# Bidirectionally kicked Rydberg atoms: Population trapping near the continuum

W. Zhao,[1] J. J. Mestayer,[1] J. C. Lancaster,[1] F. B. Dunning,[1] C. O. Reinhold,[2,4] S. Yoshida,[3] and J. Burgdörfer[3,4]
[1]*Department of Physics and Astronomy, Rice University, 6100 Main Street, Houston, Texas 77005-1892, USA*
[2]*Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6372, USA*
[3]*Institute for Theoretical Physics, Vienna University of Technology, A1040 Vienna, Austria*
[4]*Department of Physics, University of Tennessee, Knoxville, Tennessee 37996-1200, USA*
(Received 4 October 2005; published 13 January 2006)

The behavior of high-$n$ Rydberg atoms subject to a periodic sequence of short half-cycle pulses (HCPs) that alternate in sign is reexamined. Each HCP delivers an impulsive momentum transfer or "kick" to the electron and the net average field it experiences is zero. The data show that the excited electron energy distribution initially moves toward the continuum, i.e., toward states of higher $n$, but then becomes transiently trapped near the edge of the continuum leading to sizable overall survival probabilities. The physical mechanisms responsible for this are discussed with the aid of classical trajectory Monte Carlo simulations.

DOI: 10.1103/PhysRevA.73.015401 PACS number(s): 42.50.Vk, 32.80.Rm

Much interest centers on the control and manipulation of Rydberg atoms using one (or more) pulsed unidirectional electric fields, termed half-cycle pulses (HCPs), of duration $T_p \ll T_n$, where $T_n$ is the classical electron orbital period [1]. Indeed, it has been suggested recently that trains of periodic attosecond-duration HCPs can be produced through high harmonic conversion using a two-color laser field and that such trains might be used to engineer the wave functions of atoms in ground or low-lying excited states [2]. For such freely propagating HCP trains, the net average electric field is zero. This, therefore, has stimulated interest in how the dynamical response of an atom to a train of HCPs depends on the average field it experiences.

The response of atoms to a variety of HCP sequences has been investigated in this laboratory using high-$n$ Rydberg atoms and HCPs produced by applying voltage pulses to a nearby electrode. With this approach, HCP trains with zero average field can be generated by using a periodic train of HCPs that alternate in sign [3] or a periodic train of unidirectional HCPs and a superposed offset field [4,5]. The latter technique has been used recently to examine, in some detail, the effect of the net field on the atomic dynamics. The data show that when the net field experienced by the excited electron is zero, the atom is able to survive a large number of HCPs. This results because the distribution of electron energies, $E$, after initially moving toward higher energies, i.e., toward larger values of principal quantum number $n$, becomes transiently localized and trapped near $E=0$. Those electrons with $E<0$ correspond to bound states with very large values of $n$, typically $\gtrsim 800$, and account for the large overall survival probability. This trapping of the electron near threshold is the result of transport of the electron into a region of phase space where its response to the external HCP train resembles that of a quasifree electron, which suffers no net transfer of energy or momentum [6]. We have also shown recently that trapping is not associated with the existence of stable islands in phase space [5]. Rather, it is due to a slowing of the diffusion process, the very-high-$n$ states ultimately becoming ionized after a large number of HCPs.

In this Brief Report, we reexamine the problem of atoms subject to trains of HCPs that alternate in sign within the context of freely propagating pulses. We provide new data for both high-$n$ $p$ states and quasi-one-dimensional (quasi-1D) states which further demonstrate trapping of the electron near threshold. We examine this behavior using classical trajectory Monte Carlo (CTMC) simulations together with its sensitivity to aperiodicities in the HCP train. Such aperiodicities lead to marked changes in the model predictions and, when taken into account, yield much improved agreement with experiment.

The present apparatus is described in detail elsewhere [1,3,4]. Briefly, potassium atoms contained in a collimated beam are photoexcited in near-zero ($\lesssim 50$ $\mu$V cm$^{-1}$) electric field to the $351p$ state using an extra-cavity doubled CR699-21 Rh6G dye laser. Experiments are conducted in a pulsed mode. The output of the laser is chopped into a train of pulses of $\sim 1$ $\mu$s duration using an acousto-optic modulator. Following excitation, the atoms are subject to the train of $N$ pairs of alternating HCPs diagrammed in the inset in Fig. 1. This pulse sequence was generated by using a combiner to superpose a train of $N$ positive HCPs with a similar (delayed) train of $N$ negative HCPs and applying the resultant wave form to a nearby electrode. The profiles of the applied pulses were monitored using a fast probe and storage oscilloscope. Measurements revealed an interesting artifact associated with each of the superposed HCP trains. Whereas the amplitudes and profiles of the positive and negative pulses were essentially identical and constant throughout the pulse train, the separation between the first two positive pulses and the separation between the first two negative pulses was systematically $\sim 16\%$ larger than that between succeeding pairs of positive and of negative pulses. As illustrated in the inset, this leads to an anomalously large separation between the first negative and second positive pulses. (The separations between all other neighboring positive and negative pulses are equal.) The number of atoms that survive the HCP train, and their excited state distribution, is determined using selective field ionization (SFI) by applying a slowly varying ramped electric field. Because atoms in different Rydberg states ionize at different applied fields, i.e., at different times during the ramp, observation of the time dependence of the

