2002

Rydberg-Atom Population Transfer By Population Trapping in a Chirped Microwave Pulse

J. Lambert

Michael Noel
Bryn Mawr College, mnoel@brynmawr.edu

T. F. Gallagher

Custom Citation

J. Lambert, M.W. Noel and T.F. Gallagher, *Phys. Rev. A* **66**, 53413 (2002).

Rydberg-atom population transfer by population trapping in a chirped microwave pulse

J. Lambert,[1] Michael W. Noel,[1,2] and T. F. Gallagher[1]
[1]*Department of Physics, University of Virginia, Charlottesville, Virginia 22904*
[2]*Department of Physics, Bryn Mawr College, Bryn Mawr, Pennsylvania 19010*
(Received 8 August 2001; revised manuscript received 16 August 2002; published 22 November 2002)

We demonstrate that Rydberg atoms can be transferred to states of lower principal quantum number by exposing them to a frequency chirped microwave pulse. Specifically, we have transferred $n=75$ atoms to $n=66$ with a 400-ns pulse chirped from 7.8 to 11.8 GHz. In spite of the large number of coupled levels, using a simplified model we can describe the process reasonably well as a sequence of adiabatic rapid passages.

DOI: 10.1103/PhysRevA.66.053413 PACS number(s): 32.80.Rm

Adiabatic rapid passage is a robust method for transferring population from one energy level to another [1,2]. If the frequency of radiation coupling the two levels is swept through the resonance at a rate small compared to the square of the Rabi frequency, the efficiency of population transfer is very nearly 100%. The high efficiency of each step allows population to be transferred through many levels by sequential adiabatic rapid passages. Two examples are the production of circular Rydberg states and vibrational ladder climbing using chirped laser pulses [3–5]. At first glance, changing the energy, or the principal quantum number *n*, of a Rydberg atom by a chirped microwave pulse approximately resonant with the changing Kepler frequency appears to be similar. Upon closer examination, however, it is apparent that passing through a sequence of *n* states by a series of adiabatic rapid passages differs from the previous two problems in that there is not a single sequence of levels, but a sequence of groups of levels, i.e., all the ℓ states of the same *m* which are coupled together. Here ℓ and *m* are the orbital and azimuthal angular-momentum quantum numbers; we assume the microwave field to be linearly polarized.

In addition to the intrinsic interest of this problem it may have practical importance as well. Meerson and Friedland [6] suggested that using a microwave pulse, initially at the Kepler frequency and chirped to lower frequency, would transfer atoms to a higher *n* state, leading to ionization at a lower microwave field. Recent work of Bensky *et al.* [7] and Wesdorp *et al.* [8] suggests that it might be more interesting to chirp the frequency in the other direction. They have demonstrated that it is possible to induce electron-ion recombination into high-lying Rydberg states with half cycle pulses, and the technique could be a way to produce antihydrogen [9]. However, the recombined atoms are left in high-lying, $n=200$, states, and it would be useful to move them to lower-lying states.

Here we report the first use of strong, chirped microwave pulses to move population from higher to lower Rydberg states. Specifically, we have moved Li Rydberg atoms from $n=75$ to $n=66$ using chirped 7.8–11.8 GHz pulses, at the two-photon resonance between states differing in *n* by 1. In principle the population transfer would probably be more easily effected using the one-photon $\Delta n=1$ resonances, but we are not able to produce adequate microwave powers at the frequencies required for the *n* states we can resolve with our dye laser, $n<80$. Furthermore, in earlier work we observed strong coupling between levels at the two-photon $\Delta n=1$ resonance [10]. We review the principle of the method, describe the experimental technique, present our results, outline a simple quantum-mechanical model, and identify outstanding issues.

Before describing our experiments it is useful to present a simplified picture to illustrate the principle of the adiabatic rapid passage. The important simplification is that we assume that there is a single level for each *n*, whereas in reality there are *n* levels of $m=0$. With this assumption it is straightforward to understand the process using a picture based on the avoided crossings of levels dressed by microwave photons, a picture commonly used for Landau-Zener transitions [11]. Since we use the two-photon resonant $\Delta n=-1$ transitions we are interested in the energy levels with an even number of microwave photons added or subtracted, and in Fig. 1(a) we show a plot of the dressed hydrogenic energy levels from $n=60$ to 80 for the frequency range 7.5–12 GHz. To plot Fig. 1 we have assumed that the energy of each *n* state, in the absence of any coupling, is given in atomic units by

$$W=-\frac{1}{2n^2}-(n-70)2\omega, \tag{1}$$

where ω is the microwave frequency. Unless units are explicitly given, we use atomic units. The $n=70$ energy has no photons added, $n=69$ has two photons added, $n=71$ has two photons subtracted, etc. There is nothing special about $n=70$, and we use it in Eq. (1) to center the graphs of Fig. 1 at $n=70$ and make them as readable as possible. We recall that it is only the difference in slopes of levels which matters for Landau-Zener transitions, not the absolute slopes.

