Transporting Rydberg Electron Wave Packets with Chirped Trains of Pulses

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A protocol for steering Rydberg electrons towards targeted final states is realized with the aid of a chirped train of half-cycle pulses (HCPs). Its novel capabilities are demonstrated experimentally by transporting potassium atoms excited to the lowest-lying quasi-one-dimensional states in the $n_i = 350$ Stark manifold to a narrow range of much higher-n states. We demonstrate that this coherent state transfer is, to a high degree, reversible. The protocol allows for remarkable selectivity and is highly efficient, with typically over 80% of the parent atoms surviving the HCP sequence.

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Engineering the quantum states of microscopic and mesoscopic objects is a goal in many current fields of research ranging from nanomechanical devices to quantum information processing. Atoms in high-lying Rydberg states with large values of principal quantum number n provide a valuable laboratory in which to explore, control, and manipulate quantum states of mesoscopic size. Relatively recent additions to the arsenal of electromagnetic pulses available for controlling such states are carefully tailored sequences of half-cycle pulses (HCPs) [1,2]. Atoms react very differently to HCPs than to laser or microwave pulses. Here we demonstrate the remarkable control that can be achieved using a *chirped* train of HCPs.

Localized quantum states can be formed by constructing wave packets made up of a coherent superposition of stationary states [3]. Such localization in free atoms is, however, transient because the energy levels are not equispaced, leading to dephasing. Localized nondispersive wave packets, however, can be created and ''trapped'' with the help of a periodic external driving field [4,5]. Trapping results because the presence of the driving field gives rise to sizable stable islands in an otherwise chaotic phase space from which the wave packet cannot escape. The positions of these islands depend on the amplitude and period of the driving field. Recent theoretical work [6] suggests that by adiabatically varying the frequency of a HCP train a localized and trapped wave packet can be transported to preselected regions of phase space. Here we report the first experimental realization of such coherent transfer by a *chirped*, i.e., frequency-modulated, sequence of HCPs. Localized Rydberg wave packets with $n \simeq 350$ are transported to a narrow distribution of very-high-n Rydberg states with $n \simeq 700$ and then returned to $n \simeq 350$. This range of n exceeds that achieved using chirped microwave radiation [7]. The present approach improves on earlier HCP protocols devised to transport atoms to high-n states [8] in that it produces a much narrower distribution of final n states, better preserves wave packet localization, is remarkably efficient, and is reversible.

The Heisenberg quantum break time, τ_H, beyond which classical and quantum dynamics diverge increases rapidly with n ($\tau_H \propto n^4$) and reaches the microsecond regime by $n \sim 400$. This allows protocols to coherently control the motion of electron wave packets to be developed based on considerations of classical dynamics [4,6,7]. The response of a Rydberg atom to a train of N identical HCPs with profile $F_{\mathrm{HCP}}(t)$ directed along the z axis is governed by the Hamiltonian

$$H(t) = \frac{p^2}{2} - \frac{1}{r} + z \sum_{j=1}^{N} F_{\mathrm{HCP}}(t - t_j), \tag{1}$$

where $\vec{r}$ and $\vec{p}$ are the position and momentum of the Rydberg electron. Each HCP comprises a pulsed unidirectional electric field of duration $T_p \ll T_n$, where T_n ($= 2\pi n^3$ a.u.) is the electron orbital period. (Unless otherwise noted, atomic units are used throughout.) In this limit each HCP simply delivers an impulsive momentum transfer or ''kick'', $\Delta p = -\int F_{\mathrm{HCP}}(t)dt$, to the excited electron. Within the framework of the classical trajectory Monte Carlo (CTMC) method, Hamilton's equations of motion for Eq. (1) are solved for an ensemble of electrons representing the initial wave packet.

Consider initially a train of pulses with fixed period $T = t_{j+1} - t_j$ (frequency $\nu = 1/T$) such that $F_{\mathrm{HCP}}(t - t_j) \simeq -\Delta p \delta(t - jT)$. Under such periodic driving atoms display a mixture of regular and chaotic dynamics [1] that can be visualized using Poincaré surfaces of section (see Fig. 1). (All CTMC simulations of wave packet evolution employ the experimentally measured HCP profiles rather than δ-function kicks.) A series of stable islands are evident embedded in a chaotic sea. Electrons whose initial phase space points lie in a stable island remain trapped therein forming a nondispersive wave packet [4,5]. Those whose initial phase points reside in the chaotic sea undergo rapid diffusion to the continuum. We focus here on the largest (period-one) island which is the most effective in trapping electrons. For an atom to survive many kicks the electron

