Intense field stabilization in circular polarization: 3D time-dependent dynamics

Dae-Il Choi[†] and Will Chism[‡]
[†]*Laboratory for High Energy Astrophysics, NASA Goddard Space Flight Center, Greenbelt, MD 20771*
[‡]*Center for Statistical Mechanics, The University of Texas, Austin, TX 78712*
(September 13, 2001)

We use a fully three dimensional time-dependent approach to investigate the quantum dynamics of the Rydberg atom subject to high intensity, short wavelength laser pulses of circular polarization. We find enhanced survival probability as the field is increased under fixed pulse conditions. We also confirm wavepacket behaviors seen in prior two dimensional time-dependent computations.

PACS numbers: 32.80.Fb,32.80.Wr,42.50.Hz

The advent of lasers producing electric fields at or above inter-atomic electric fields has led to the discovery of many unforeseen highly nonlinear phenomena [1]. As nonlinear laser-atom physics has matured, computational approaches employed by researchers in the field have naturally been applied to scenarios not easily realized in experiments. One such scenario is the interaction of an intense, *high frequency* laser with the single-electron hydrogen atom. Currently available tabletop pulsed laser systems readily produce atomic "intensities" $I \approx 10^{16} W/cm^2$, but available output frequencies are well below the "atomic unit" frequency, given by the hydrogen ground state ionization energy ($|E_o| = 13.6eV$). In the case where the driving frequency is at or above the ionization energy a remarkable phenomena known as "stabilization" occurs. This is characterized by a decrease in the ionization probability as the laser intensity increases. For constant driving field, stabilization is manifest as an increase in dressed state *lifetime*. However, due to the initial increase in ionization rate for increased fields as the interaction turns on, increased steady-state lifetimes do not necessarily translate into increased end-of-pulse survival probabilities. This is commonly known as the "death-valley" problem.

Nevertheless, indications of stabilization have been seen by tailoring the initial state to act as an effective ground state [2–5]. In particular, the high frequency condition has been realized using dipole forbidden circular Rydberg states ($n = 4, 5$) of neon ($|E_o| < 1eV$), and a driving laser of $\hbar\omega = 2eV$ [2]. In this experiment, a decrease in total ionization yield was observed with increasing peak laser intensity, while fluence was held constant [3]. Fermi's golden rule predicts the total ionization depends only on the fluence, so this result clearly indicates non-perturbative stabilization [4,5]. With this notable exception, however, stabilization remains experimentally unconfirmed, and detailed studies have been confined to the realm of simulation.

The vast majority of these studies have concentrated on the case of linearly polarized (LP) fields, where much has been learned about the dynamics of stabilization [6]. Studies considering the case of circularly polarized (CP) fields have also noted comparable or enhanced stabilization [7–10]. However, the physical mechanism for stabilization in the CP case remains open, as the electron dynamics are dramatically different from the dynamics in LP fields. Recent investigations of the CP Rydberg system, restricted to the two-dimensional plane of polarization, have found "ring-like" probability distributions, among a variety of waveform behaviors. This ring structure is generally found to rotate in phase with the laser field, about a point displaced from core. A probability maximum may also occur at the center of the rotating ring. All prior fully three dimensional studies have been limited to the case of LP systems [11].

In this Letter, we investigate the quantum dynamics of stabilization in the CP Rydberg system using a three dimensional time-dependent approach. In this system, the electron wavefunction moves under the influence of the Coulomb force, and circularly polarized radiation field. For the fields and frequencies considered herein, the interaction is appropriately described in the dipole approximation, and radiation field may be assumed classical. Adopting atomic units ($\hbar = 1 = e = m$), the Hamiltonian for the electron may be written

$$H = -\frac{1}{2}\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2}\right) - \frac{1}{r} + F(t)(\hat{x}\cos(\omega t) + \hat{y}\sin(\omega t))$$