Using the dressed levels given by Eq. (1) the energy levels of states differing in *n* by 1 would cross at the two-photon resonance, but when the two-photon $\Delta n=1$ couplings are included the levels have avoided crossings, as shown in Fig. 1. There are, of course, higher-order multiphoton resonances, but they are too weak to produce visible avoided crossings in Fig. 1 and are always traversed diabatically.

As the frequency of the pulse is chirped, atoms traverse the avoided crossings at the two-photon $\Delta n=1$ resonances adiabatically if the Landau-Zener criterion [11]

$$4n^2\Omega^2 \gg S \tag{2}$$

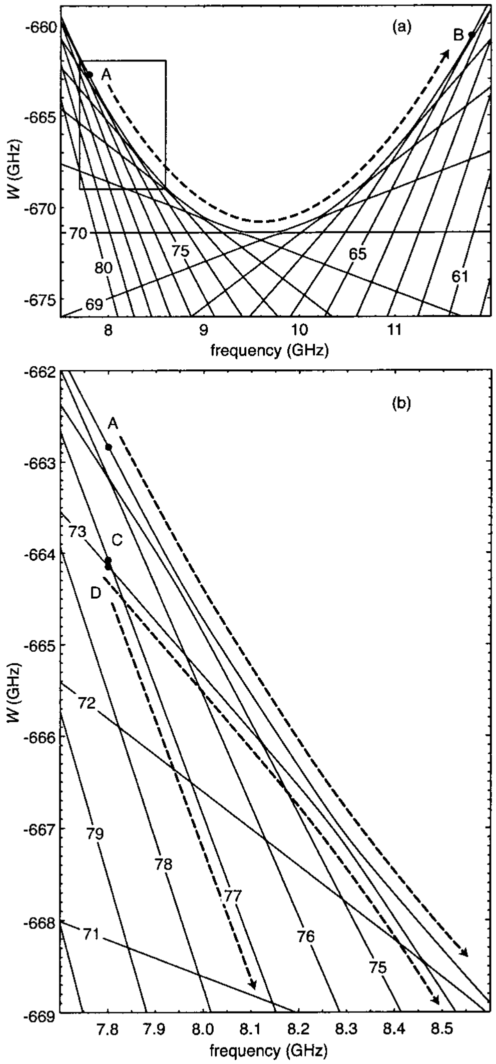

FIG. 1. (a) Microwave dressed energy levels for $n=60$ to 80 versus microwave frequency for constant amplitude microwave field. Even numbers of microwave photons are added to or subtracted from the energy of each state so that the states are degenerate at the two-photon $\Delta n=1$ resonances. Due to the two-photon coupling of the microwave field there are avoided crossings at these $\Delta n=1$ resonances. In a microwave pulse chirped from 7.8 to 11.8 GHz population initially placed in the $n=75$ state, point *A*, is transferred to the $n=65$ state, point *B*, via the highest-lying adiabatic curve, as shown by the broken arrow. (b) An expanded view of the left-hand portion of (a) showing the avoided crossings at low frequency more clearly and the fate of atoms initially placed in $n=75$ and nearby states. Atoms placed initially in $n=75$, point *A*, follow an adiabatic path, shown by its broken arrow, first passing to the $n=74$ level at 7.95 GHz, then to the $n=73$ level at 8.30 GHz, etc., eventually arriving at $n=65$ as shown in (a). An atom initially placed in the $n=77$ level, point *C*, remains in the $n=77$ level as shown by its broken arrow. An atom initially placed in the $n=73$ level makes the transition to $n=74$ and remains there, as shown by its broken arrow.

is met, where Ω is the two-photon matrix element (in GHz) and S is the slew rate of the microwave frequency (in GHz/ns). Assuming that the two-photon avoided crossings are traversed adiabatically let us now consider the effect of a microwave pulse chirped from 7.8 to 11.8 GHz on atoms in different initial states. Atoms initially placed in $n=75$, shown by point *A* in Fig. 1(a), follow the highest-lying adiabatic curve and are transported to point *B*, $n=65$. The atoms undergo a sequence of adiabatic rapid passages through the $\Delta n=-1$ resonances until they reach $n=65$. The frequency chirp stops before reaching the $n=65$ to $n=64$ resonance at 11.90 GHz, and it is for this reason that the atoms are left in $n=65$. While Fig. 1(a) gives a good global view of the process, the details of the avoided crossings are not completely evident, and in Fig. 1(b) we show an expanded view of the left-hand side of Fig. 1(a). As shown in Fig. 1(b) an atom placed initially in $n=75$, at point *A*, undergoes an adiabatic rapid passage to $n=74$ at 7.95 GHz followed by a second one to $n=73$ at 8.30 GHz, and, as shown in Fig. 1(a), this process continues to $n=65$. Now consider the fate of an atom initially placed in $n=77$, at point *C* of Fig. 1(b). It stays in $n=77$ since the microwave frequency is always higher than half the $n=77$ to $n=76$ interval, so the atoms never leave $n=77$; all higher-order multiphoton avoided crossings are traversed diabatically. On the other hand, if the atoms are in $n=73$ at the beginning of the pulse, shown by point *D* in Fig. 1(b), the atoms undergo one adiabatic rapid passage to $n=74$, and are left in $n=74$. In sum, only for $n=75$ do we expect atoms to be transported over a range of n states.

