STUDYING ATOMIC DYNAMICS WITH DESIGNER PULSES

C. O. Reinhold and J. Burgdörfer
Oak Ridge National Laboratory*, Oak Ridge, TN 37831
Dept. of Physics & Astronomy, University of Tennessee, Knoxville, TN 37996-1200

M. T. Frey and F. B. Dunning
Dept. of Physics and Rice Quantum Institute, Rice University, 6100 S. Main St.
Houston, TX 77005-1892

to be published in proceedings of

XX International Conference on the Physics of Electronic and Atomic Collisions
Vienna, Austria
July 23-29, 1997

World Scientific Publishing Company

*Managed by Lockheed Martin Energy Research Corp. under contract DE-AC05-96OR22464 with the U.S.D.O.E.

"The submitted manuscript has been authored by a contractor of the U.S. Government under contract No. DE-AC05-96OR22464. Accordingly, the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes."
This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, make any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.
STUDYING ATOMIC DYNAMICS WITH DESIGNER PULSES

C O REINHOLD, J BURGDÖRFER
Oak Ridge National Laboratory, Oak Ridge, TN 37831-6375, USA and University of Tennessee, Department of Physics and Astronomy, Knoxville, TN 37996-1200, USA

M T FREY, F B DUNNING
Dept. of Physics and the Rice Quantum Institute, Rice University, 6100 S. Main Street, Houston, TX 77005-1892, USA

We present a brief review of recent experimental and theoretical progress on the dynamics of Rydberg atoms using short half-cycle pulses. We discuss new possibilities in coherent control and non-linear dynamics of atoms which have lately become possible using various superpositions of such pulses.

1 Introduction

In the past few years, the ionization and excitation of Rydberg atoms by "half-cycle" pulses (HCPs) has been investigated extensively. In contrast to short laser pulses which extend over several optical cycles, HCPs are characterized by a strong unidirectional electric field $F(t)$ confined to a short time interval, $T_p$, corresponding to only one half of a cycle. These characteristics make HCPs very similar to the transverse electric field pulse experienced by an atom in a fast ion-atom collision.

Experiments have recently reached the regime in which the effective duration of the pulses, $T_p$, and the peak fields, $F_p$, are of the order of the classical electron orbital period, $T_{ni} = 2\pi n_i^3$, and the Coulomb electric field, $F_{ni} = n_i^{-4}$ in the atom, respectively (where $n_i$ is the initial principal quantum number and atomic units are used throughout)\(^1,2,3,4,5\). This has been accomplished, for example, by using subpicosecond pulses and moderately low $n_i \approx 30$ Rydberg levels\(^1\) or, alternatively, by using nanosecond pulses and very high $n_i \sim 400$ states\(^3\). These experimental advances have stimulated a large number of theoretical investigations using quantum and classical approaches\(^6,7,8\).

The effect of a single HCP on an atom differs significantly from that of an ultrashort laser pulse because the integral of the applied electric field is finite. If the field varies very slowly as a function of time, ionization results primarily from escape over the potential barrier generated by the atomic and applied fields. Whether or not ionization occurs is determined by the ratio $F_p/F_{ni}$ (the ionization threshold is proportional to $n_i^{-4}$). In contrast, application of a very short electric field pulse with duration $T_p \ll T_{ni}$ lowers the Coulomb barrier only momentarily. In this limit, the HCP simply delivers a "kick" or
impulsive momentum transfer

\[ \Delta \vec{p} = -\int_{-\infty}^{\infty} \vec{F}(t) \, dt \] (1)

leading to an energy transfer (i.e. change in electron kinetic energy)

\[ \Delta E = \frac{(\vec{p} + \Delta \vec{p})^2}{2} - \frac{\vec{p}^2}{2} = \frac{(\Delta \vec{p})^2}{2} + \vec{p} \cdot \Delta \vec{p} \] (2)

where \( \vec{p} \) is the momentum of the electron prior to the kick. Ionization can still occur if \( \Delta E \) is greater than the original binding energy. The momentum transfer required to induce ionization in the short pulse regime scales as \( n_i^{-1} \) (i.e. as the orbital momentum of the electron \( p_n = n_i^{-1} \)).

Very recently, experiments using "designer" pulses have become feasible: i.e. trains of HCPs which can be shaped at will\(^9,10,11,12\). Such pulses can be used to mimic various processes taking place in fast ion-atom, ion-molecule and ion-solid interactions. In addition, they provide an excellent tool to study problems of fundamental interest within other areas of atomic physics. In this work we discuss two examples which illustrate the utility of designer pulses in (i) producing and probing coherent atomic states and (ii) investigating non-linear atomic dynamics. In the following we assume \( \Delta \vec{p} = \Delta \vec{F} \). Details about our calculations and experimental setup can be found elsewhere\(^6,3,10,11,12\).