 

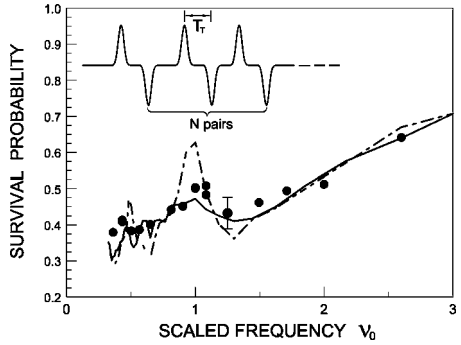


FIG. 1. Survival probability for $K(351p)$ atoms subject to $N=20$ pairs of alternating HCPs delivering scaled impulses $|\Delta p_0|=0.21$ as a function of scaled frequency $\nu_0$ (see text). The results of CTMC simulations are included which assume (– – – – –) a "perfect" HCP train and (———) a 16% increase in separation between the first two positive and first two negative kicks (see text). Atoms with $n \geqslant 1200$ are presumed to be ionized by stray fields. The inset illustrates the HCP sequence used in this work.

ionization signal provides a measure of the initial excited state distribution. Measurements in which no HCPs are applied are interspersed at routine intervals during data acquisition to monitor the number of Rydberg atoms initially created. The Rydberg atom survival probability is then determined by taking the ratio of the SFI signals observed with and without the HCPs applied.

Figure 1 shows the survival probability for $K(351p)$ atoms subject to $N=20$ pairs of HCPs that deliver scaled impulses or "kicks" of magnitude $\Delta p_0 \equiv n\Delta p = n\int_0^\infty F_{HCP}(t)dt = 0.21$, where $F_{HCP}(t)$ describes the field produced by each ~600 ps full width at half maximum HCP (atomic units are used throughout), as a function of the scaled HCP frequency $\nu_0 \equiv \nu_T/\nu_n = T_n/T_T$, where $T_n = 2\pi n^3$ and $T_T(\equiv 1/\nu_T)$ is the separation between successive pulses in the HCP train. The general increase in survival probability observed with increasing scaled frequency is not unexpected. As the scaled frequency increases, the separation between successive kicks becomes shorter and, in essence, the effect of one kick is immediately reversed by the next. A local maximum in the survival probability is observed for scaled frequencies $\nu_0 \sim 1$. Earlier work shows that this results because the phase space for an atom driven by a bidirectional sequence of kicks contains a sizable "leaky" torus or cantorus that is embedded in an otherwise chaotic sea [3]. For $\nu_0 \sim 1$, a fraction of the parent atoms have initial phase points that lie within this cantorus. Such phase points remain trapped therein for an extended number of kicks before drifting away and into the continuum, leading to the observed increase in survival probability.

Figure 1 also includes the results of CTMC simulations, which assume that product states with $n > 1200$ are ionized by stray residual fields present in the apparatus. These simulations were fully three-dimensional and were undertaken using the Hamiltonian

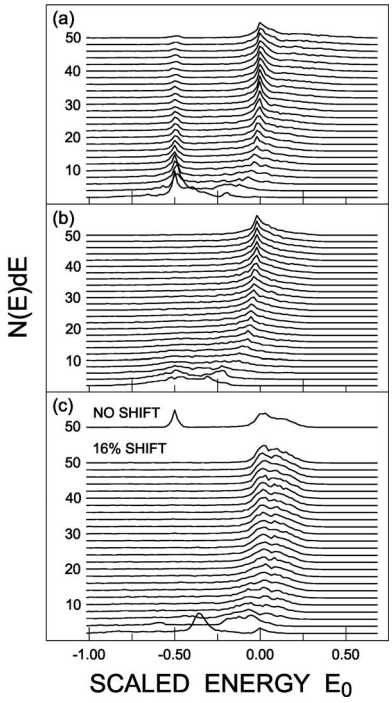


FIG. 2. Evolution of the electron energy distribution as a function of the number $N$ of HCP pairs for (a) $351p$ atoms and a scaled frequency $\nu_0=1$, (b) $351p$ atoms and $\nu_0=3$, and (c) quasi-1D $n=350$, $m=0$ atoms and $\nu_0=1$. The energy is expressed in scaled units and each HCP delivers an impulse $|\Delta p_0|=0.21$. The 16% larger separation between the first two positive and first two negative pulses is included (see text). The upper distribution in (c) is that expected for a quasi-1D atom when using a "perfect" train of $N=50$ HCPs (see text).