In the experiment a Li atomic beam passes through a piece of WR 90 (x band) waveguide above a septum in the waveguide. The atoms are excited in the waveguide by three counterpropagating 5-ns dye laser pulses tuned to the Li $2s$-$2p$, $2p$-$3s$, and $3s$-np transitions at 670, 803, and 620 nm. The last laser is polarized parallel to the direction of the microwave field so as to excite $m=0$ states.

Immediately (100 ns) after laser excitation, the atoms are exposed to the chirped microwave pulse, typically 400-ns long. Finally, a negative 110-V pulse with a 1.75 μs rise time is applied to the septum. The pulsed field ionizes the Rydberg atoms not ionized by the microwave field and ejects the electrons through a hole in the top of the waveguide. The electrons impinge upon a dual microchannel plate detector, and the time resolved signal is recorded with a digital oscilloscope. Since the electrons have negligible flight time, their arrival time is nearly coincident with the field at which they were ionized. Higher-lying Rydberg states ionize at lower fields, so the time resolved signal can be used to determine the final Rydberg state distribution.

The different feature of this experiment is the chirped microwave pulse. The oscillator is a Hewlett-Packard (HP) VTO 8430 voltage tuned oscillator (VTO) which we chirp from 3.9 to 5.9 GHz by applying a voltage ramp which rises from 0.8 V to 16.4 V. The ramp is generated by an amplified HP 8082A pulse generator. The 10 mW of oscillator power is passed through a Watkins Johnson (WJ) FD93C doubler and a 6-GHz high pass filter, resulting in 0.5 mW of power at 7.8 to 11.8 GHz. We measure the instantaneous frequency of the chirped oscillator by mixing its doubled output with that of a continuous-wave x band oscillator in a WJ M14A mixer and sending the output to an oscilloscope, which displays a pulse at the time the doubled VTO's frequency matches the local

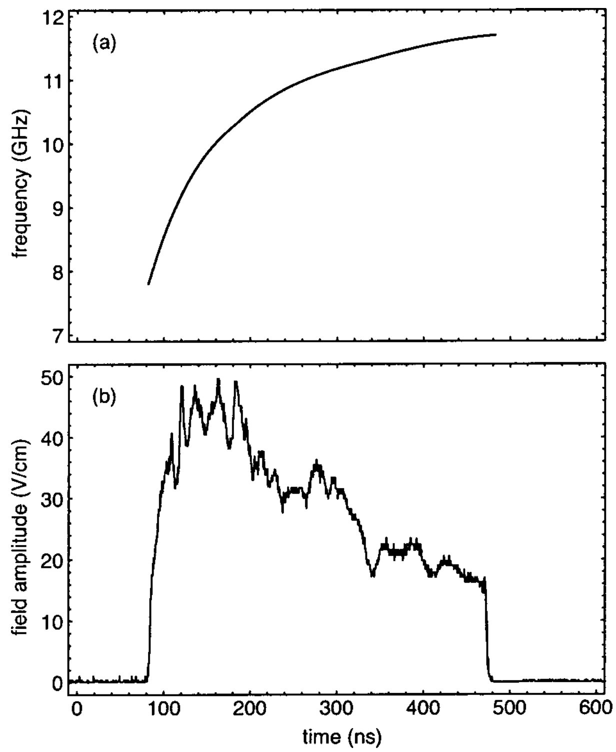

FIG. 2. The chirped microwave pulse (a) frequency dependence, and (b) microwave field amplitude calculated from the detector signal.