2 Creating and probing momentum wavepackets

The interest in creating coherent atomic states dates back to early attempts by Schrödinger\(^13\) to construct a non-dispersive minimum uncertainty wavepacket similar to coherent oscillator eigenstates. The latter follow Newton's laws of classical mechanics while the width in position and momentum remain at the minimum consistent with the Heisenberg uncertainty principle. Early attempts using electronic states in atoms failed because of the non-equidistant spacing of energy levels, which leads to rapid dephasing. Subsequently, the production of a more general class of wavepackets that consist of a dispersive coherent superposition of atomic states and do not satisfy the minimum uncertainty condition has been studied. Only in recent years has it become possible to generate transient quasi-classical wavepackets which remain well localized for a long period of time. This has been accomplished by using Rydberg states with large principal quantum numbers, \( n \), for which the deviation from equidistant levels is small (decreasing as \( n^{-1} \)) and for which the wavepacket evolution resembles that of the harmonic oscillator\(^16,17,10,11,9\).
Consider an electron in an atom whose dynamics is governed by a time-independent atomic Hamiltonian $H_{at}$ with orthonormal eigenvectors $|\chi_{\alpha}\rangle$ and eigenenergies $\epsilon_{\alpha}$. In general, the wavefunction of the electron may be written as a superposition of the $|\chi_{\alpha}\rangle$ and evolves in time as

$$|\Psi(t)\rangle = \sum_{\alpha} e^{-i\epsilon_{\alpha}t} \langle \chi_{\alpha} | \Psi(0) \rangle |\chi_{\alpha}\rangle.$$  

which is a "fully coherent" wavepacket (i.e. a pure state at each instant of time). The mean value of an observable $\hat{O}$ is given by

$$\langle \hat{O} \rangle = \sum_{\alpha,\beta} e^{-i(\epsilon_{\alpha} - \epsilon_{\beta})t} \langle \chi_{\alpha} | \Psi(0) \rangle \langle \Psi(0) | \chi_{\beta} \rangle \langle \chi_{\beta} | \hat{O} |\chi_{\alpha}\rangle$$

and oscillates in time with frequencies $\omega_{\alpha,\beta} = \epsilon_{\alpha} - \epsilon_{\beta}$, usually referred to as "quantum beats".

A wavepacket can be created through a time-dependent perturbation which acts as a "pump". Subsequently, the quantum beat frequency $\omega_{k,j}$ can be observed using another time-dependent perturbation (a "probe") with a time duration $T_p < (2\pi/\omega_{k,j})$. Such a scheme can be devised using, for example, two short HCPs. Consider the application of an ultrashort "pump" HCP to a stationary Rydberg state $|\phi_i\rangle = |n_i l_i m_i\rangle$ with well defined spherical quantum numbers $n_i, l_i, m_i$. The resulting coherent wavefunction just after the pulse

$$|\Psi(t = 0)\rangle = |\phi^P_i\rangle = e^{i\Delta E_{i}\phi} |\phi_i\rangle$$

and corresponds to a Galilei boosted initial state shifted in momentum space by $\Delta\phi$. Figure 1 displays the excitation function associated with various states with $n_i = 417$ after application of a boost. This energy distribution determines the relative weight of the time evolution phases $e^{-i\epsilon_{\alpha}t}$ in Eq. 3. Calculations employing the classical trajectory Monte Carlo (CTMC) approach are included for both an ultrashort pulse ($T_p/T_{n_i} = 0$) and a pulse of $\sim 2$ ns duration ($T_p/T_{n_i} = 0.18$) as used in experiments. The HCP leads to population of a range of higher $n$ states centered around $n_f = \sqrt{-2\langle E\rangle_{t=0}} \approx 493$ with a narrow width $\Delta n \approx 20$. The peak of the distribution is found at

$$\epsilon_{n_f} \approx \langle E\rangle_{t=0} = \langle \phi_i | H_{at} |\phi_i\rangle + \langle \phi_i | \Delta E |\phi_i\rangle$$
where $\Delta E$ is given by Eq. 2. Note that $\langle nlm|\vec{p}|nml \rangle = 0$ and, therefore, $\langle E \rangle_{t=0} = \epsilon_{n_i} + (\Delta p)^2 / 2$ where $\epsilon_{n_i}$ is the energy of the initial Rydberg state.

