Transfer of population between Rydberg states using a train of half-cycle pulses

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We discuss a protocol for efficient and quick transfer of population between nearly one-dimensional Rydberg states by a chirped train of half-cycle pulses. The chirp refers both to the time interval and relative strength between subsequent pulses. The most spectacular result obtained by the use of this protocol is the transfer of over 10% of the initial population from the high-Rydberg state of $n=50,80$ to much lower ones of $n'\approx 10$ in a short time of the order of 1 ns.

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In recent years there has been increasing interest in the generation and control of the Rydberg wave packets because of their possible use, e.g., for data storage and processing [1]. Since 1996 [2,3], creation, manipulation, and sampling of Rydberg wave packets have been realized with the use of unidirectional half-cycle electric-field pulses (HCP), usually formed in a train with each pulse of a duration much shorter than the Kepler period $T_n=2\pi n^3$ [in atomic units (a.u.)] of the n Rydberg state ($n\gg 1$). A question of particular interest, in this context, is to find an efficient protocol for transporting a Rydberg packet from the initial to the target state by the HCP train. Such a protocol could be used, e.g., for deexcitation of antihydrogen atoms produced in ATRAP and ATHENA experiments [4,5]. The atoms produced in these experiments are known to be left in their high-Rydberg states, $n>30$, while precise spectroscopic studies of fundamental properties of matter, e.g., *CPT* invariance, require antihydrogen atoms in their extremely low-lying states [6]. As antihydrogen quickly annihilates, it imposes the restriction for the protocol of deexcitation to be very quick.

In this Brief Report we present the idea of such an efficient and quick deexcitation protocol exploiting an appropriate HCP train. To simplify the presentation we use the one-dimensional (1D) model of the Rydberg atom. The conditions for such a model are known [7,8] to be well satisfied by selected states only, e.g., the extreme blue and red members of the Stark manifolds. As an approximation, the 1D model is not expected to give quantitatively exact results for fully three-dimensional (3D) atoms. According to [9–11], this model is, however, able to predict some qualitative tendencies in real 3D Rydberg atoms exposed to HCPs. The other approximation we apply is the widely used impulse approximation for the atom-pulse interaction [12,13]. With these two approximations, the transition amplitude from the initial state $|n\rangle$ to a given state $|n'\rangle$ under the action of a HCP is provided by the form factor (in a.u.) $T_{n'n}=\langle n'|\exp(iqx)|n\rangle$ in the following explicit form [14]:

$$T_{n'n}=-\frac{z\left(\lambda-\frac{2}{n}\right)^{n}\left(\lambda-\frac{2}{n'}\right)^{n'}}{\sqrt{nn'}\lambda^{n+n'}}\times\left\{\left[\frac{n-1}{\lambda-\frac{2}{n}}+\frac{n'-1}{\lambda-\frac{2}{n'}}-\frac{n+n'}{\lambda}\right]F(-n+1,-n'+1,2,z)-\frac{1}{2}(n-1)(n'-1)\left[\frac{1}{\lambda-\frac{2}{n}}+\frac{1}{\lambda-\frac{2}{n'}}\right]\times zF(-n+2,-n'+2,3,z)\right\},\quad(1)$$

where q is the momentum transferred to the Rydberg electron by HCP, $F(a,b,c,y)$ is the hypergeometric function, $\lambda=n^{-1}+n'^{-1}-iq$, and $z=-4n'n[(n-n')^2+(qn'n)^2]^{-1}$. The Rydberg wave packet created by a single HCP from the initial state $|n\rangle$ is written as $|\psi\rangle=\Sigma_{n'}T_{n'n}|n'\rangle$, where the summation runs over all states left populated after HCP. If, instead of a single HCP, one uses a train of HCPs with different q, in general, and subsequent pulse delays $\tau_1,\tau_2,\ldots,\tau_j$, where τ_j stands for the delay after the jth HCP, the Rydberg wave packet at the end of the train of K HCPs reads

$$|\psi_K\rangle=\sum_{n_K}\cdots\sum_{n_2}\sum_{n_1}T_{n_Kn_{K-1}}e^{-i\omega_{n_{K-1}}\tau_{K-1}}\cdots\times e^{-i\omega_{n_2}\tau_2}T_{n_2n_1}e^{-i\omega_{n_1}\tau_1}T_{n_1n}|n\rangle=\sum_{n_K}b_{n_K}|n_K\rangle.\quad(2)$$