oscillator's frequency. In Fig. 2(a) we show the frequency dependence of the chirped microwave pulse. The doubled VTO output is then fed into the local oscillator port of a WJ M86C mixer which is used as a switch and pulse shaper. If we apply a square 400-ns voltage pulse to the intermediate frequency port of the mixer we obtain a flat (constant power) 400-ns microwave output pulse at the radio frequency port. Usually, however, we apply an increasing voltage to compensate for the decrease in the gain of the final amplifier at high frequency. The pulse shaping was done using an arbitrary wave-form generator with 1-ns resolution. After the mixer the microwave pulse passes through HP 8449B and MITEQ MPN4 preamplifiers, and then through a Litton 624 pulsed traveling-wave tube amplifier. The amplified pulse passes through a Narda 792FF variable attenuator and a triangle microwave voltage controlled attenuator (VCA) en route to the waveguide inside the apparatus. The transmitted pulses are brought out of the waveguide, pass through a HP 354A attenuator, and are detected with a calibrated HP 8473C detector. In Fig. 2(b) we show the microwave field in the waveguide calculated from the detector signal. As shown, the field amplitude decreases during the pulse, but by no means monotonically. The sharp variations in the field amplitude are due to the fact that our microwave system does not have a flat response from 7.8 to 11.8 GHz. A particularly attractive feature of adiabatic rapid passage is that these field amplitude variations are not important. What is important is the smooth frequency chirp shown in Fig. 2(a).

When we apply a flat 400-ns microwave pulse at 7.80, 9.95, or 11.80 GHz with no chirp the atoms stay in the same state or are ionized by the field, a result consistent with previous work [10]. In contrast, when we apply a 400-ns pulse chirped from 7.8 to 11.8 GHz we observe a clear evidence for population transfer, as shown by the data of Fig. 3 in which the Li 66*p* to 77*p* states were initially populated, and the microwave field had the pulse shape of Fig. 2(b). For reference, the two-photon 65–66 and 74–75 transition frequencies are 11.71 and 7.96 GHz. A relative power of 40 dB in Fig. 3 corresponds to the pulse of Fig. 2(b). The data of Fig. 3 are obtained by repetitively scanning the attenuation of the VCA while recording the time resolved field-ionization signal on every shot of the laser. The signal amplitude is represented using a linear gray scale, and the time resolved signal for a given microwave pulse amplitude, averaged over many laser shots, can be seen by reading horizontally across the panel. In Fig. 3(a), obtained by laser excitation of the $n=66$ state, for all relative powers up to 31 dB the signal is essentially unchanged, with a peak at 285 ns. For higher powers it disappears, due to microwave ionization. The atoms apparently either remain in the $n=66$ state or are ionized if the microwave pulse is strong enough. Similar results are observed as n is increased from 66 to 70, but as can be seen in Fig. 3, the signal does move to an earlier time, as expected, with the peak occurring at 240 ns for $n=70$. For $n=71$ at relative microwave powers greater than 27 dB the signal moves to a later time, and by $n=75$ the entire signal is at a later time for relative powers exceeding 26 dB, implying complete population transfer to lower states. For $n=77$ and higher, no population transfer is observed.

From the data of Fig. 3 it is evident that for $n=75$ the population is moved to lower n by the chirped pulse. A better determination of the final state can be made by examining the final-state distributions of Fig. 3 at the relative microwave power of 26 dB. In Fig. 4(a) we show, for reference, the $n=75$ state signal at 4 dB. In Figs. 4(b), 4(c), 4(d), and 4(e) we show the final-state distributions obtained when exciting the $n=75$, 70, 66, and 65 states at the relative power of 26 dB. The $n=75$ distribution of Fig. 4(b) matches the $n=66$ but not the $n=65$ distribution, indicating that the final state is $n=66$, not the expected $n=65$ state. It is also interesting that the final-state distribution for $n=70$, Fig. 4(c), is at higher n than for $n=75$, Fig. 4(b). As shown by Fig. 3(e) the $n=70$ final-state distribution does not depend on the microwave field, i.e., an initial $n=70$ population is not moved by the chirped pulse.

The observations shown in Figs. 3 and 4 are clearly consistent with the process of adiabatic rapid passage shown in Fig. 1. Further support for this interpretation comes from using only the last 300 ns of the pulse shown in Fig. 2, in which the frequency was chirped from 9.8 to 11.8 GHz. Using this pulse, which is initially resonant with the $n=70-69$ transition, population is readily transferred from $n=70$ to $n=66$, whereas using the 400-ns 7.8–11.8 GHz pulse no population transfer from $n=70$ to lower n is observed. In the discussion of Fig. 1 we pointed out that atoms initially in $n=73$ do not undergo population transfer to $n=65$ but are expected to undergo a $\Delta n=+1$ transition. The same reasoning applies to atoms initially placed in $n=70$ in the 400-ns pulse.