The peak in the energy distribution is completely equivalent to the well-known Bethe ridge in atomic collision physics\(^{14}\). Remarkably, this structure also exists in the density of bound states after a short HCP. The width of the Bethe ridge is determined by the linear term $\vec{p} \cdot \Delta \vec{p}$ in equation 2 and depends on the quantum number (and the geometry) of the initial state. This width determines the width $\Delta n$ of the "wavepacket". Since $1 \ll \Delta n < n_f$, the convergence of the quantum evolution (Eq. 3) to the classical limit can be treated in detail.

The wavepacket generated by a short HCP can be understood in terms of the time evolution of the momentum of the electron (Fig. 2) which is initially boosted to $\langle p_x \rangle_{t=0} = \Delta p$. Subsequently, $\langle p_x \rangle$ beats in time with the classical orbital frequency $\omega_{n_f}$ (i.e. with period $T_{n_f} = 2\pi / \omega_{n_f}$). On general grounds\(^{15}\), one expects the quantum and the classical evolution to depart from each
other at a characteristic "break" time $t_c \simeq 2\pi/\Delta \epsilon$, where $\Delta \epsilon$ is the smallest level spacing (i.e. $t_c \sim T_{nm}$). Remarkably, classical-quantum correspondence extends for times beyond $t_c$ (see Fig. 2). This extended classical-quantum correspondence can be traced to the particular properties of the Coulomb-Kepler problem and of the HCP-generated wavepackets. The energy differences in the Rydberg series determining quantum beats are given to second order in $\delta n/n_f$ by

$$\epsilon_{n_f+\delta n} - \epsilon_{n_f} \simeq \delta n \omega_{n_f} \left[ 1 - \frac{3}{2} \frac{\delta n}{n_f} + 2 \left( \frac{\delta n}{n_f} \right)^2 \right]$$

(7)

where $|\delta n| \leq \Delta n/2$. To leading order, the spectrum compares locally to that of a harmonic quantum oscillator, with $\omega_{n_f} = n_f^{-3}$ being the classical orbital frequency. For a harmonic oscillator, quantum and classical expectation values agree. The damping of the beats is caused by the "anharmonic" correction in Eq. 7. The fact that classical dynamics can reproduce the dephasing occurs because, when $\Delta n$ is large, the approximation of a large but discrete set by a continuous distribution is valid. Classical dynamics will fail, however, at times approaching the revival time $t_R \simeq (n_f/3)T_{nm}$ of the wavepacket. 

Figure 2: Time development (expressed in units of $T_{nm}$) of the scaled expectation value $\langle p_z \rangle /p_{n_i} = n_i(p_z)$ following application of a HCP to hydrogen atoms in the $100s$ state. The scaled momentum transfer $\Delta p/p_{n_i} = 0.53$.

The evolution of such a classical wavepacket, i.e. of the phase space density which initially mimics a quantum circular state is illustrated in Figure 3. At $t = 0$ the wavepacket is well localized in the $(\rho, z)$ plane ($\rho = \sqrt{x^2 + y^2}$) and the HCP delivers a kick towards the positive $z$ axis. For $t > 0$ the wavepacket approximately follows the trajectory of an electron initially in a circular orbit contained in the $(x, y)$ plane. The spreading of the wavepacket originates in the energy distribution (Fig. 1). At $t = T_{nm}/2$ the wavepacket is moving towards
the $z < 0$ direction ($p_z < 0$). If a second HCP with $\Delta p > 0$ is applied at this time, the linear $\Delta p$ contribution to the energy transfer in Eq. 2 tends to decrease the final binding energy and stabilizes the atom. Similarly, for a second HCP with $\Delta p > 0$ applied at $t = T_{n_f}$, the two sequential momentum transfers $\Delta p$ add up leading to an increase of energy and an enhanced ionization probability.

Figure 3: Time evolution of a circular state, $H(n_i = 417, l_i = m_i = 416)$, subject at $t = 0$ to an ultrashort HCP with $\Delta p_0 = 0.53$. The dots represent a scatter plot of the probability density of the classical wavepacket. The thick gray line is the trajectory followed by an electron in a perfectly circular orbit initially in the $(x, y)$ plane. The cylindrical coordinates of the electron are scaled to the initial orbital radius: i.e., $z_0 = z_0/n_i^2$, $p_0 = p_0/n_i^2$.