where $\hat{x}$ and $\hat{y}$ are position operators, and $F(t)$ describes the laser field envelope. The Schrödinger equation is integrated using Crank-Nicholson finite differencing and absorbing boundary conditions [12]. In order to avoid the singularity at the origin, we offset the position of points on the grid by half the distance between adjacent gridpoints. This places gridpoints symmetrically about the origin, and poses no problem provided the spatial discretization level is sufficient to resolve the maximal components of momentum k_{max} attained by the wavefunction. This can be estimated with confidence from the high harmonic generation (HHG) cutoff law since harmonics produced under stabilization conditions are known to be suppressed [1]. The HHG cutoff energy is given by $\hbar\omega_{HHG} \approx |E_o|+3U_p$, where $U_p = F^2/4\omega^2$ is the pondermotive energy of an electron in a LP field. Thus k_{max} is proportional to F/ω. Setting the laser frequency $\omega = 1.2$, and considering fields up to $F \approx 4.0$, we find a discretization of $\delta x \approx 0.1667$ is sufficient to resolve all components of the wavefunction. To our knowledge this is the largest field considered in studies of stabilization for circular polarization.

To determine the survival probability, we start with a stationary gaussian wavepacket centered at the origin with width $\sigma = 1.5$, and pulse the laser interaction with an envelope $F(t) = F\sin^2(\frac{\pi t}{6\tau})$, over the time interval $0 < t < 6\tau$, where τ is the optical cycle. A portion of the wavefunction ionizes off the grid during the laser turn-on, and at the end of the pulse, remaining probability essentially occupies the atomic ground state. As the field strength is increased, we find the total probability remaining on the grid at the end on the pulse increases, characteristic of stabilization. This is demonstrated in Fig. 1, where we show the time dependent survival probability on the grid over the 6-cycle pulse, for the field strengths $F = 2.5$, $F = 3.0$, $F = 3.5$, and $F = 4.0$. The probability on the grid remains close to unity during the first half of the pulse for all field strengths. This corresponds to the time it takes components of the wavefunction to traverse the grid and ionize. At a point near two laser cycles the probability begins to drop significantly, with lower total probability occuring for larger field strengths. Therefore, an increase in total ionization occurs during the laser ramp-up for higher fields. However, during the following two cycles when the laser field is closest to its maximum, ionization occurs more slowly, or at a reduced rate, for more intense fields. The total ionization for the lower fields then catches up to and surpasses that of the higher fields. This occurs just beyond four laser cycles for each of the fields we consider, and this trend continues to the end of the laser pulse, resulting in enhanced survival probability as a function of field strength.

FIG. 1. Time dependent survival probabilities for F=2.5, 3.0, 3.5, & 4.0, over the six cycle pulse. As the laser field strength increases from F=2.5 to F=4.0, the end-of-pulse survival probability also increases, characteristic of stabilization.

FIG. 2. Snapshots of the wavepacket during the laser pulse, for field F=3.5. The first three shots show the time development from approximately 3 to 3.5 cycles. The spiral probability distribution has closed, and rotates in phase with the laser field, producing an apparent displaced ring structure. The last frame shows the end-of-pulse hydrogen ground state.

While Fig. 1 confirms the presence of the stabilization for the fields and frequency we consider, the fundamental stabilization mechanism remains obscure. In order to address this issue we must examine the time dependent wavepacket dynamics. Now, when a CP laser field of sufficient intensity to induce direct ionization is turned on, the electron probability will generally flow away from the core along a spiral path to eventual ionization. However, for the fields we consider, an interesting twist on this effect is seen. The spiral path begins to close back upon itself. This produces a probability trapping effect, as the ionization channel is closed. This is evidenced by the formation a ring-like probability distribution.

FIG. 3. Probability profile along the x axis for after approximately 3 cycles for each of the field strengths we consider. Each profile exhibits three maxima: One at the core, one along the laser field (at positive x), and one on the "back-side" (at negative x). As the laser field strength increases from F=2.5 to F=4.0, the spiral closes, as exhibited by the shift of the back-side maxima toward the origin.

In Fig. 2 we show a series of snapshots near the the peak of the pulse exhibiting this behavior. The field strength is $F = 3.5$ and the time is $t = 15.544$, approximately 3 laser cycles. We find a ring-like structure of enhanced probability. Probability on this ring is asymmetrically distributed with maximum occuring along the direction of the rotating laser field. This peak follows a circular orbit about the core, with approximate radius F/ω^2. The entire ring structure also rotates about the core, with one edge intersecting the core. Thus, the ring is displaced along the field, and rotates, locked in phase with the field, about the origin. This behavior cannot be predicted or observed in time-independent (Floquet) or time-averaged (Kramers-Henneberger) approaches. Specifically, if one takes a time average of the probability, it may result in a ring-like distribution, but this will always be centered on the origin and cannot exhibit the displacement along the laser field evident in time dependent-studies.