Here, ω_j are the state eigenfrequencies ($-1/2n_j^2$ in a.u.), b_{n_K}—the amplitudes of the states that remained populated after the Kth pulse, and the exponential functions describe free evolution of the states in between two subsequent HCPs. In the following, we assume pulses that transfer the negative momentum to the electron in the 1D atom, thus pushing the electron toward the nucleus. To take into account the possibility of ionization, Eq. (1) needs to be adapted in the following way. First, we replace $1/\sqrt{n'}$ before the brace by $\sqrt{2\pi k/[1-\exp(-2\pi/k)]}/(ik)$, and then, replace all other n' by $1/(ik)$, where $k\geq 0$ stands for the momentum (in a.u.) of the continuum-state electron. The use of Eq. (1) means that

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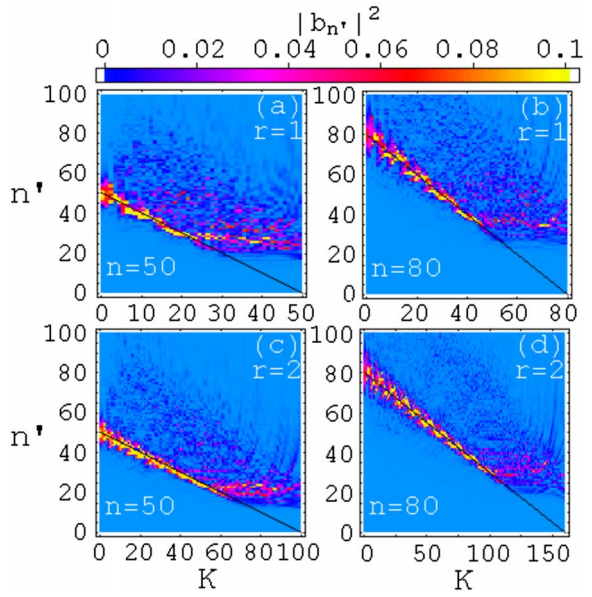

FIG. 1. (Color online) Rydberg-state distribution, $|b_{n'}|^2$, versus the number K of identical HCPs (each of $q=-0.1/n$) in the train of spacing $\tau_K=[1-K/(nr)]^3T_n$, with $r=1$ [parts (a) and (b)] and $r=2$ [parts (c) and (d)], for the initial states $n=50$ [parts (a) and (c)] and $n=80$ [parts (b) and (d)]. The black lines indicate the comparative states $n_C=|n-K/r\rangle$.

we treat the problem in a way different than by other authors [15,16].

A series of detailed calculations, exploiting Eqs. (1) and (2), has led us to the conclusion that Rydberg wave packet exposed to an appropriately chirped train of equal-momentum pulses tends to be localized around the Rydberg state whose Kepler period is equal to the delay between subsequent pulses. We shall show this by assuming the momentum transfer by a single HCP in the amount $q=-0.1/n$ and the chirping of delay between pulses in the form

$$\tau_K=[1-K/(nr)]^3T_n, \tag{3}$$

where T_n stands for the Kepler period of the initial state n. Under the action of such a train, the Rydberg wave packet is shown in Fig. 1 to be concentrated around the most occupied state $n'=n-K/r$ of the Kepler period close to $T_{n'}=T_{n-K/r}=\tau_K$. By different colors we present in Fig. 1 the calculated Rydberg-state distribution, $|b_{n'}|^2$, after K pulses of the train, for two chosen values of parameter r [$r=1$ in parts (a) and (b), and $r=2$ in parts (c) and (d)]. Parts (a) and (c) are the distributions from the initial state $n=50$ while parts (b) and (d) are those from $n=80$. The chosen principal quantum numbers correspond to those observed in the production of antihydrogen atoms [4]. Each part includes the black line corresponding to the target Rydberg states $n_C=n'=n-K/r$ with their Kepler periods $T_{n'}=\tau_K$. In all cases shown in Fig. 1, the packet, until some number of HCPs, is moved almost monotonously toward low-lying states, and the compatibility between the most populated state (the brightest points) and n_C is very good—the largest-population state is really that close to n_C. As seen, the main population moves down from the initial state $n=50$ to $n'=28$ after 24 pulses for $r=1$, and