A comparison of calculated and measured survival probabilities of K(417p) is shown in Fig. 4 as a function of the time delay between the pump HCP and a probe HCP. The survival probability oscillates as a function of the time delay between the two HCPs reflecting the time evolution of the wavepacket which, in turn, follows the time development of the expectation value of the $z$ component, $p_z$, of momentum of the electron after the first HCP (see ($p_z$) on the right hand axis). The experimental data agree well with the calculations.
3 Dynamical stabilization of the "kicked" Rydberg atom

The interaction of Rydberg atoms with a sequence of many half-cycle pulses opens up the opportunity to experimentally and theoretically study the non-linear dynamics of the "kicked Rydberg atom". The kicked atom together with its counterpart, the kicked rotor, represent the two fundamental model systems for irregular classical dynamics in periodically perturbed Hamiltonian systems. The time evolution for kicked systems reduces to a sequence of discrete maps between adjacent kicks. This simplification permits detailed numerical studies of the long-term evolution using both classical and quantum dynamics, and hence, of the classical-quantum correspondence in microscopic systems that feature regular and chaotic dynamics.

Figure 5 displays the first experimental and theoretical evidence for the dynamical stabilization of a periodically kicked Rydberg atom. The figure shows the survival probability measured following application of 10 and 50 HCPs of 50 mV/cm amplitude as a function of the frequency, $\nu_p$, of the train of pulses (i.e. the time interval between adjacent pulses). As expected, the application of more HCPs leads to a decrease in the survival probability. The size of the decrease, however, depends sensitively on $\nu_p$ and a pronounced maximum in the survival probability is evident at scaled frequencies $\nu_0 = 2\pi n^2 \nu_p \geq 1$ for 50 HCPs. The agreement between the experiment and the CTMC simulation indicates that the stabilization signalled by the increased survival probability for $\nu_0 \geq 1$ is classical in origin. Note that in the limit of high frequencies, indicated by the horizontal arrow, the train of pulses becomes equivalent to a
Figure 5: Rydberg atom survival probability following application of ten and fifty HCPs with a peak field of 50 mV/cm as a function of the pulse repetition frequency. Experimental data for K(388p), ■, results of CTMC calculations for K(388p), —— CTMC results for the impulsively driven H(388p) atom with $n_p \Delta \nu = 0.3$, ———. The pulse repetition frequency is displayed in MHz and in scaled units $\nu_0 = \nu_p 2\pi n_p^2$. Vertical arrows indicate positions of stable islands for the kicked hydrogen atom (see text). The horizontal arrow marks the high frequency limit for ten pulses.

In order to examine the origin of the broad maximum in Fig. 5 we have studied a simplified model of hydrogen atoms subject to a sequence of $\delta$-function impulses. The corresponding Hamiltonian is

$$H = H_{at} - z \Delta \nu \sum_{j=1}^{N} \delta(t - j \nu_p^{-1}).$$

(8)

The evolution governed by this Hamiltonian during one period of the perturbation is given by a map of phase space coordinates

$$M(\vec{r}, \vec{p}) = \underbrace{M_{Coul}} \circ \underbrace{M_{\Delta \nu}}(\vec{r}, \vec{p})$$

(9)

where $M_{\Delta \nu}$ describes the kick and $M_{Coul}$ describes the evolution in the Coulomb orbit. The evolution of the phase space coordinates after a sequence of $N$ pulses is given by $M^N(\vec{r}, \vec{p})$. For this simplified system it is computationally
feasible to investigate the long-term stability in the limit $N \to \infty$ (typically $N \geq 10^6$) and to perform a detailed analysis of the classical phase space. As demonstrated in Fig. 5, the simplified model reproduces the observed general structure in the survival probability although the stability is somewhat reduced. This results because the the finite width of each experimental HCP cuts off the higher frequencies in the perturbation.

For a large number $N$ of kicks, the maximum in the survival probability is found to comprise a series of overlapping peaks. For reference, those at scaled frequencies $\nu_0 \sim 1.3, 0.6, 2.1$ are marked with arrows in Fig. 5 . The maximum is therefore due to a large, only marginally unstable, region of phase space which has stable islands at its core. Figure 6, which shows calculated survival probabilities as a function of $N$ for several values of $\nu_0$, demonstrates that for frequencies within the maximum, atoms can be either dynamically stable or unstable. Within the numerical accuracy, for $\nu_0 \sim 1.3$ about 3% of the Monte Carlo ensemble of trajectories with angular momentum $l = 1$ remain stable for more than $10^6$ kicks. In fact, depending on the initial state of the electron, stabilization probabilities much larger than those by Fig. 6 can be obtained. This is especially true for blueshifted and for nearly circular states ($L = L_z$).