$$H(t) = H_{at} + z\sum_{j=1}^{2N} (-1)^{j+1} F_{HCP}(t - j\nu_T^{-1}), \qquad (1)$$

where $H_{at}$ is the (hydrogenic) atomic Hamiltonian. The calculations assume an isotropic microcanonical ensemble with classical angular momenta $1 \leqslant L \leqslant 2$ corresponding to the initial laser-excited $l=1$ state with a statistical population of $m$ substates. The experimentally measured HCP profile is used for $F_{HCP}$.

One simulation in Fig. 1 was undertaken assuming a "perfect" strictly periodic HCP train and predicts a pronounced peak in the survival probability at a scaled frequency $\nu_0 \sim 1$, which is somewhat larger than that observed. Furthermore, it exhibits a second local maximum near $\nu_0 \sim 0.5$, which is not visible in these, or earlier, measurements. The second simulation includes the anomalous time separation between the first negative and second positive pulses discussed earlier. Inclusion of this effect leads to pronounced changes in the model predictions. The size of the feature at $\nu_0 \sim 1$ is dramatically reduced and the feature at $\nu_0 \sim 0.5$ essentially disappears. The agreement with experiment, however, is much improved.

The effects of population trapping near the continuum are clearly evident when the evolution of the electron energy distribution is considered as a function of the number, $N$, of

HCP pairs. This is shown in Fig. 2(a) for a scaled frequency $\nu_0=1$ and kick strength $|\Delta p_0|=0.21$. The energy is expressed in scaled units $E_0=n^2E$. As $N$ increases, the distribution first broadens but then breaks into two relatively narrow features, one located near the energy of the parent atoms ($E_0=-0.5$), the other centered near the edge of the continuum at $E=0$. Those electrons in this higher-energy feature having $E<0$ remain bound in states with values of $n$ typically $\gtrsim 800$. The feature near $E_0=-0.5$ is associated with atoms trapped (temporarily) in the cantorus discussed earlier that is associated with the local maximum in survival probability seen in Fig. 1 at $\nu_0 \sim 1$.

Figure 2(b) shows the time evolution of the electron energy distribution for $K(351p)$ atoms when the scaled frequency is increased to $\nu_0=3$. In this case, the distribution first broadens but then subsequently narrows and moves toward the continuum. The motion toward the continuum with increasing $N$, however, is slower than that seen for $\nu_0=1$. This results because at high scaled frequencies the effects of successive HCPs better cancel. The overall survival probability following some fixed number of pulses is, therefore, increased, consistent with the behavior seen in Fig. 1. No feature at energies characteristic of the parent state is evident. This is not unexpected as the initial state lies entirely within the chaotic sea.

Studies were also undertaken using quasi-1D atoms created by photoexciting, in a weak dc electric field ($\sim 250\ \mu\mathrm{V\ cm^{-1}}$), a mix of the lowest-lying red-most states in the $n=350$, $m=0$ Stark manifold [7]. The calculated evolution of the electron energy distribution for such a mix of states when subject to a train of HCPs with $\nu_0=1$ is shown in Fig. 2(c). In these simulations the initial state is represented by a statistical mixture of 36 oriented Stark states centered on the parabolic quantum number $n_1=320$ (see Ref. [7]). Again, as the number of HCPs is increased, the energy distribution becomes localized and trapped near $E_0=0$. No feature is observed at energies in the vicinity of the parent state. This was unexpected as our earlier phase space studies suggested that (quasi-) 1D atoms whose initial phase points overlap the observed cantori should be transiently trapped, resulting in a pronounced feature in the electron energy distribution at $E_0 \sim -0.5$ (see Ref. [3]). This apparent discrepancy was traced to the anomalous time separation between the first negative and second positive HCPs in the HCP train. As illustrated in Fig. 2(c), simulations using a "perfect" HCP train show that even after $N=50$ pulse pairs, a pronounced feature is present at energies close to the parent state.