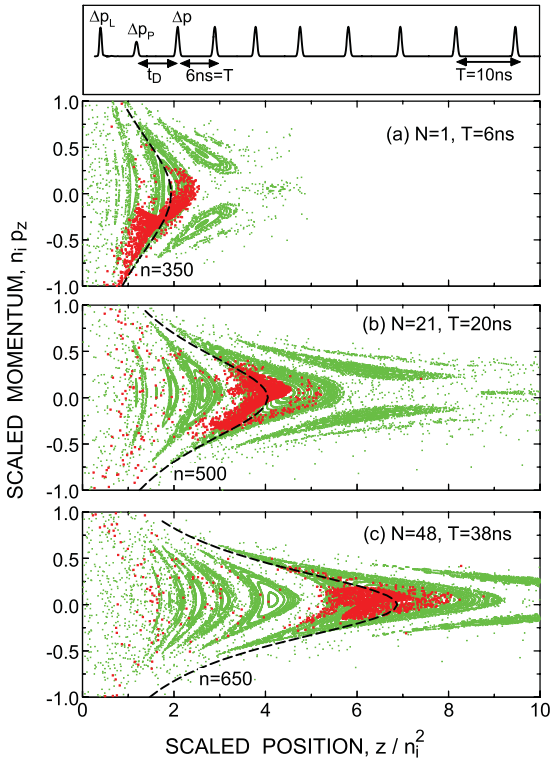

FIG. 1 (color online). Upper panel: profile of applied HCP sequence. The pulse spacing in the chirped train is incremented linearly as $t_{j+1} - t_j = 6$ ns + 0.67$(j - 1)$ ns. Lower panels: Dark points, phase space distribution of the present wave packet, projected on the z, p_z plane, immediately before the 1st, 21st, and 48th kick in the chirped HCP train. The axes are labeled in scaled units. Light points, corresponding Poincaré surfaces of section for the 3D kicked hydrogen atom with $T = 6$, $T = 20$, and $T = 38$ ns, and δ-function kicks with scaled strength $\Delta p_0 = n_i \Delta p = -0.1$. The cuts are taken immediately before each kick at $\rho_0 = \rho/n_i^2 \sim 0.5 \pm 0.2$, $p_{\rho_0} = n_i p_\rho \sim 0 \pm 0.1$. The dashed lines indicate stationary tori corresponding to states with $n = 350$, 500, and 650 ($\rho_0 = 0.5$, $p_\rho = 0$).

energy must be little changed following each kick, i.e., the z component of the electron momentum immediately before the kick must fulfill the condition $p_{zi} = -\Delta p/2$, whereupon its value immediately after the kick becomes $p_{zf} = +\Delta p/2 = -p_{zi}$, i.e., p_z simply reverses sign without any change in the electron kinetic energy. p_z must then evolve through orbital motion such that it again becomes $\sim -\Delta p/2$ immediately before the next kick. Near the center of the dominant island the electron motion is a series of segments of a Coulomb orbit cut short by a kick, the period of the orbit T_n being tied to that of the HCP train, T (a time slice of such a periodic orbit is visualized in Fig. 1(a) as a point around $(z_0, p_{z0}) = (z/n^2, p_z n) \simeq (2, -\Delta p_0/2)$). If the period T is changed, the position of the islands is also changed. If the rate of change is sufficiently slow a wave packet initially localized in the island can respond adiabatically and remain trapped in the island as it "moves", thereby allowing it to be transported to a different location in phase space, i.e., to a different location in coordinate space, z, and to different quantum numbers n. The adiabaticity criterion [9] suggests that this can be accomplished if $\delta T \ll T$, where the chirp δT is the change in the time interval between adjacent kicks. This is demonstrated in Fig. 1, where a classical ensemble of phase space points that simulates the wave packet created here is transported from $z_0 \simeq 1.8$ to $z_0 \simeq 6.5$. The spacing between adjacent pulses is incremented linearly as $t_{j+1} - t_j = 6$ ns + $(j - 1)\delta T$ using $\delta T = 0.67$ ns $\ll T_{n_i}$.