Initially we expected to need a higher field at the higher

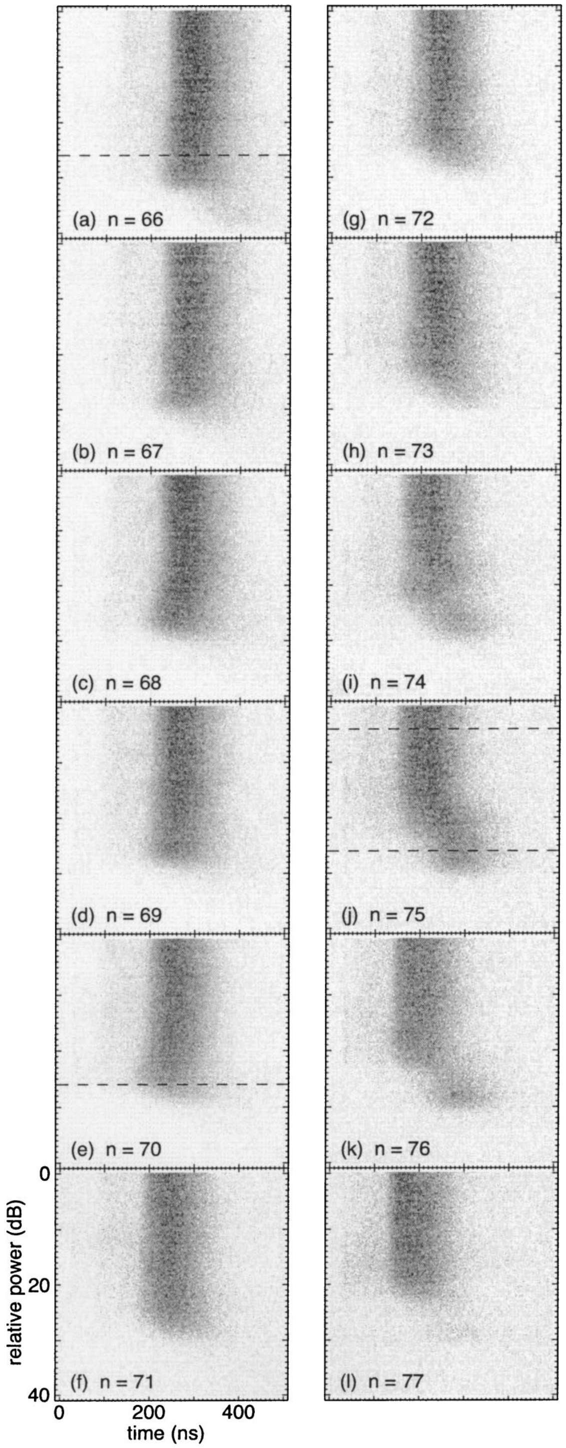

FIG. 3. Observed field-ionization signals when $n=66$ to $75p$ states are initially populated. The horizontal axis is the time, or equivalently the field, at which the field ionization occurs. The vertical axis is relative microwave power, and 40 dB corresponds to the pulse shown in Fig. 1. The signal intensity is shown by a gray scale. For atoms initially excited to $n=66$ to 70 field ionization occurs at the same time until the signal disappears due to microwave ionization, and the entire signal moves to earlier time in each panel as n increases. Beginning with $n=71$ the signal moves to later times, or lower n, as the microwave power is increased, a phenomenon which is most apparent for $n=75$. For $n=77$ no shift in the time of the signal is observed.

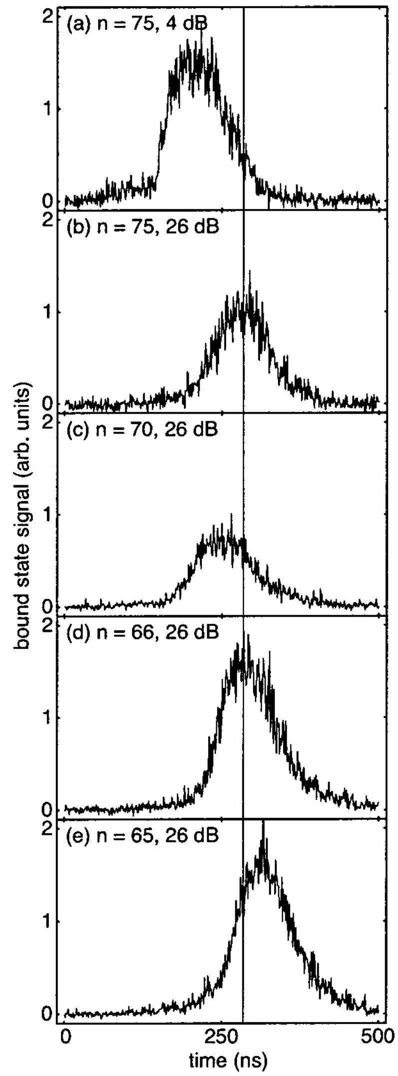

FIG. 4. Time resolved traces for initial excitation of $n=75$ at (a) low relative power, 4 dB and (b) high relative power, 26 dB. Time resolved traces for initially excited (c) $n=70$, (d) $n=66$, and (e) $n=65$ at high relative power, 26 dB. It is apparent that the arrival times of the signals in (b) and (d) are essentially the same, as shown by the solid line through all the given panels, indicating population transfer from $n=75$ to $n=66$. The $n=65$ and $n=70$ signals of (e) and (c) are later and earlier, respectively.