In Fig. 3, we illustrate the displacement and closure of the spiral arm explicitly with a cross sectional slice of the probability distribution. We take a cross section for each field strength, near the peak of the pulse. We choose $t \approx 3\tau$, so that the laser field is aligned along the x-axis. Each profile exhibits three maxima: one near the core, one along the laser field at positive x, and one on the "back-side" at negative x. These maxima correspond to probability remaining near the core, and the two locations at which the spiral tail intersects the x-axis. As the field increases, the maxima at positive x shifts outward along the field direction, and the "back-side" maxima shifts toward the core. This corresponds to the displacement of the ring along the field, and closing of the tail, which forms the ring-like structure. Similar displaced ring structures have been noted in time dependent two dimensional studies of stabilization [9,10,12]. Another interesting feature of Fig. 3 is the decrease in probability amplitude at the spiral tail crossing points, relative to the probability amplitude at the core, seen as the field increased. This a laser ramping effect, *i.e.* the limited time available for the probability to spread over the ring leads to unequal population on the ring. In this case, as we increase the field, the maximum probability shifts along the ring, from the point located along the laser field, back to the core. However, the location of the probability peak may be adjusted, provided the laser interaction is ramped quickly to avoid complete ionization.

In conclusion, for a Rydberg atom subject to an intense, high frequency laser pulse of circular polarization, we have found a decrease in total ionization yield as the field strength is increased under fixed pulse envelope conditions. For this stabilization phenomena in circularly polarized fields, the characteristic waveform is a ring-like structure, displaced along the laser electric field. This entire displaced ring structure is locked in phase with the rotating field. This behavior can only observed in time-dependent approaches. Our fully three dimensional investigation confirms prior results obtained using a two-dimensional time dependent approach, and indicates the two dimensional time-dependent approach is likely to provide the necessary physical insight into stabilization while maintaining reasonable computational effort. These ring-like waveforms are typically asymmetrically populated, and population of these stabilized structures may be achieved from the ground state with sufficiently short pulses.

---

[1] *Atoms in Intense Laser Fields*, edited by M. Gavrila (Academic Press, San Diego, 1992); *Super-Intense Laser-Atom Physics*, edited by B. Piraux *et al.* (Plenum Press, New York, 1993); K. Burnett, V. C. Reed, and P. L. Knight, J. Phys. B **26**, 561 (1993); M. Protopapas, C. H. Keitel, and P. L. Knight, Rep. Prog. Phys. **60**, 389 (1997).

[2] R. J. Vos and M. Gavrila, Phys.Rev.Lett. **68**, 170 (1992).

[3] M. P. de Boer, *et al.*, Phys. Rev. Lett. **71**, 3263 (1993); M. P. de Boer, *et al.*, Phys. Rev. A **50**, 4085 (1994).

[4] N. J. van Druten, *et al.*, Phys. Rev. A **55**, 622 (1997).

[5] B. Piraux and R. M. Potvliege, Phys. Rev. A **57**, 5009 (1998).

[6] R. Grobe and C. K. Law, Phys. Rev. A **44**, R4114 (1991).

[7] J. Zakrzewski and D. Delande, J. Phys. B **28**, L667 (1995).

[8] M. Pont and M. Gavrila, Phys. Rev. Lett. **65**, 2362 (1990); M. Pont, Phys. Rev. A **40**, 5659 (1989).

[9] A. Patel *et al.*, Phys. Rev. A **58**, R2652 (1998); A. Patel, N. J. Kylstra and P. L. Knight, Optics Express **4**, 496 (1999).

[10] Will Chism, Dae-Il Choi, and L. E. Reichl, Phys. Rev. A **61**, 054702 (2000).

[11] K. C. Kulander, K. J. Schafer, and J. L. Krause, Phys. Rev. Lett **66**, 2601 (1991).

[12] Dae-Il Choi, Univ. of Texas at Austin Ph.D. thesis (1998).