. This is a good reflection of *what has contributed* to the HHG spectrum.

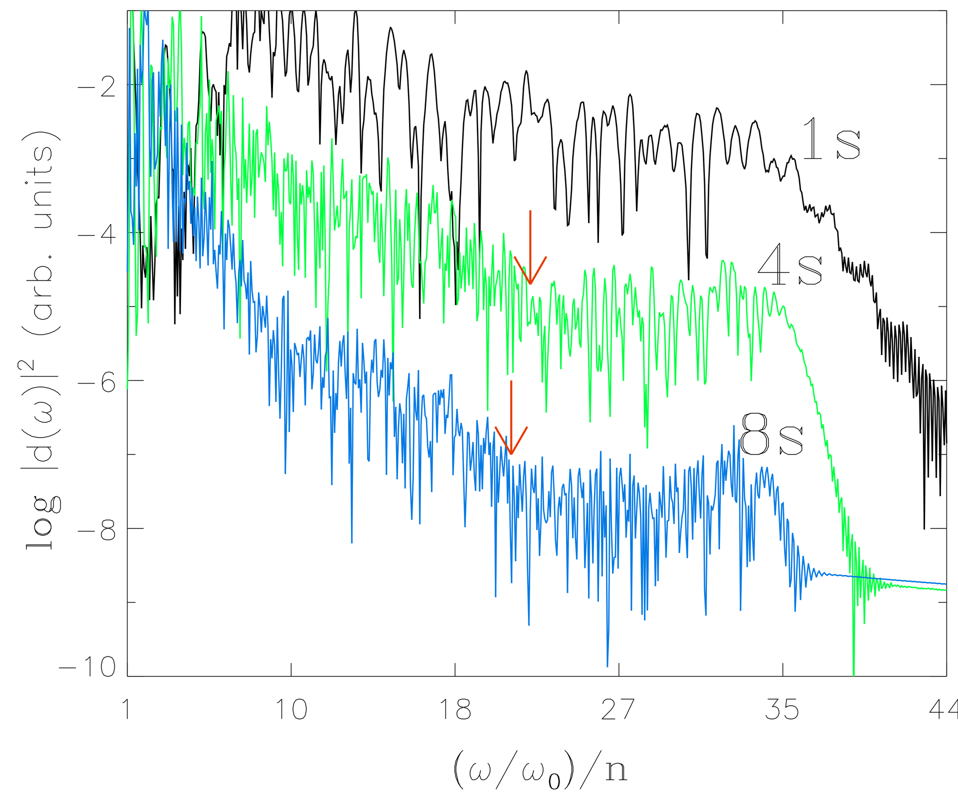


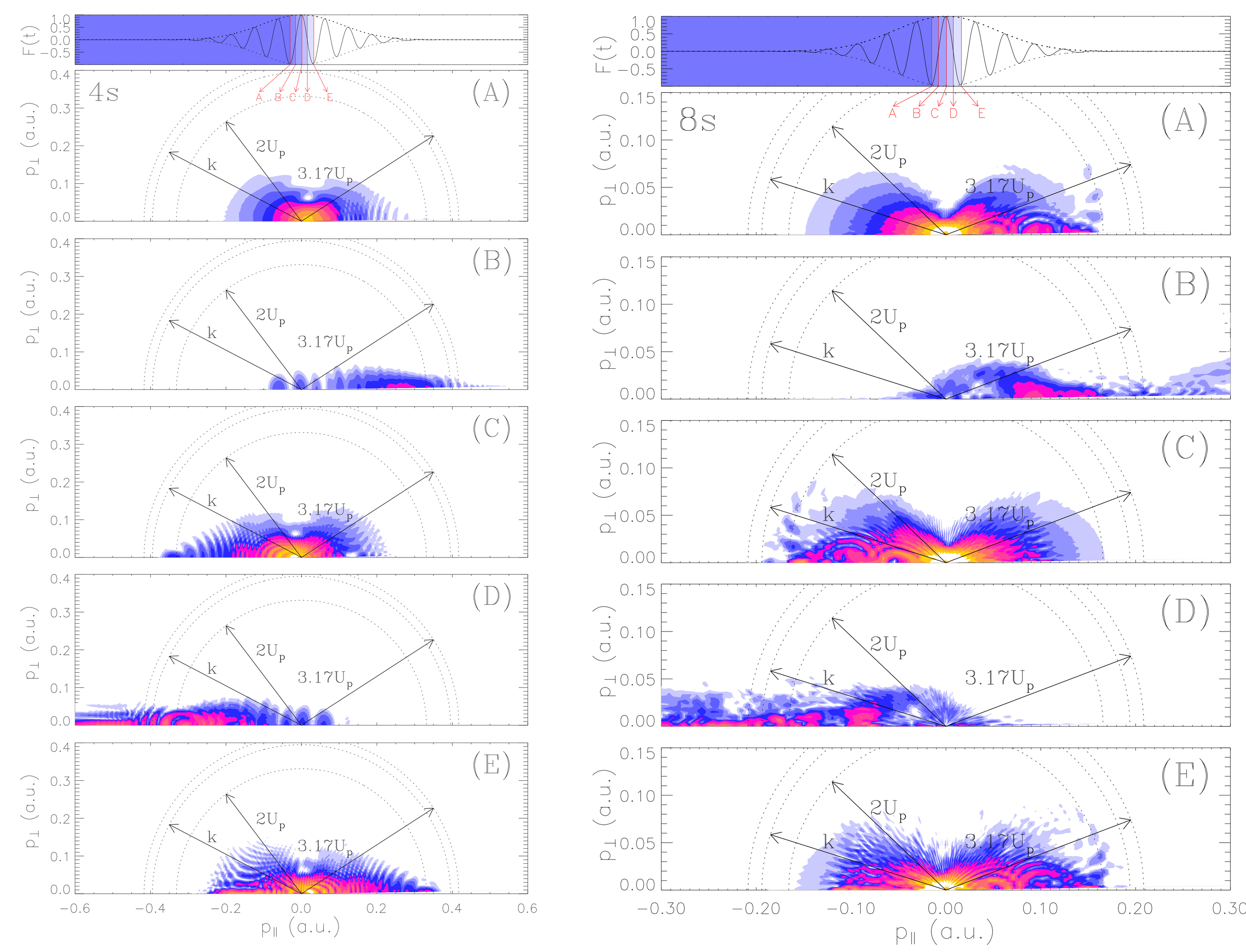
The original cut-off at $\pm 3.17U_p$ and the secondary cut-off (labeled as k) are clearly visible. Ionization mostly happens **along the laser polarization axis** as in typical strong field HHG from ground states.