The model predictions were confirmed experimentally through SFI studies, the results of which are shown in Fig. 3. For reference, Fig. 3(a) shows SFI spectra measured with the laser tuned to excite a number of selected high-$n$ states. SFI spectra recorded for the parent $351p$ state and following application of $N=20$ HCP pairs with $\nu_0=1$ are presented in Fig. 3(b). As predicted by the simulations, after application of the HCP train the SFI spectrum displays two peaks. The small feature seen at late times results from ionization of atoms with values of $n$ close to that of the parent atoms. The larger feature observed at early times corresponds to ionization of atoms in very-high-$n$ states. For these states, the electron orbital period is rather long ($T_n \sim 150$ ns at $n \sim 1000$) and the

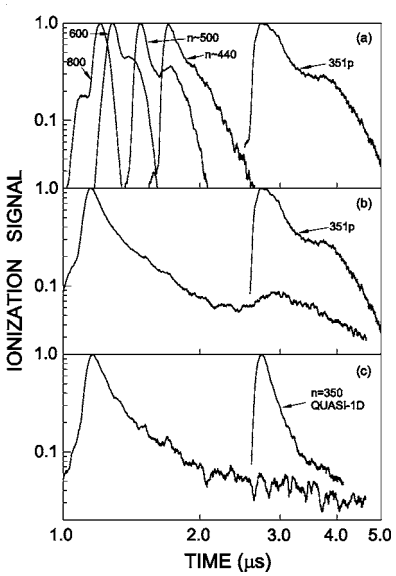


FIG. 3. (a) SFI calibration spectra recorded with no applied dc field and the laser tuned to excite states with the values of $n$ indicated. (b) SFI spectra for parent $351p$ atoms and following application of $N=20$ HCP pairs with $|\Delta p_0|=0.21$ and $\nu_0=1$. (c) Same as in (b) only using quasi-1D $n=350$, $m=0$ atoms. To facilitate comparison the various profiles are normalized to the same peak height.

electron is typically located at large distances ($\gtrsim 50\ \mu$m at $n \sim 1000$) from the Rydberg core ion. The electron thus behaves much as a free electron, which, because the average electric field experienced during the HCP train is zero, gains neither energy nor momentum from the field [6]. It can, therefore, remain trapped close to the continuum for extended periods. The overall Rydberg atom survival probability, however, decreases as $N$ increases because, as seen in Fig. 2(a), the electron energy distribution broadens further into the continuum and an increasing fraction of the atoms are no longer bound. As apparent from Fig. 3(c), no feature associated with the ionization of atoms having values of $n$ close to that of the parent states is observed in the SFI spectrum for quasi-1D atoms. Rather, this comprises a single feature that corresponds to ionization of very-high-$n$ states. This is consistent with the simulations shown in Fig. 2(c).

The present work provides further evidence that when a Rydberg atom is subject to a periodic sequence of HCPs whose net average field is zero the excited electron energy distribution initially moves toward states of higher $n$ but then becomes trapped near the edge of the continuum leading to sizable overall survival probabilities. This stabilization mechanism should be quite general. Indeed, similar behavior has been seen in microwave ionization of Rydberg atoms [8] and in simulations of the behavior of Rydberg states when perturbed by random kicks during transport through solids [9].

This research is supported by the NSF under Grant No. PHY-0353424 and by the Robert A. Welch Foundation. C.O.R. acknowledges support by the OBES, U. S. DOE to Oak Ridge National Laboratory, which is managed by the UT-Batelle LLC under Contract No. DE-AC05, 00OR22725. J.B. and S.Y. acknowledge support from the FWF (Austria). J.C.L. also received support from the ONR.

[1] See, for example, F. B. Dunning, J. C. Lancaster, C. O. Reinhold, S. Yoshida, and J. Burgdörfer, Adv. At., Mol., Opt. Phys. (to be published).
[2] E. Persson, S. Puschkarski, X. M. Tong, and J. Burgdörfer, in *Ultrafast Optics*, edited by F. Krausz (Springer, Berlin, 2004).
[3] B. E. Tannian, C. L. Stokely, F. B. Dunning, C. O. Reinhold, S. Yoshida, and J. Burgdörfer, Phys. Rev. A **62**, 043402 (2000).
[4] W. Zhao, J. C. Lancaster, F. B. Dunning, C. O. Reinhold, and J. Burgdörfer, J. Phys. B **38**, S191 (2005).
[5] S. Yoshida, C. O. Reinhold, J. Burgdörfer, W. Zhao, J. J. Mestayer, J. C. Lancaster, and F. B. Dunning, Phys. Rev. A (to be published).
[6] T. Grozdanov and H. S. Taylor, J. Phys. B **20**, 3683 (1987).
[7] C. L. Stokely, J. C. Lancaster, F. B. Dunning, D. Arbo, C. O. Reinhold, and J. Burgdörfer, Phys. Rev. A **67**, 013403 (2003).
[8] M. W. Noel, W. M. Griffith, and T. F. Gallagher, Phys. Rev. Lett. **83**, 1747 (1999).
[9] C. O. Reinhold, J. Burgdörfer, J. Kemmler, and P. Koschar, Phys. Rev. A **45**, R2655 (1992).