Coherent Population Transfer in an Atom by Multiphoton Adiabatic Rapid Passage

H. Maeda, J. H. Gurian, D. V. L. Norum, and T. F. Gallagher
Department of Physics, University of Virginia, Charlottesville, Virginia 22904-0714, USA
(Received 2 August 2005; published 23 February 2006)

Coherent population transfer in an atom using a sequence of adiabatic rapid passages through single-photon resonances is well-known, but it requires that the frequency sweep match the changing frequencies of the atomic transitions. The same population transfer can be effected via a single multiphoton adiabatic rapid passage, which requires only a small frequency sweep, if it is possible to select the desired multiphoton transition from the many possible transitions. Here we report the observation of population transfer between Rydberg states by high order multiphoton adiabatic rapid passage.

DOI: 10.1103/PhysRevLett.96.073002 PACS numbers: 32.80.Rm, 32.80.Bx, 32.80.Qk

Adiabatic rapid passage (ARP) is an approximately 100% efficient way to transfer population from one state to another, which makes coherent population transfer using a sequence of ARP's practical. Examples are using a chirped infrared-laser pulse to make a sequence of vibrational transitions in a slightly anharmonic diatomic molecule [1–3], the production of circular states by a sequence of $\Delta m = +1$ or -1 microwave (MW) transitions [4], and using a chirped MW pulse to change the principal quantum number n of atomic Rydberg states [5–7]. In the above examples the frequency of the atomic or molecular motion, i.e., the single-photon transition frequency, follows the changing frequency of the radiation. Consider the example of Fig. 1(a), a Rydberg atom initially in the state of $n = 72$, which has a Kepler or $\Delta n = 1$ transition frequency of 17.3 GHz. If this atom is exposed to a MW pulse chirped from 17.5 to 12 GHz it undergoes a sequence of ARP's up in n to the $n = 82$ state, which has a Kepler frequency of 12.2 GHz.

Here we report an alternative method of coherent population transfer, in which we replace the sequence of ARP's of single-photon transitions with ARP of a single multiphoton transition. For example, replacing the sequence of one-photon ARP's of Fig. 1(a) by ARP of the ten-photon $n = 72$ to $n = 82$ transition at 15.2 GHz [see Fig. 1(b)]. Using a multiphoton transition necessitates higher power, but, since there is only one transition, the range of the frequency sweep can be dramatically reduced. The advantages of using ARP's of multiphoton transitions for coherent population transfer were first suggested by Oreg *et al.* [8], and more recently by Gibson [9]. A well-known example is the "counterintuitive" pulse sequence [10], which leads to coherent population transfer in three-level systems by ARP of a two-photon transition, as demonstrated by Broers *et al.* [11]. The measurements reported here can be thought of as a multiphoton generalization of the counterintuitive pulse sequence, and they demonstrate that ARP using multiphoton transitions is, in fact, quite robust. In the sections which follow we outline the essential idea, describe our experiments, and discuss the implications.

A useful way of describing ARP is as an adiabatic traversal of an avoided crossing of Floquet levels [12]. We calculate the Floquet energy levels using a one-dimensional model for the atom in which the energy W is given by $W = -1/2n^2$ and the matrix element coupling adjacent n states by $\langle n|x|n+1\rangle = 0.3n^2$. We use atomic units, unless specified otherwise. A one-dimensional model provides a good description of Rydberg atoms in strong, linearly polarized MW fields [13,14].

In zero MW field the Floquet (or dressed-state) energy of each n state is given by

$$W = -1/2n^2 - (n - 75)\omega, \tag{1}$$

where ω is the MW angular frequency. The $n = 75$ energy is frequency independent, and the $n = 73$ energy, for example, increases twice as rapidly as the MW frequency [see Fig. 2(a)]. In Fig. 1(b) we show the $n = 72$ and $n = 82$ Floquet levels as a function of MW frequency near the ten-photon $n = 72$–82 resonance at 15.2 GHz. In zero MW field the two levels cross, as shown by the broken lines, and as shown by the solid lines, in a MW field of 3 V/cm there is an avoided crossing of magnitude $\Omega_{10} \approx 0.5$ GHz, which is the ten-photon Rabi frequency. ARP from $n = 72$ to 82 can be effected by sweeping the frequency through the ten-photon resonance in either direction as shown by the two arrows in Fig. 1(b).