Three-dimensional Quantum Calculations

(Left column) We solve the full 3d time-dependent Schrödinger equation starting from the initial states $1s$, $4s$, and $8s$ for the same set of laser parameters.

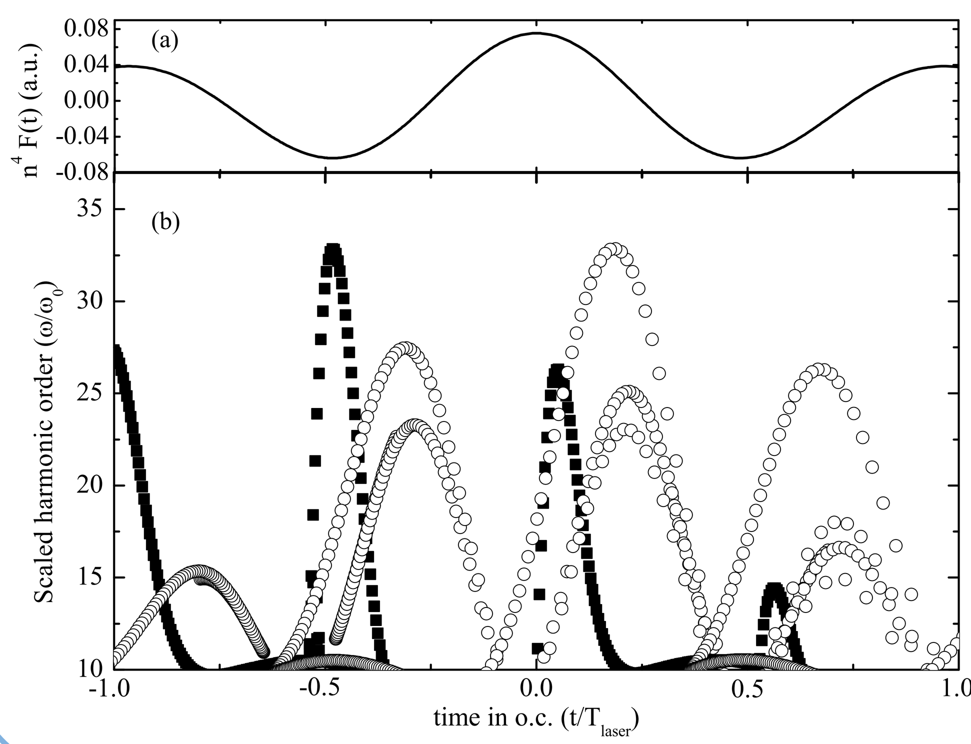
The flat secondary plateau at small ω followed by the sharp secondary cut-off seen in the 1d spectra is replaced by an enhancement of lower harmonics which is skewed towards the lower harmonics. This is facilitated by the various different AC Stark shifts experienced by different ℓ-states in a given n-manifold. This makes it possible for the state population to drift towards even more highly excited states, giving out lower harmonics more efficiently upon recombination. (Right columns) Momentum maps at select few points during the peak cycle of the pulse, beyond the peak of the combined Coulomb/laser potential at $1/\sqrt{F}$ along the laser polarization.





The uneven enhancement in the second plateau, in contrast with the uniform profile predicted from the 1d models, suggests that schemes for composing more intense attosecond pulses relying on the excited states, such as two-color driven HHG, may not be as promising as previous 1d models predicted.

Classical trajectories



The total energy of an electron in the laser field when it comes back to the nucleus calculated from the classical equations of motion as a function of release (filled squares) and return times (open circles).

Classical equations of motion scale exactly. The trajectories contributing to the the higher ω plateau are those that extend above the scaled harmonic order ~24 (for the $n = 1$, 2, 4, and 8 batch). This marks the limit where excitation to $n \to \infty$ assists the ionization step of the HHG.

Acknowledgements

I.Y., E. A. and Z. A. was suported by BAPKO of Marmara University. T. T. was supported by the Office of Basic Energy Sciences, US Department of Energy.