Population transfer in the Na *s-p* Rydberg ladder by a chirped microwave pulse

H. Maeda,[1,2] J. H. Gurian,[1,*] and T. F. Gallagher[1]

[1]*Department of Physics, University of Virginia, Charlottesville, Virginia 22904-4714, USA*

[2]*Department of Physics and Mathematics, Aoyama Gakuin University, Fuchinobe, Sagamihara, Kanagawa 252-5258, Japan*

While quantum defects of $n\ell$ Rydberg states of Na with $\ell \geqslant 2$ are nearly equal to zero, the quantum defects of ns and np states of Na are quite large due to the finite size of its ionic core. Since the Na ns and np states are energetically isolated from the higher angular momentum states of the same n, we can think of the Na ns and np Rydberg states as a specific example of a simple multilevel ladder system consisting of only alternating s and p angular momentum states. Here we report that population transfer in the Na s-p Rydberg ladder can be effectively achieved using a frequency-chirped microwave pulse, which couples only s and p states under suitable conditions.

I. INTRODUCTION

The transfer of population of Rydberg atoms from an initial state of principal quantum number n to one of a very different quantum number n' by a chirped radiation field is of intrinsic and potentially practical interest [1]. By a chirped radiation field we mean one that is rapidly swept in frequency. Meerson and Freidland proposed that hydrogen atoms could be easily ionized by very small chirped microwave fields [2]. Specifically, they considered a one-dimensional, classical hydrogen atom. They showed theoretically that if an atom is exposed to a linearly polarized microwave field at the Kepler frequency $\omega = \omega_K$, the electron's orbital motion becomes phase locked to the microwave field. In terms of the Poincaré surface of section, the orbit is trapped in a moving resonance island. If the frequency is slowly reduced, the electron's motion remains phase locked, reducing the frequency of the electron's orbit and increasing its amplitude. If the microwave field amplitude is unchanged, the electron's motion becomes chaotic, and the atom is ionized. An early experiment along these lines was that of Lambert *et al.* who transported population from Li Rydberg states of $n \cong 75$ to $n \cong 66$ using a microwave field chirped from 7 to 11 GHz. They used a microwave field at half the Kepler frequency (i.e., $\omega = \omega_K/2$), and they described their observations in quantum-mechanical terms as a sequence of adiabatic rapid passages (ARPs) through two-photon $n \to n-1$ resonances of a one-dimensional hydrogenic atom [3]. Maeda *et al.* transported Li atoms from $n = 70$ to 80 using microwave fields chirped from 19 to 13 GHz [4]. At $n = 70$ the Kepler frequency is 19 GHz, and at $n = 80$ it is 13 GHz. Their results are easily understood as a sequence of $n \to n+1$ ARPs through one-photon resonances, roughly analogous to the description given by Lambert *et al.* [3]. In addition, they showed that the electron's motion remains phase locked to the microwave field throughout the frequency chirp, so the experiment also corresponds to the original suggestion of Meerson and Freidland [2], except that the transfer is not to a high enough n that ionization occurs. This experiment, and a subsequent

one, demonstrate that ARP is equivalent to classical phase locking [5]. Mestayer *et al.* have moved K atoms from the extreme Stark states of $n = 350$ to the analogous states of $n = 500$ using a chirped train of unidirectional field pulses [6]. The train was chirped in that the time between the identical pulses was increased. Initially the time between the pulses was equal to the Kepler period at $n = 350$, and at the end of the train it was equal to the Kepler period for $n = 500$. This procedure has the same result as chirping a sinusoidal microwave field over the same frequency range, and they described their results in terms of trapping the electron orbit in a resonance island, a description equivalent to that given by Meerson and Freidland [2].