The probability of ARP through an isolated k-photon resonance with a linear frequency sweep S is given by $P_k = \exp(-\pi^2\Omega_k^2/kS)$, where Ω_k is the magnitude of the avoided crossing in GHz and S is given in GHz/ns. The requirement for ARP is

$$\Omega_k > \frac{\sqrt{kS}}{\pi}. \tag{2}$$

In our experiments $S = 0.012$ GHz/ns, so Ω_k for $k = 1$ and 10 Eq. (2) requires $\Omega_1 > 35$ MHz and $\Omega_{10} > 110$ MHz, respectively. For $k = 1$ $\Omega_k = 0.3n^2E$, in which E is the MW-field amplitude, and for $k > 1$ an approximate requirement is that $0.3n^2E/\Delta_d \approx 1$ where Δ_d is the largest detuning from an intermediate-state resonance. For a

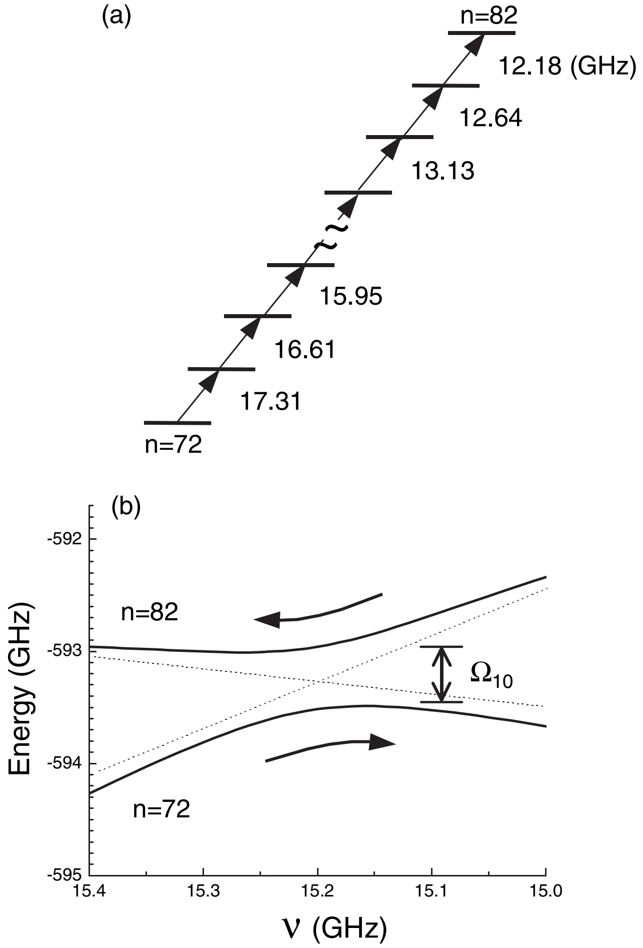

FIG. 1. (a) $n = 72 \rightarrow 82$ transition by a sequence of single-photon ARPs. (b) $n = 72$ and $n = 82$ Floquet states near the ten-photon resonance plotted as a function of MW frequency ν (solid curves). In zero MW field the two levels cross, as shown by the broken lines.

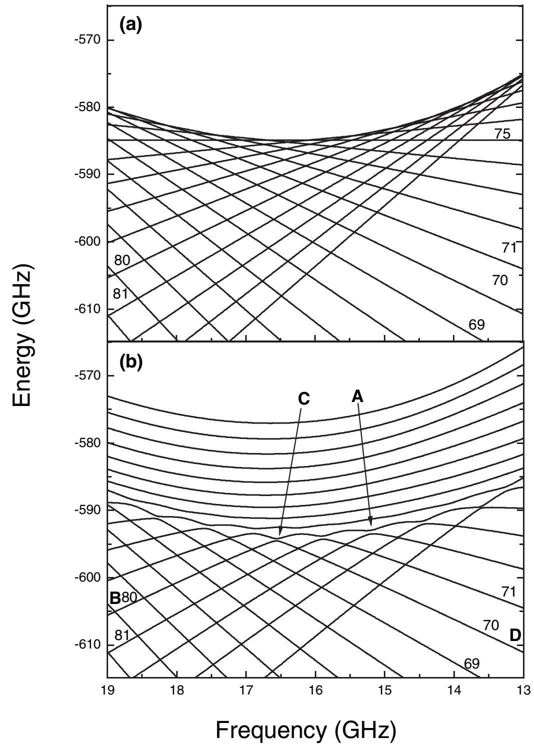

FIG. 2. (a) Floquet energy levels as defined in Eq. (1) for $60 \leq n \leq 84$ vs MW frequency in zero MW field. The $n = 69$, 70, 71, 75, 80, and 81 levels are labeled. The $\Delta n = 1$ resonances are the highest lying level crossings, and $\Delta n > 1$ crossings are lower in energy. (b) With a MW amplitude of 3 V/cm, the $\Delta n = k$, $k \leq 8$ avoided crossings become smooth curves, the $\Delta n = 10$ avoided crossings are recognizable as isolated avoided-level crossings, and the $\Delta n \geq 11$ avoided crossings are invisible on this scale. $n = 72$–82 ten-photon avoided crossing is denoted as A at 15.2 GHz. With a $19 \rightarrow 13$ GHz chirped pulse the atoms in level B pass to D through C.