We have identified various stable islands by Poincare surface of sections for slices in phase space $^12$. The identification of the periodic orbits around which the stable islands are organized proceeds by noting that the binding energy must be unchanged following some integer number $k$ of applied kicks,
i.e. $M_k(E) = E$. For $k = 1$, this implies $\Delta E = 0$ in Eq. 2 or, equivalently, $p_z = -\Delta p/2$. The periodic orbit which corresponds to the center of the main stable island is indeed found at this $p_z$ value and can be identified with help of its projection onto the $(z, p_z)$ plane, as shown in Fig. 7. The trajectory has been followed for 20 kicks involving segments of 20 Coulomb orbits having the same binding energy but with different angular momenta. In the $(z, p_z)$ plane and for $\rho \ll |z|$, all segments appear to trace each other. They can, however, be resolved using a projection onto the $(z, \rho)$ plane where one obtains a picture resembling an orbit precession in a weak DC electric field (i.e. a Stark orbit). This "DC" field is provided by the time-averaged field of the train of pulses ($F_{DC} = \nu_p \Delta p$).

![Figure 7: Projection of a stable orbit onto the $(z, p_z)$ plane (solid line) starting with $L_z = 2.5 \times 10^{-4}$, $\rho = 0.1$, $p_p = 0$ for $\nu_0 = 1.26$, $\Delta p_0 = 0.32$, $p_z = -\Delta p/2$, $z = 2.27$. The dashed line indicates the segment of the Coulomb orbit that is not reached by the electron.](image)

A kick maps a point $P = (z, p_z = -\Delta p/2)$ in a given Kepler orbit onto a point $Q = (z, p_z = \Delta p/2)$ of another orbit, as illustrated by the vertical arrow in Fig. 7. The subsequent unperturbed Coulomb evolution transports the point $Q$ along the direction of the arrow back to point $P$, $M_{coul}(Q) \simeq P$. The periodic orbit is therefore nothing but a sequence of segments of full Coulomb orbits cut short by the kick. For this to occur, the time separation $T_s$ between kicks (pulses) must be less than the Kepler period, i.e., $T_s = \alpha T_n$, with $\alpha < 1$. (At the maximum in Fig. 5, $\nu_0 = \alpha^{-1} \sim 1.3$). Fig. 7 suggests the existence of a family of stable periodic orbits with scaled frequencies $\nu^*_s = (\alpha + s)^{-1}$ ($s = 0, 1, 2, \ldots$) corresponding to orbits that complete $s$ full periods and one segment prior to the next kick mapping $P$ onto $Q$. For $\nu_0^* \sim 1.3$, the scaled frequencies corresponding to $s = 1, 2, 3\ldots$ are $\nu_0 \sim 0.57, 0.35, 0.26\ldots$ and agree well with positions of the peaks in the calculated survival probability in Fig 5.
4 Summary

Half-cycle pulses provide a direct correspondence between atomic collisions and atoms in strong electromagnetic fields, as clearly evidenced by recent work using fast highly charged ions and recoil-ion momentum spectroscopy. In fact, fast ions provide the strongest and shortest form of half-cycle pulses which can be used to multiply ionize atoms in the ground state.

The availability of trains of well defined half-cycle pulses which strongly perturb Rydberg atoms opens new possibilities in atomic physics. Well controlled atomic manipulation and experimental realization of the "kicked atom" on a microscale where quantum effects should modify the long-time evolution have been, up to recently, difficult. Study of these problems affords the opportunity to analyze the correspondence between the classical and quantum dynamics from a new point of view and in great detail. One goal of future studies will be to determine possible signatures of the breakdown of classical-quantum correspondence and of quantum modification of irregular classical dynamics in the experimental ionization signal. Another possibility could be using long-lived Stark beats as a tool for investigating elastic electron-molecule scattering at electron energies down to a few microelectronvolts.

Acknowledgments

The experimental work was supported by the NSF and the Robert A. Welch Foundation. The theoretical work was funded by the U. S. DoE, OBES, Div. of Chem. Sciences, under Contract No. DE-AC05-96OR22464 with ORNL managed by LMERC and by the NSF. It is a pleasure to acknowledge the assistance of B. Tannian in data acquisition.

References

13. E. Schrödinger, Naturwissenschaften 14, 664 (1926).
20. See, e.g. J. Ullrich, these proceedings.
Report Number (14) ORNL/CP-4784
CONF-970710

Publ. Date (11) 199709

Sponsor Code (18) DOE/ER, NSF, XF

UC Category (19) UC-400; UC-000, DOE/ER