The efficiency of the present protocol is maximized by carefully loading the island starting with a quasi-one-dimensional (quasi-1D) state localized along the z axis. The pulse sequence, shown in Fig. 1, comprises a "localizing" HCP Δp_L, a "positioning" HCP Δp_P, and a chirped train of N identical HCPs. The first localizing HCP provides a kick of scaled strength $\Delta p_{L0} = n_i \Delta p_L = -0.085$ directed towards the nucleus. (A pulsed field in the $+z$ direction delivers a kick in the $-z$ direction.) This creates a wave packet which after a time delay of $\sim$6.5 ns ($\sim T_{n_i}$) undergoes strong transient localization [10] into a region of phase space located near the outer classical turning point $((z, p_z) \sim (2n_i^2, 0))$ that is smaller than the dominant island. The positioning pulse, Δp_P (applied at the time of optimum localization) is used, in conjunction with the time delay t_D (see Fig. 1) to optimally position the wave packet within the island [8] at the start of the chirped HCP train. The values employed ($\Delta p_{P0} = n_i \Delta p_P = -0.05$, $\tau_D = 7.4$ ns) were optimized through both simulation and experiment.

The chirped HCP train was provided by a programmable pulse pattern generator (PPPG) that divides time into a series of bins and in each outputs a voltage of 0 or V. The bin width was set at 0.67ns and V was chosen to deliver a scaled impulse $\Delta p_0 = n_i \Delta p = -0.1$. The electron is initially in a stationary quasi-1D Rydberg state [11] created by photoexciting potassium atoms to the lowest-lying, red-shifted states in the $n_i = 350$ Stark manifold in a weak ($\sim$250 μV cm^{-1}) dc field directed along the z axis. The atoms are initially oriented along the $+z$ axis. They are then subject to the HCP sequence which was produced by using a combiner to superpose the output of the PPPG with those of other fast pulse generators and applying the resulting waveform to a nearby electrode. The number of atoms that survive, and their excited state distribution, is determined by selective field ionization (SFI) in which a slowly-varying ramped electric field is applied to the atoms and the liberated electrons detected. Since atoms in different n states ionize at different fields, i.e., at different times during the ramp, measurement of the electron arrival time distribution provides a measure of the initial excited state distribution.

The calculated evolution of the electron position, momentum and energy distributions during the chirped train is shown in Fig. 2. Even though the initial chirp rate $\delta T/T$ is sizeable ($\sim$0.1), the overlap between the islands associated

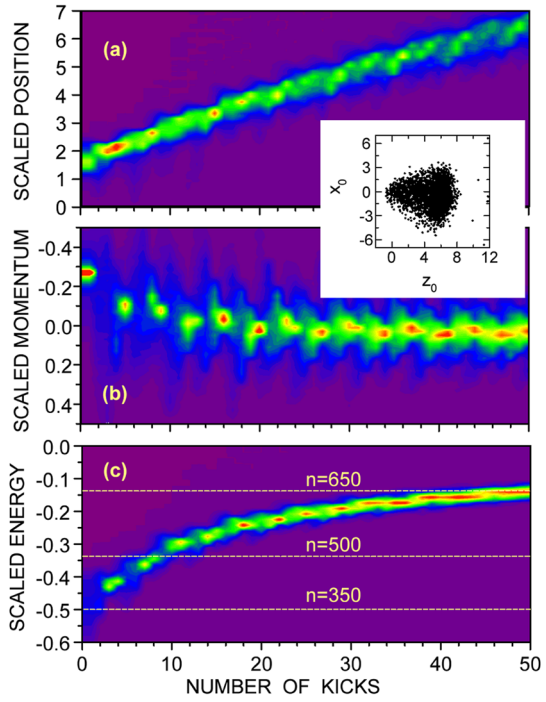

FIG. 2 (color online). Calculated evolution of the position (a), momentum (b), and electron energy (c) distributions (expressed in scaled units $z_0 \equiv z/n_i^2$, $p_{z0} \equiv n_i p_z$, $E_0 \equiv n_i^2 E$) immediately before each kick for quasi-1D $n_i = 350$ atoms subject to the train of linearly chirped HCPs shown in Fig. 1. Three n values are indicated in (c) by horizontal lines. The inset shows the final electron spatial distribution following 50 HCPs projected onto the xz plane.

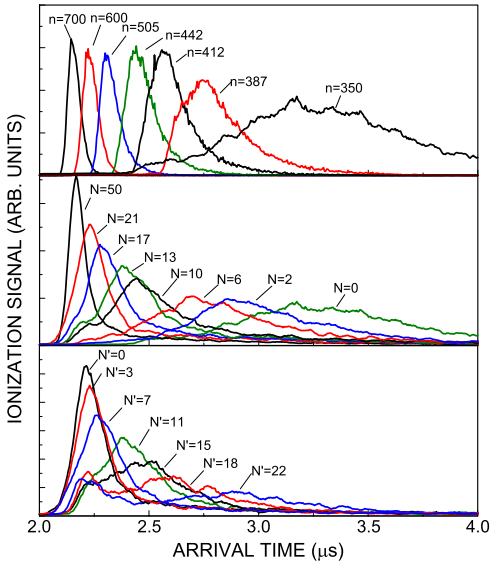

FIG. 3 (color online). (a) SFI calibration spectra recorded with no applied dc field and the laser tuned to excite states with the values of n indicated. (b) SFI spectra recorded following application of the numbers N of HCPs indicated in the down-chirped train shown in Fig. 1. (c) SFI spectra recorded following application of the number N' of up-chirped HCPs indicated (see text).