k-photon transition the variation in the $\Delta n = 1$ frequency, $1/n^3$, leads to $\Delta_d = 3k/2n^4$ and $E = 5k/n^6$. Thus the requisite MW field E increases linearly with k, for $k > 1$. This simple model suggests that for $n = 75$ the required fields are 18 mV/cm and 1.4 V/cm for $k = 1$ and 10. From Eq. (1) it is apparent that the minimum required frequency sweep is

$$\Delta \nu \approx \frac{2\Omega_k}{k}, \tag{3}$$

which decreases as $1/\sqrt{k}$ and is $\sim$22 MHz for our slew rate and $k = 10$. For the avoided crossing shown in Fig. 1(b), with $E = 3$ V/cm, the minimum sweep is ≈ 100 MHz.

The ten-photon avoided crossing shown in Fig. 1(b) does not exist in isolation but is surrounded by other level crossings of both higher and lower order. Whether or not this avoided crossing is accessible is a crucial question. To address it we show in Fig. 2 the calculated Floquet energy levels for $n \sim 75$ vs the frequency of the MW field. As shown in Fig. 2(a) in zero MW field the k-photon, $\Delta n = k$ resonances appear as level crossings. The one-photon $\Delta n = 1$ resonances lie along the top of the energy levels shown in Fig. 2(a). The $k > 1$ resonances lie below them. The $\Delta n = 10$ resonance between the $n = 72$ and $n = 82$ levels of Fig. 1(b) occurs where the levels cross at 15.2 GHz. In Fig. 2(b) we show the same levels with a MW field of amplitude 3 V/cm. All level crossings become avoided crossings, and at this field the sequences of $\Delta n = k$ avoided crossings for $k \leq 8$ become smooth curves. For $\Delta n = k = 10$ there are recognizable avoided crossings, and for $\Delta n \geq 11$ the size of the avoided crossings decreases by an order of magnitude for an increase in Δn of one, producing avoided crossings invisible on the scale of Fig. 2. As shown by point A in Fig. 2(b) the ten-photon avoided crossing of Fig. 1(b) is by no means isolated, but we can use it to effect population transfer. The requirement is that it be traversed adiabatically and all other avoided crossings diabatically. An obvious approach is a Gaussian pulse swept from 15.1 to 15.3 GHz with a peak amplitude of 3 V/cm.

In the experiment a beam of ground state Li atoms passes through a WR62 waveguide where the atoms are excited to np states by three 5 ns laser pulses using the sequence $2s \rightarrow 2p \rightarrow 3s \rightarrow np$. They are then exposed to a frequency swept MW pulse. Finally, a voltage ramp rising in 1100 ns is applied to a septum in the waveguide for selective field ionization. The electrons resulting from field ionization are ejected through a hole in the top of the waveguide and are detected with a dual microchannel-plate detector. Since the electrons have negligible flight time and atoms ionize at $F = 1/9n^4$ [15], the time-resolved electron signal allows us to determine the final n-state distribution.

To produce the chirped MW pulse we use a voltage-controlled oscillator, whose frequency varies from 13 to 19 GHz as the control voltage is changed from 2 to 15.5 V. We use its maximum sweep rate of 0.012 GHz. The typical output power is 10 mW. Using a control pulse from an arbitrary waveform generator and a pair of mixers in series we form the output into a swept pulse from 50 to 500 ns long. The pulse is amplified to powers as high as 300 mW with a solid-state amplifier and transported to the waveguide in the vacuum system.