frequencies occurring later in the pulse, because the principal quantum number is lower and the matrix elements are smaller. However, population transfer with the pulse shown in Fig. 2(b) was more efficient than with a flatter pulse, presumably due to the decreasing slew rate of the pulse. We have also investigated the effect of a pulse chirped in the opposite direction, from high to low frequency. While we expected to see population move from higher to lower states, no population transfer was observed. The origin of the difference between chirping up and down is not yet clear.

There are n $m=0$ levels in the Rydberg state of principal quantum number n, so the adiabatic population transfer from $n=75$ to $n=66$ is an inherently complicated problem. At the same time, our experimental results can be understood using the adiabatic rapid passage picture of Fig. 1 which was con-

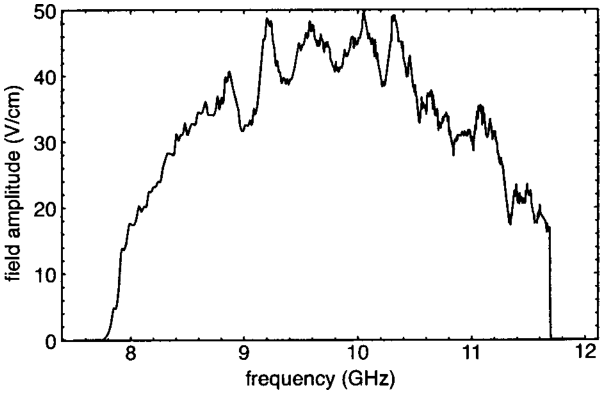

FIG. 5. Microwave field amplitude versus frequency for the pulse shown in Fig. 2.

structed assuming that there is one level for each n. In an effort to provide a simple picture which takes both of these considerations into account we have developed the following model, which reproduces the essential feature of our observations, the adiabatic rapid passage. We have used a hydrogenic model because it is most tractable, and it is a reasonable approximation for several reasons. First, at $n=70$ the average quantum defect of the Li $m=0$ levels is 0.004 [12]. Second, the effect of the core coupling in a field scales as $1/n^4$ while the electric dipole moments scale as n^2, making the electric-field coupling increasingly dominant at high n. Finally, results of microwave ionization experiments with high-lying states of Li closely resemble those obtained with H [10,13].

The energy level diagram of Fig. 1 and the accompanying discussion are based on the assumption that there is one level per n, which is of course not correct, as we have already pointed out. There are many levels and each n level is coupled to at least two $n-1$ levels, which ensures that there must be interference in a sequence of $\Delta n=-1$ transitions. Although interference is missing from such a model, the model is still instructive.

We can use parabolic or spherical coordinates to calculate the two-photon $\Delta n=1$ matrix elements [12,14], and we have chosen to use spherical coordinates because they are more familiar, and there are strict $\Delta \ell = \pm 1$ dipole selection rules. In spherical coordinates a typical two-photon matrix element is the $ns \rightarrow (n-1)s$ matrix element Ω given by

$$\Omega = \sum_{n'} \frac{\langle ns|\mu_z|n'p\rangle\langle n'p|\mu_z|(n-1)s\rangle E^2}{\Delta_{n'}}, \qquad (3)$$

where $\Delta_{n'}$ is the detuning of the real intermediate $n'p$ state from the virtual intermediate state. We have calculated the radial matrix elements required for Eq. (3) using a Numerov algorithm [15], and the largest contributions to the sum of Eq. (3) come from the contributions of the np and $(n-1)p$ intermediate states due to their large matrix elements and small detunings. Their contributions have opposite signs and largely cancel, to roughly 10%. Even so, their residual coupling dominates the contributions from other intermediate states, which fall off very rapidly.