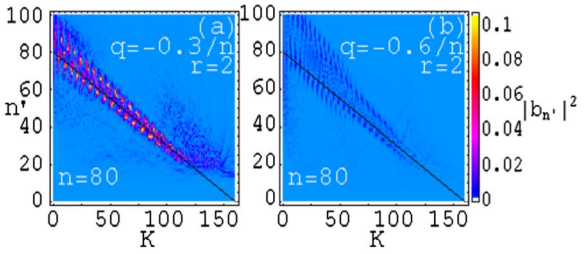

FIG. 2. (Color online) Rydberg-state distribution, $|b_{n'}|^2$, versus the number K of identical HCPs in the train of spacing $\tau_K=[1-K/(nr)]^3T_n$, with $r=2$, for the initial states $n=80$. Momentum of each pulse is $q=-0.3/n$ [part (a)] and $q=-0.6/n$ [part (b)]. The black lines indicate the comparative states $n_C=|n-K/r\rangle$.

to $n'=20$ after 60 pulses for $r=2$. For the initial state $n=80$, the main population is deexcited to $n'=35$ after 47 pulses for $r=1$ and to 26 after 108 pulses for $r=2$, respectively. For a given n, the parameter r is, thus, responsible for the speed of deexcitation and, by Eq. (3), it can be understood as the number of pulses the electron needs to be moved one state down. Figure 1 shows that deexcitation stops for a train long enough. The packet submits then delocalization in the sense that it includes many states of comparable populations. These states have diverse Kepler periods, different from the delay between the pulses and, therefore, the packet can no longer be efficiently deexcited. In Fig. 1, we straightforwardly see the difference between state distributions for K located in the area of efficient deexcitation and K outside this range. For K outside the efficient area, the populations of the preferred states, n_C, are seen to be clearly lower. As seen, the higher the r the lower the n' at which the breakdown of deexcitation occurs, which translates into more efficient process of deexcitation for higher r. The disadvantage of higher r is, however, an increase in the length of the train. In the moment of the breakdown of deexcitation, the length amounts to 8.8×10^6 a.u.$=0.21$ ns for $r=1$ and 1.9×10^7 a.u.$=0.45$ ns for $r=2$ if the initial state is $n=50$, and 5.7×10^7 a.u.$=1.4$ ns for $r=1$ and 1.3×10^8 a.u.$=3.0$ ns for $r=2$ if the initial state is $n=80$. A possible source of cessation of deexcitation for large enough K is increasing anharmonicity of level separation when approaching the nucleus. In Fig. 2, we show Rydberg-state distribution, $|b_{n'}|^2$, after K pulses in the train of Eq. (3), for the initial state $n=80$ and $r=2$, but with momentum q transferred by each pulse different than that in Fig. 1(d). By comparison with Fig. 1, we see that the increase in q, when all pulses in the train are identical, results in diminishing the amount of deexcited population. Moreover, the lack of yellow points in Fig. 2(b) can be attributed to an increase in the ionization.

The way to overcome the disadvantageous effect of increasing anharmonicity in energy separation of low states is suggested, to some extent, by the behaviors of the amplitudes $T_{n'n''}$ versus decreasing n' and n'' for the fixed pulse strength $q=-0.1/n$. The surviving amplitude $T_{n'n'}$ of the state n' is found to become more and more dominant one when approaching the lower states n'. The two other amplitudes of jumping up ($n''=n'+1$) and down ($n''=n'-1$), respectively, remain nearly the same when $n'\gg1$ but for diminishing n' the former amplitude gets higher with respect to the latter

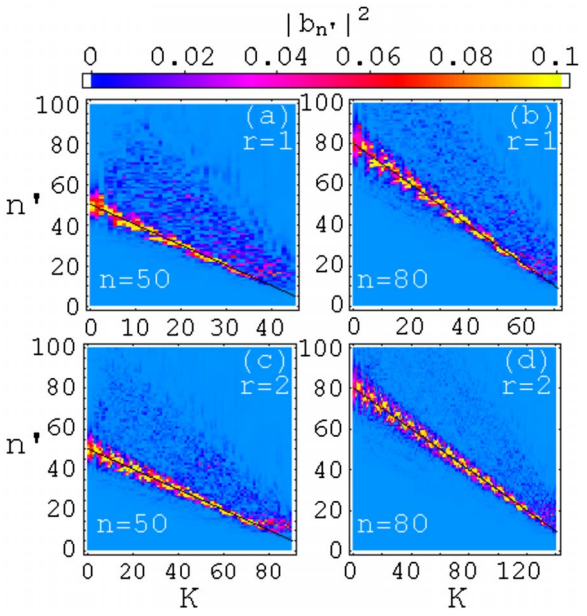

FIG. 3. (Color online) Rydberg-state distribution, $|b_{n'}|^2$, versus the number K of HCPs in the train of spacing $\tau_K=[1-K/(nr)]^3T_n$ and the momentum transferred by the Kth pulse $q_K=-0.1/(n-K/r)$. The initial states and parameters r as in Fig. 1. The black lines indicate the comparative states $n_C=|n-K/r\rangle$.