with successive pulse spacings is sufficient to allow the electron wave packet to remain trapped moving linearly up in z as N, and the pulse separation, increases. The final two-dimensional spatial distribution following 50 HCPs is shown in the inset in Fig. 2. This was calculated 0.5 μs after the last HCP to allow the product wave packet to reach its quasistationary state. Remarkably, the final state is nearly as strongly polarized as is the initial quasi-1D state. As N increases, the atom also moves steadily up in n but the electron energy and momentum distributions remain narrow [see Figs. 2(b) and 2(c)]. More than 95% of the parent atoms are predicted to survive the HCP sequence and be transported to very-high-n states.

The calculated energy distribution following 50 HCPs is strongly peaked at $n \sim 660$ with a width at half height of $\Delta n \sim \pm 30$, which is significantly narrower than those found for previous HCP protocols [8]. SFI studies confirmed this behavior. For reference, Fig. 3(a) shows SFI spectra recorded with no applied dc field and the photoexcitation laser tuned to produce selected high-n states. As expected, as n is increased the spectra move towards earlier times, i.e., ionization occurs at lower fields. SFI spectra obtained following application of different numbers of HCPs are presented in Fig. 3(b). These move steadily towards earlier times as N increases, consistent with transfer to higher-n states. Their widths remain comparable to those seen following excitation of single Rydberg states pointing to a narrow final n distribution. By extending the chirp to larger values of T it should, in principle, be possible to generate localized groups of states with arbitrarily high n. This could not be verified experimentally as stray fields and field inhomogenities in the apparatus begin to ionize atoms with values of n above ~800–900. For large N a small tail is evident on the SFI features that extends to later times, indicating that a small fraction of the parent atoms escape the island. Such escape could account for the measured overall survival probability, typically ~80%–90%, being somewhat lower than predicted by simulation and can be attributed to uncertainties in the alignment of the initial states, i.e., in the orientation of the initial Stark field, induced by stray fields.

Coherent state manipulation using a chirped HCP train is also reversible. This was demonstrated by first driving parent $n_i = 350$ Stark states to higher-n ($n \sim 540$) states using the first 25 HCPs in the "down-chirped" train shown in Fig. 1. This was followed by 10 pulses with constant separation $T = 22$ ns before applying 25 "up-chirped" HCPs with linearly decreasing separation ($\delta T = -0.67$ ns). The calculated evolution of the electron energy distribution during this pulse sequence is shown in Fig. 4. As the pulse separation decreases the wave packet is driven steadily towards states of lower n, but remains narrow in energy. Overall, more than 90% of the parent atoms survive the pulse sequence. This reduction in n was confirmed by SFI studies [see Fig. 3(c)]. Towards the end of the pulse train, however, an early-time SFI feature becomes apparent indicating that some atoms are left in very-high-n states.

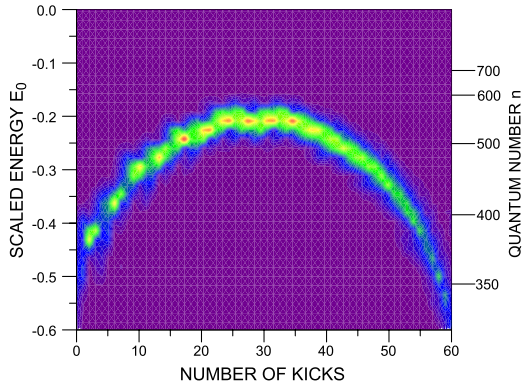

FIG. 4 (color online). Calculated evolution of the electron energy distribution for quasi-1D $n_i = 350$ atoms subject to a chirped HCP train in which the pulse separation is linearly increased for the first 25 HCPs, held constant for 10 HCPs, and then linearly decreased (see text). The spacing between the first two pulses was 6 ns and was incremented in units of ± 0.67 ns.