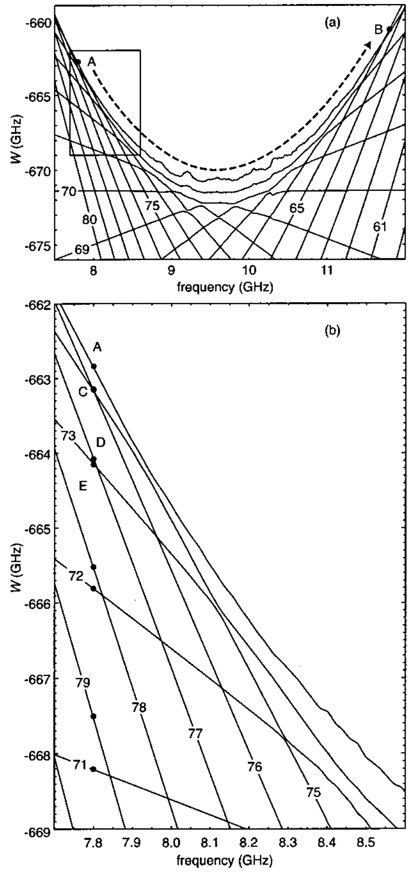

FIG. 6. (a) Microwave dressed energy levels for $n=60$ to 80 versus microwave frequency for the pulse shape of Fig. 5 and a relative power of 25 dB, a peak field of 7 V/cm. Even numbers of microwave photons are added to or subtracted from the energy of each state so that the states are degenerate at the two-photon $\Delta n=1$ resonances. Due to the strong two-photon coupling produced by the microwave field there are large, overlapping avoided crossings at these $\Delta n=1$ resonances, leading to the parabolic shape of the uppermost adiabatic curve. In a microwave pulse chirped from 7.8 to 11.8 GHz population initially placed in the $n=75$ state, point A, is transferred to the $n=65$ state, point B, via the highest-lying adiabatic curve, as shown by the broken arrow. It is clear that the noise in the curve, due to the microwave field variation during the pulse, is unimportant since the adiabatic curves are so far apart. There are also strong avoided crossings at the four-photon $\Delta n=2$ resonances, and the six-photon $\Delta n=3$ resonances, leading to the two approximately parabolic curves under the highest-lying one. These curves can also support adiabatic population transfer over almost ten states. (b) An expanded view of the left-hand portion of (a) showing the avoided crossings at low frequency more clearly and the fate of atoms initially placed in $n=75$ and nearby states. Atoms placed initially in $n=75$, point A, follow an adiabatic path, shown by its broken arrow of (a), first passing to the $n=74$ level at the avoided crossing at 7.95 GHz, then the adiabatic level becomes a parabola in the rising field of the pulse, and the atoms follow it to $n=65$, as shown in (a). Atoms initially placed in $n=74$, point E, follow an adiabatic path to $n=66$. Atoms initially in $n=73$, point D, cross the six-photon $\Delta n=3$ resonance diabatically, then proceed adiabatically. The $n=72$ level, point F, makes an adiabatic transition to $n=75$ and remains there. An atom placed in $n=76$, point E, traverses all avoided crossings diabatically and remains in $n=76$.

We can use the matrix elements of Eq. (3) to compare our observations to this simplified model. For $n=70$, the center of the chirp of our pulse, and a field amplitude of 1 V/cm we calculate a two-photon matrix element $\Omega = 0.0095$ GHz using Eq. (3). Inspecting Fig. 3 we can see that the onset of the population transfer from $n=75$ occurs at a relative power of 25 dB, corresponding to a field of 7 V/cm at the peak of the microwave pulse. With this field the squared $n=70$ to $n=69$ two-photon matrix element of Eq. (3) is given by $\Omega^2 = 0.22\ \mathrm{GHz}^2$. From Fig. 2 we can see that the frequency slew rate at the beginning of the pulse is $S=0.04$ GHz/ns, so the experimentally observed onset of population transfer occurs when $\Omega^2 \approx 5$ s, a value of Ω^2 far exceeding that expected from Eq. (2). However, to have adiabatic passage through ten such two-photon $\Delta n=1$ transitions requires that $4\pi^2\Omega^2 \approx 10S$, or $\Omega^2 = S/4$, a value of Ω^2 which is still twenty times smaller than observed. Apparently a higher two-photon coupling is needed than we would expect from this simple model.

To generate Fig. 1, we diagonalized a Hamiltonian matrix including one level per n for the $n=60$–80 states dressed by an even number of microwave photons. We included the two-photon $\Delta n=1$ matrix element calculated for $n=70$ for all the two-photon couplings. The microwave field amplitude used to calculate Fig. 1 is 2.5 V/cm. While this model has obvious shortcomings, it can be used to explore several of the nonideal aspects of our experiment. First, the microwave pulse is not flat, which is underscored by Fig. 5, a plot of the microwave field versus frequency for the pulse of Fig. 2, at a relative power of 40 dB. Second, a higher microwave field is required than we expected. Figure 6 is a plot analogous to Fig. 1 for the pulse of Fig. 5 at a relative power of 25 dB, i.e., a 7-V/cm peak field. Inspecting Fig. 6 makes several points apparent. First, at the high, relative to Fig. 1, field of Fig. 6 the highest adiabatic curves are essentially parabolas, due to the fact that the avoided crossings overlap each other. Although the usual Landau-Zener criterion of Eq. (2) is no longer applicable, it is clear that adiabatic passage is ensured. Second, the variation in the microwave field amplitude only produces a visible effect at the peak of the pulse on the highest adiabatic curve, the one originating from the two-photon $\Delta n=1$ coupling. This curve is so far removed from the others that the small energy variations are unimportant. Finally, population transfer appears possible not only from $n=75$ to $n=65$, from point A to point B in Fig. 6(a) but from the $n=73$ and 74 states as well using the two parabolalike adiabatic curves below the uppermost one of Fig. 6(a). These two curves become parabolas due to the $\Delta n=2$ and $\Delta n=3$ couplings. While we are not computing these correctly, it is clear that they are important, and Fig. 6(a) shows their qualitative effect.