one. These behaviors disturb the process of deexcitation and when the deexcitation stops the Rydberg packet stays in nearly the same level, as seen in Fig. 1. However, one can enhance the deexcitation process by increasing slightly the strength of subsequent pulses, still chirping the delay between them in the way given by Eq. (3), because the increasing $|q|$ makes the down transitions more effective than the up transitions. Thus, the improved protocol of deexcitation reads

$$\tau_K=[1-K/(nr)]^3T_n,$$

$$q_K=-\frac{0.1}{n[1-K/(nr)]}. \tag{4}$$

Figure 3 shows the Rydberg-state distribution as a function of K, for the train with τ and q chirped as in Eq. (4). We have retained the same values for r and n as in Fig. 1. As compared with Fig. 1, we see an improvement in the deexcitation process—now efficient deexcitation maintains for higher K, and the state with the largest population at the moment of the breakdown of deexcitation is lower than before. As follows from Fig. 3, for the initial state $n=50$, the packet stops its deexcitation after $K=37$ for $r=1$ and after $K=80$ for $r=2$. In these cases the states with the highest population are $n'=14$ and $n'=11$, respectively. For the initial state $n=80$, the state with the highest population is $n'=17$ after 64 pulses for $r=1$ and $n'=13$ after 136 pulses for $r=2$. The deexcitation from the state $n=80$ to the state $n'=13$ means that the mean lifetime of the Rydberg atom drops down from 30 ms to 5.4 μs [17]. In this case, the deexciting train lasts 1.3×10^8 a.u.$=3.2$ ns only. These deexcitation results are better than our previous ones [18] obtained with chirping τ only.

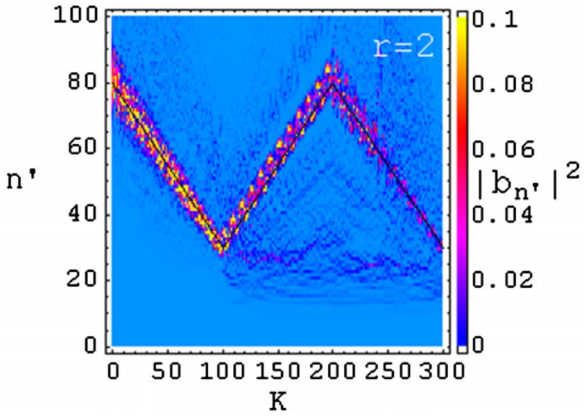

FIG. 4. (Color online) Rydberg-state distribution, $|b_{n'}|^2$, versus the number K of pulses in the chirped train given by Eq. (5), for the initial state $n=80$. The black line indicates the comparative state.

An appropriately chirped train can be used not only as a tool for the deexcitation but also for the controlled excitation of Rydberg states. Figure 4 shows the Rydberg-state distribution as a function of the number K of pulses in the train acting on the initial state $n=80$, with τ and q being chirped as

$$\tau_K=[1-K/(2\times80)]^3T_{80},$$

$$q_K=-\frac{0.1}{n[1-K/(2\times80)]}\quad \text{for } K\le100,$$

$$\tau_K=[1+(K-100)/(2\times30)]^3T_{30},$$

$$q_K=-\frac{0.1}{n[1+(K-100)/(2\times30)]}\quad \text{for } 100<K\le200,$$

$$\tau_K=[1-(K-200)/(2\times80)]^3T_{80}, \tag{5}$$

$$q_K=-\frac{0.1}{n[1-(K-200)/(2\times80)]}\quad \text{for } K>200.$$

As compared with Eq. (4), $r=2$ now. The above train shifts the population from the initial state $n=80$ to the surroundings of $n'=30$ for the first 100 pulses ($n_C=80-K/r$); for the next 100 HCPs it excites the population to the surroundings of $n=80$ [$n_C=30+(K-100)/r$], and for the last 100 pulses it deexcites the population to $n'=30$ again [$n_C=80-(K-200)/r$]. However, during the excitation step, a part of the population is seen separating from the rest and becomes trapped in low-lying states. We have compared the results of Fig. 4 with those for the train with q kept constant ($q=-0.1/n$) and only τ being chirped as in Eq. (5). In the latter case, we have observed narrower distribution for a given K and weaker excitations in the middle step. The reversibility of the population-transfer process, shown in Fig. 4, agrees with the recent result of Mestayer *et al.* [16] found by a different method for the much higher quasi-1D initial state exposed to a different HCP train with only τ being chirped.

