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

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We investigate the stabilization of a hydrogen atom in circularly polarized laser fields. We use a time-dependent, fully three dimensional approach to study the quantum dynamics of the hydrogen atom subject to high intensity, short wavelength laser pulses. We find enhanced survival probability as the field is increased under fixed envelope conditions. We also confirm wavepacket dynamics seen in prior time-dependent computations restricted to two dimensions.

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The advent of lasers producing electric fields at or above inter-atomic electric fields has led to the discovery of many new and highly nonlinear phenomena. 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. Commercially available pulsed laser systems readily produce intensities at or above the atomic unit frequency, which is given by the ground state ionization energy \( \hbar \omega \). In the case where the driving frequency is near or above the ground state ionization energy, \( \hbar \omega \geq |E_o| \), a remarkable phenomena known as “stabilization” may occur. Stabilization 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 poses a significant challenge to experimental observation of stabilization and 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. 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 \( \hbar \omega = 2eV \). In this experiment, a decrease in total ionization yield was observed with increasing peak laser intensity, while fluence was held constant. Fermi’s golden rule predicts the total ionization depends only on the fluence, so this result clearly indicates non-perturbative stabilization. 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. Studies considering the case of circularly polarized (CP) fields have also noted comparable or enhanced stabilization. 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 time-dependent investigations of the CP Rydberg system, restricted to the two-dimensional plane of polarization, have found “ring-like” probability distributions. This ring structure is generally found to rotate in phase with the laser field, about a point displaced from core. Thus the motion resembles the motion of a hula-hoop. 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.

**Computational Model** – In this Letter, we investigate the quantum dynamics of stabilization in the CP hydrogen 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 with classical radiation field. Adopting atomic units \( (\hbar = 1 = e = m) \), the Hamiltonian for the electron may be written

\[
H = -\frac{1}{2} \nabla^2 - \frac{1}{r} + F(t)(\hat{x}\cos(\omega t) + \hat{y}\sin(\omega t))
\]

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. 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 wave-
function. 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 \( \text{[reference]} \). The HHG cutoff energy is given by \( \hbar \omega \approx |E_0| + 3 U_p \), where \( U_p = F^2/\omega^2 \) is the ponderomotive energy of an electron in a LP field. Thus \( k_{\text{max}} \) is essentially proportional to \( F/\omega \). Setting the laser frequency \( \omega = 1.2 \), and considering fields up to \( F \approx 4.0 \), we find a spatial 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 to date in studies of stabilization for circular polarization.

**Survival Probability** – To determine the survival probability, we start with a stationary gaussian wavepacket centered at the origin with width \( \sigma = 1.5 \), and introduce the laser interaction with a pulse envelope \( F(t) = F \sin^2(\pi t/\tau) \), over the time interval \( 0 < t < 6 \tau \), where \( \tau \) 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 of the pulse increases, characteristic of stabilization. This is demonstrated in Fig. 5, 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 occurring 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.

**Wavepacket Dynamics** – While Fig. 3 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. In Fig. 3 we show a series of snapshots near 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 occurring along the direction of the rotating laser field. This peak follows a circular orbit about the core, with approximate radius \( F/\omega^2 \). 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-averaged (Kramers-Henneberger) approaches. Specifically, if one takes a time average of the probability, it may result in an apparent “ring-like” distribution, but this will necessarily be rotationally invariant about the origin, and cannot exhibit the displacement along the laser field evident in time dependent-studies. Thus the actual “hula-hoop” motion will be concealed by the time averaging. The last frame of Fig. 3 also shows the end-of-pulse probability distribution, which has overlap with the hydrogen ground state to within \( 10^{-5} \).

**Shifts in Ring Structure** – 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. These displaced ring structures have been noted in time dependent two dimensional studies of stabilization \( \text{[reference]} \). Another interesting feature of Fig. 3 is the decrease in probability amplitude seen at the spiral tail crossing points, relative to the probability amplitude at the core, 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 distribution on the ring. In this case, as we increase the field, the maximum probability shifts along the ring, from a point located along the laser field, backwards along the spiral arm, toward the core. This is indicative of the fact that the probability distribution on the stabilized ring structure may be adjusted by the pulse ramping.

In conclusion, for a hydrogen 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 con-
ditions. 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, executing a “hula hoop” type motion. This behavior can only be 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.

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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, $|\psi(x, y, z = 0)|^2$, during the laser pulse, for field F=3.5 on Z = 0 plane. 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 (with relative probability scale of 1/7 w.r.t to other 3 frames).
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.