In Fig. 6(b) we show an expanded view of these curves at the beginning of the pulse, where the amplitude is rising rapidly, as shown by Fig. 5. Starting in $n=74$ or 73, points E and D, respectively, there are clear adiabatic passages leading to $n \approx 65$. For $n>75$, avoided crossings are traversed purely diabatically, as shown by points E and C, representing atoms initially in $n=76$ and 77. For atoms initially put in $n=72$, point F, the avoided crossing with $n=75$ is traversed adiabatically, and the atoms remain in $n=75$. Atoms initially placed in lower-lying states have a similar fate.

While the picture we have presented above gives a reasonable qualitative description of our observations in terms of adiabatic rapid passage, there are problems which are not yet understood. The major one is the issue of interference. In particular there are typically three $\Delta n=1$ transitions, $\Delta\ell = 0, \pm 2$, leading to interference in transitions through successive n states, which we have ignored. A similar situation arises in pulsed field ionization, [16,17] but the present problem appears to be more tractable. There are, of course other open questions, for example, why does the pulse of Fig. 2 work better than a flat pulse?

In conclusion, by exposing Rydberg atoms to a chirped microwave pulse we have been able to transfer the population to lower Rydberg states. The technique appears robust enough to be practical, and it is quite possible that by using a frequency equal to the Kepler frequency the population transfer can be effected with lower microwave power. The adiabatic rapid passage model provides a reasonable qualitative description, but it is evident that the model we have used is not quantitatively correct. We attribute its shortcomings to our ignoring the interference occurring in successive $\Delta n=1$ transitions. Extending our approach to take all the interferences into account is possible, but messy, suggesting that a new way of thinking about the problem would be useful.

This work has been supported by the National Science Foundation. It is a pleasure to acknowledge useful discussions with R. R. Jones and W. M. Griffith.

[1] A. Abragam, *The Principles of Nuclear Magnetism* (Oxford University Press, London, 1961).
[2] E. Murgu, F. Ropke, S. M. Djambova, and T. F. Gallagher, J. Chem. Phys. **110**, 9500 (1999).
[3] R. G. Hulet and D. Kleppner, Phys. Rev. Lett. **51**, 1430 (1983).
[4] P. Nussensveig, F. Bernadot, M. Brune, J. Hare, J. M. Raimond, S. Haroche, and W. Gawlik, Phys. Rev. A **48**, 3991 (1993).
[5] D. J. Maas, D. I. Duncan, R. B. Vrijen, W. J. van der Zande, and L. D. Noordam, Chem. Phys. Lett. **290**, 75 (1998).
[6] B. Meerson and L. Friedland, Phys. Rev. A **41**, 5233 (1990).
[7] T. J. Bensky, M. B. Campbell, and R. R. Jones, Phys. Rev. Lett. **81**, 3112 (1998).
[8] C. Wesdorp, F. Robicheaux, and L. D. Noordam, Phys. Rev. Lett. **84**, 3799 (2000).
[9] G. Gabrielse, D. S. Hall, T. Roach, P. Yesley, A. Khabbaz, J. Estrada, C. Heimann, and H. Kalinowsky, Phys. Lett. B **455**, 311 (1999).
[10] M. W. Noel, W. M. Griffith, and T. F. Gallagher, Phys. Rev. A **62**, 063401 (2000).
[11] J. R. Rubbmark, M. M. Kash, M. G. Littman, and D. Kleppner, Phys. Rev. A **23**, 3107 (1981).

[12] T. F. Gallagher, *Rydberg Atoms* (Cambridge University Press, Cambridge, 1994).
[13] P. M. Koch and K. A. H. van Leeuwen, Phys. Rep. **255**, 289 (1995).
[14] H. A. Bethe and E. A. Salpeter, *Quantum Mechanics of One and Two Electron Atoms* (Springer, Berlin, 1957).
[15] M. L. Zimmerman, M. G. Littman, M. M. Kash, and D. Kleppner, Phys. Rev. A **20**, 2251 (1979).
[16] D. A. Harmin, Phys. Rev. A **44**, 433 (1991).
[17] F. Robicheaux, C. Wesdorp, and L. D. Noordam, Phys. Rev. A **62**, 043404 (2000).