In summary, we have presented a protocol enabling efficient and quick transfer of population between Rydberg states in the 1D model atom exposed to a HCP train. Crucial for the protocol was the appropriate chirping of the train. We have shown that, by changing the interval between subsequent identical pulses, the Rydberg packet evolves in such a way that the most populated state in it is that whose Kepler period matches the interval between pulses. Therefore a shortening of this intervals leads to deexcitation. The initial states $n=50$ and $n=80$ were transferred to states $n'=20-30$ with the efficiency higher than 10% in a short time of the order of 1 ns. Even better results were obtained when chirping referred to both intervals between the pulses and the momentum transferred by subsequent pulses to the Rydberg +electron. By this way, the electron initially in the state $n=50$ was deexcited to the state $n'=11$, and the initial state $n=80$ was deexcited to $n'=13$, with efficiency higher than in the case when the chirping concerned the delay between the pulses only. Because of the employed 1D approximation, the above results cannot be directly compared with those of other authors [19–21] considering different methods of deexcitation of 3D atoms. However, our results are expected to indicate how to improve the deexcitation process in real atoms.

[1] J. Ahn, T. C. Weinacht, and P. H. Bucksbaum, Science **287**, 463 (2000).
[2] R. R. Jones, Phys. Rev. Lett. **76**, 3927 (1996).
[3] C. O. Reinhold, J. Burgdörfer, M. T. Frey, and F. B. Dunning, Phys. Rev. A **54**, R33 (1996).
[4] G. Gabrielse *et al.*, Phys. Rev. Lett. **89**, 213401 (2002).
[5] M. Amoretti *et al.*, Nature (London) **419**, 456 (2002).
[6] M. H. Holzscheiter and M. Charlton, Rep. Prog. Phys. **62**, 1 (1999).
[7] T. F. Gallagher, *Rydberg Atoms* (Cambridge University Press, New York, 1994).
[8] C. L. Stokely, J. C. Lancaster, F. B. Dunning, D. G. Arbó, C. O. Reinhold, and J. Burgdörfer, Phys. Rev. A **67**, 013403 (2003).
[9] M. T. Frey, F. B. Dunning, C. O. Reinhold, S. Yoshida, and J. Burgdörfer, Phys. Rev. A **59**, 1434 (1999).
[10] E. Persson, S. Yoshida, X. M. Tong, C. O. Reinhold, and J. Burgdörfer, Phys. Rev. A **68**, 063406 (2003).
[11] C. O. Reinhold, W. Zhao, J. C. Lancaster, F. B. Dunning, E. Persson, D. G. Arbó, S. Yoshida, and J. Burgdörfer, Phys. Rev. A **70**, 033402 (2004).
[12] P. Krstic and Y. Hahn, Phys. Rev. A **48**, 4515 (1993); **50**, 4629 (1994); Phys. Lett. A **192**, 47 (1994).
[13] R. Parzyński and M. Sobczak, Phys. Rev. A **65**, 045401 (2002).
[14] I. Bersons and R. Veilande, Phys. Rev. A **69**, 043408 (2004).
[15] S. Yoshida, C. O. Reinhold, E. Persson, J. Burgdörfer, and F. B. Dunning, J. Phys. B **38**, S209 (2005).
[16] J. J. Mestayer, W. Zhao, J. C. Lancaster, F. B. Dunning, C. O. Reinhold, S. Yoshida, and J. Burgdörfer, Phys. Rev. Lett. **99**, 183003 (2007).
[17] E. S. Chang, Phys. Rev. A **31**, 495 (1985).
[18] T. Kopyciuk and R. Parzyński, Phys. Rev. A **75**, 055402 (2007).
[19] S. X. Hu and L. A. Collins, Phys. Rev. A **69**, 041402(R) (2004).
[20] T. Topçu and F. Robicheaux, Phys. Rev. A **73**, 043405 (2006).
[21] A. Wetzels, A. Gürtler, L. D. Noordam, and F. Robicheaux, Phys. Rev. A **73**, 062507 (2006).