For low-n states ($n < 450$) the SFI profile after $N' = 22$ is similar to that for $N = 2$ (the time-reversed partner would be $N = 3$). Overall, approximately 70%–80% of the parent atoms are stable against ionization during the entire pulse sequence, ~70% of which return to $n < 450$ states (i.e., the combined return probability is ~50%).

The narrow width of the final momentum distribution was confirmed experimentally by applying a probe HCP immediately following the last pulse in a train of $N = 25$ chirped pulses. As shown in Fig. 5, the survival probability falls steadily with increasing probe strength displaying a reasonably sharp steplike drop near $\Delta p_0 \simeq 0.7$, which is significantly steeper than that seen for the initial stationary state. The steplike decrease provides clear evidence that

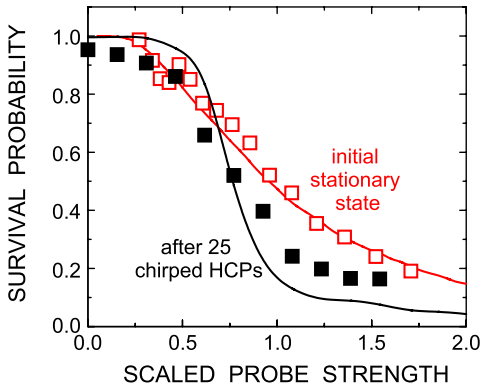

FIG. 5 (color online). Dependence of the survival probability on the scaled strength $\bar{n}\Delta p_{\text{probe}}$ of a probe kick, where $\bar{n}$ is the average principal quantum number of the states being probed, when the probe impulse is both applied directly to the initial $n = 350$ quasi-1D state and following the first 25 HCPs in the train shown in Fig. 1 such that $\bar{n} = 540$. The experimental results (symbols) are compared with CTMC simulations (lines) assuming that stray fields and field inhomogenities in the experiment ionize atoms with $n > 800$.

transport by a chirped HCP sequence preserves the localization of the wave packet in both position and momentum [10]. Again, a reduction in initial alignment can account for the difference between theory and experiment.

Application of the present protocol is not restricted to quasi-1D atoms nor does it necessarily require prelocalization. Transport without prelocalization was explored through both simulation and experiment using K(351p) atoms (rather than quasi-1D atoms) for which there is reasonable initial overlap with the main island. While the efficiency with which parent atoms can be transported to the targeted (higher) n states is reduced, the results showed that some 25% can still be transferred to the desired levels.

This work demonstrates the level of control over atomic spatial and n distributions that can be exercised using a carefully designed sequence of chirped HCPs. The ability to generate strongly polarized quasi-1D atoms in arbitrarily high n states might provide new opportunities to study phenomena such as quantum localization predicted at high scaled frequencies [12]. The ability to transport the electron to states of lower n might be important for schemes to deexcite Rydberg states of antihydrogen and access low-lying states [13]. Chirped HCP trains will also allow wave packets to be diabatically switched between different islands [5] and, possibly, to be better focused by "dithering" the position of the island so as to filter out those portions of the wave packet located near the island edge [6].

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[1] F. B. Dunning *et al.*, Adv. At. Mol. Opt. Phys. **52**, 49 (2005).
[2] R. R. Jones, D. You, and P. H. Bucksbaum, Phys. Rev. Lett. **70**, 1236 (1993).
[3] J. A. Yeazell and C. R. Stroud, Jr., Phys. Rev. Lett. **60**, 1494 (1988).
[4] A. Buchleitner, D. Delande, and J. Zakrzewski, Phys. Rep. **368**, 409 (2002).
[5] C. O. Reinhold *et al.*, J. Phys. B **34**, L551 (2001).
[6] S. Yoshida *et al.*, J. Phys. B **38**, S209 (2005).
[7] H. Maeda, D. V. L. Norum, and T. F. Gallagher, Science **307**, 1757 (2005); M. Kalinski and J. H. Eberly, Opt. Express **1**, 216 (1997).
[8] W. Zhao *et al.*, Phys. Rev. Lett. **97**, 253003 (2006); W. Zhao *et al.*, *ibid.* **95**, 163007 (2005).
[9] A. Messiah, *Quantum Mechanics* (North-Holland, New York, 1970), Vol. 2.
[10] D. G. Arbó *et al.*, Phys. Rev. A **67**, 063401 (2003).
[11] C. L. Stokely *et al.*, Phys. Rev. A **67**, 013403 (2003).
[12] S. Yoshida, C. O. Reinhold, and J. Burgdörfer, Phys. Rev. Lett. **84**, 2602 (2000).
[13] G. Gabrielse *et al.*, Phys. Rev. Lett. **89**, 213401 (2002).