As noted earlier, a Gaussian pulse should be nearly ideal, and using a 1 V, 50-ns-long Gaussian control pulse from the arbitrary waveform generator we have generated a swept pulse centered at 15.2 GHz. In Fig. 3 we show the population transfer observed when starting with $n = 72$ atoms and exposing them to this pulse. Figure 3 is composed of oscilloscope traces of the time-resolved field-ionization signals observed with no pulse (dashed line), and pulses with a peak amplitudes of 2 V/cm (dotted line) and 3 V/cm (solid line). For no pulse, and pulses of peak amplitude <1 V/cm no population transfer is observed and the signal is observed at $t = 800$ ns. For a peak amplitude of 2 V/cm, almost half the signal is observed at $t = 640$ ns, corresponding to population transfer to $n \approx 83$. For a peak amplitude of 3 V/cm > 80% of the population is transferred to $n \approx 83$. At higher fields the population transfer decreases, as expected. We have changed n by ~11 using a MW pulse which is only chirped by 600 MHz in 50 ns. Using the Gaussian pulse used in the population transfer of Fig. 3 leads to about half as much transfer for initial $n = 71$ and 73 states and no transfer for initial $n = 70$ and 74 states.

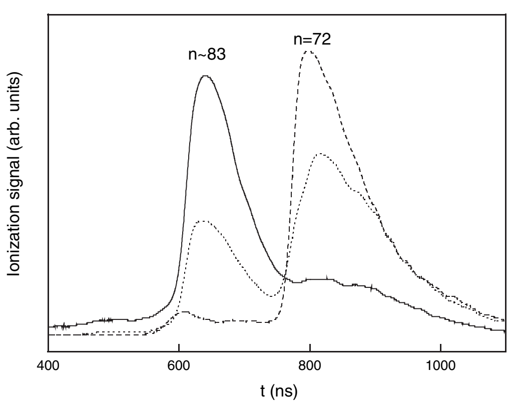

FIG. 3. Time-resolved field-ionization signals obtained subsequent to exposing $n = 72$ atoms to a 50 ns, +0.012 GHz/ns chirped pulse centered at 15.2 GHz, so that only ~600 MHz of chirp is required. For pulse amplitude zero (dashed line), the atoms stay in the $n = 72$ state. For pulse amplitude 2 V/cm (dotted line), roughly 40% of the atoms are transferred to $n \approx 83$. With pulse amplitude 3 V/cm (solid line), more than 80% of the atoms are transferred to $n \approx 83$.

One of the reasons for using a short frequency sweep is to minimize the number of avoided crossings encountered to ensure that only the desired avoided crossing is traversed adiabatically, but the sweep need not be short. With properly chosen pulses swept over 6 GHz the atom will find the desired avoided crossing itself. In Fig. 4 we show the population transfers observed with 500-ns-long constant amplitude pulses swept in both directions between 13 and 19 GHz. The data shown in each panel are gray-scale representations of time-resolved field-ionization signals for amplitudes of MW field E from 0.015 to 15 V/cm. In Fig. 4(a) we show the result of exposing $n = 80$ atoms to a $19 \rightarrow 13$ GHz chirp. As E increases from 0.1 to 3 V/cm the change in n increases from 0 to 10. How this population transfer occurs when $E \approx 3$ V/cm may be understood with the aid of Fig. 2(b). The atoms pass diabatically from point B to C, where ARP occurs, followed by a diabatic passage to point D. Figure 4(b) shows the analogous result for $n = 73$ atoms exposed to a pulse chirped from 13 to 19 GHz. In Figs. 4(a) and 4(b) the change in n increases with the MW-field amplitude and is approximately equal to the number of levels coupled together by the MW field, i.e., the number of smooth energy-level curves at the top of Fig. 2(b). It is as if the atoms follow diabatic trajectories which are reflected from the smooth curves of Fig. 2(b). This observation can be understood by considering the requirement for an adiabatic passage given by Eq. (2). In the chirped pulses used in obtaining the data of Figs. 4(a) and 4(b) the first and only avoided crossing to be traversed adiabatically is the one just below the smooth curves. By calculating the Floquet level structure, and thus Ω_k, for different microwave fields we can predict the Δn of the population transfer for a given field amplitude. The results of these calculations for the conditions of, for example, Fig. 4(a) are in good agreement with our observations, as shown. With a 6 GHz sweep we can select Δn, independent of n over a range of n, by the MW-field amplitude.

The multiphoton ARP approach described here allows rapid, efficient population transfer over many n states with easily generated pulses. One can envision using several such pulses, centered at different frequencies, to effect still larger changes in n on a 1 μs time scale, which could be quite useful for transporting recombined antihydrogen to

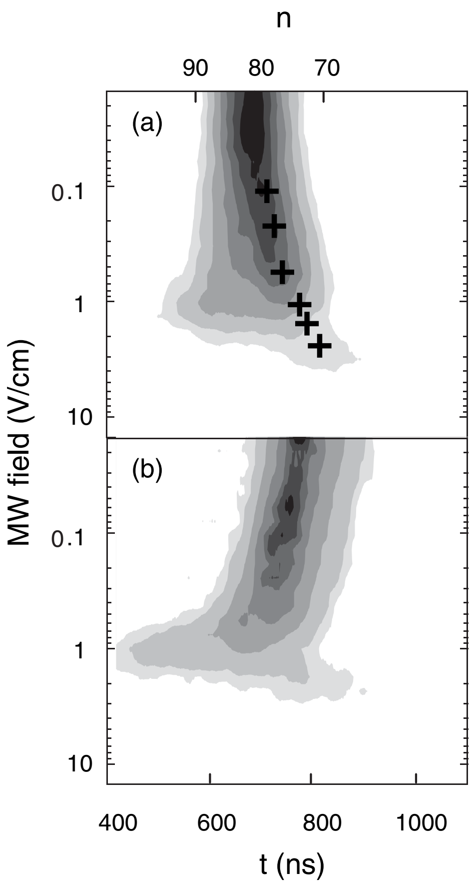

FIG. 4. Gray-scale renderings of the final-state distributions subsequent to exposure to constant amplitude MW pulses. Each panel is built up from 26 oscilloscope traces of the field-ionization signals; $t = 0$ corresponds to the beginning of the field ramp. (a) Atoms initially in $n = 80$ exposed to 19 to 13 GHz chirped pulses. The calculated Δn transfers are shown by crosses (+). Population transfer to n as low as ~ 72 is observed. (b) Atoms initially in $n = 73$ exposed to 13 to 19 GHz chirped pulses. Population transfer to $n \approx 77$ is efficient, and transfer to very high n states is observed.

lower lying states [16]. More generally, this work suggests that it may actually be simpler to use a single multiphoton resonance than a sequence of single-photon resonances since only a small chirp is required. Furthermore, this process is robust; it works in spite of the presence of many levels which we have ignored. Since it is straightforward to generate tailored laser pulses [17–19], especially ones with Gaussian intensity profiles and prescribed chirps, this approach should be applicable to other physical systems [2,9]. For example, laser excitation of a high vibrational state of a diatomic molecule using one multiphoton transition rather than a sequence of single-photon transitions is a case almost identical to this one.

It is a pleasure to acknowledge stimulating discussions with L. A. Bloomfield and R. R. Jones. This work has been supported by the National Science Foundation under Grants No. PHY-0244320 and No. CHE-0215957.

[1] S. Chelkowski, A. D. Bandrauk, and P. B. Corkum, Phys. Rev. Lett. **65**, 2355 (1990).
[2] D. J. Maas *et al.*, Chem. Phys. Lett. **290**, 75 (1998).
[3] D. M. Villeneuve *et al.*, Phys. Rev. Lett. **85**, 542 (2000).
[4] R. G. Hulet and D. Kleppner, Phys. Rev. Lett. **51**, 1430 (1983).
[5] B. Meerson and L. Friedland, Phys. Rev. A **41**, 5233 (1990).
[6] J. Lambert, M. W. Noel, and T. F. Gallagher, Phys. Rev. A **66**, 053413 (2002).
[7] H. Maeda, D. V. L. Norum, and T. F. Gallagher, Science **307**, 1757 (2005).
[8] J. Oreg, F. T. Hioe, and J. H. Eberly, Phys. Rev. A **29**, 690 (1984).
[9] G. N. Gibson, Phys. Rev. A **72**, 041404 (2005).
[10] N. V. Vitanov *et al.*, Annu. Rev. Phys. Chem. **52**, 763 (2001); N. V. Vitanov *et al.*, Adv. At. Mol. Opt. Phys. **46**, 55 (2001).
[11] B. Broers, H. B. van Linden van den Heuvell, and L. D. Noordam, Phys. Rev. Lett. **69**, 2062 (1992).
[12] J. H. Shirley, Phys. Rev. **138**, B979 (1965).
[13] R. V. Jensen, S. M. Susskind, and M. M. Sanders, Phys. Rep. **201**, 1 (1991).
[14] A. Buchleitner, D. Delande, and J. Zakrewski, Phys. Rep. **368**, 409 (2002).
[15] T. F. Gallagher, *Rydberg Atoms* (Cambridge University Press, Cambridge, 1994), p. 103.
[16] G. Gabrielse *et al.*, Phys. Rev. Lett. **89**, 213401 (2002).
[17] T. C. Weinacht, J. Ahn, and P. H. Bucksbaum, Phys. Rev. Lett. **80**, 5508 (1998).
[18] G. Vogt *et al.*, Phys. Rev. Lett. **94**, 068305 (2005).
[19] E. Wells *et al.*, Phys. Rev. A **72**, 063406 (2005).