All of the above cases have been described as one-dimensional systems. In the experiment of Mestayer *et al.*, starting from the extreme Stark states ensures a propensity to remain in the extreme Stark states, but it is not truly a one-dimensional system. A Li atom in a linearly polarized microwave field is certainly not a one-dimensional system since a linearly polarized field at the Kepler frequency couples all the $\ell \geqslant 1$ states of $m = 0$. Here ℓ and m are the orbital and azimuthal angular momentum quantum numbers. In any alkali atom but Li the $ns \to np \to (n+1)s \to (n+1)p \to \cdots$ system, which we term the s-p ladder, is, in a sense, a one-dimensional system. For all the alkali atoms but Li the quantum defects of the ns and np states, δ_s and δ_p, are nonzero and differ by one half. The difference between the Li and Na energy levels is shown explicitly in Fig. 1. In Fig. 1(a) we show the energy levels of Li. In Li $\delta_s = 0.40$, and $\delta_{\ell \geqslant 1} \leqslant 0.05$. In the Li experiments the np states were excited by a laser, and a microwave field at the Kepler frequency coupled all the nearly degenerate states of $\ell \geqslant 1$. In the one-dimensional models the $(n-1)$-fold degeneracy of the $\ell \geqslant 1$ states is ignored [2,7]. In the Na atom, shown in Fig. 1(b), the ns and np quantum defects are $\delta_s = 1.35$ and $\delta_p = 0.85$, while $\delta_{\ell \geqslant 2} \leqslant 0.015$. In Na it is relatively easy to excite either the ns or nd states. Exciting the nd states it should be possible to effect population transfer by chirping a field at the Kepler frequency since the Na $\ell \geqslant 2$ states are completely analogous to the Li $\ell \geqslant 1$ states. On the other hand, exciting the Na ns states and chirping a field at half the Kepler frequency should allow population transfer through the s-p ladder without entering the $\ell \geqslant 2$ states. Here we describe population transfer via the Na s-p

———————
[*]Present address: Laboratoire Aimé Cotton, CNRS, Université Paris-Sud, Bât. 505, Campus d' Orsay, F-91405 Orsay, France.
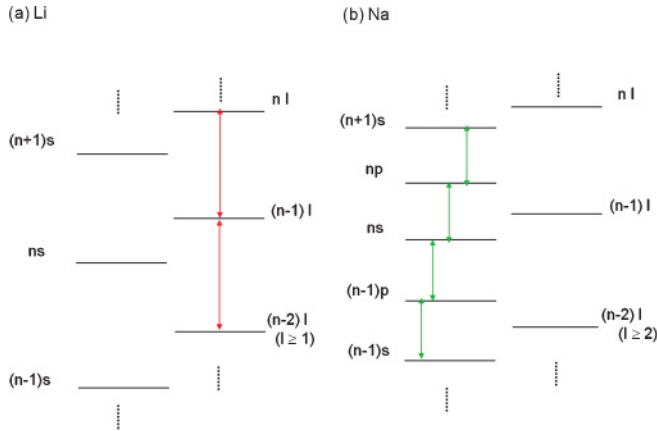
FIG. 1. (Color online) Energy level structure of (a) Li and (b) Na.

ladder to higher-lying n states. This system is one-dimensional in the sense that, over a range of microwave field strengths, only one dipole coupling is possible in each step, from s to p, or p to s. Other couplings, such as p to d, do not occur. It thus has the potential to be an excellent test system for population transfer schemes. In the sections which follow we present a Floquet picture of the population transfer, describe the experimental approach, present our findings, and outline their implications.

II. FLOQUET DESCRIPTION OF POPULATION TRANSFER

In Fig. 2 we show the Na ns and np levels for $58 \leqslant n \leqslant 65$. As shown, the transition frequencies decrease almost monotonically from $n = 58$ to 65. The sequence is not quite monotonic since the ns-np frequency is slightly lower than the np-$(n + 1)s$ frequency. If the sequence of frequencies decreased monotonically, population transfer could occur via a sequence of single photon ARPs [i.e., $ns \rightarrow np \rightarrow (n + 1)s \rightarrow (n + 1)p \rightarrow \cdots$]. Although the frequencies of the sequence of Fig. 2 do not change monotonically, efficient population transfer is still possible. Why the nonmonotonicity
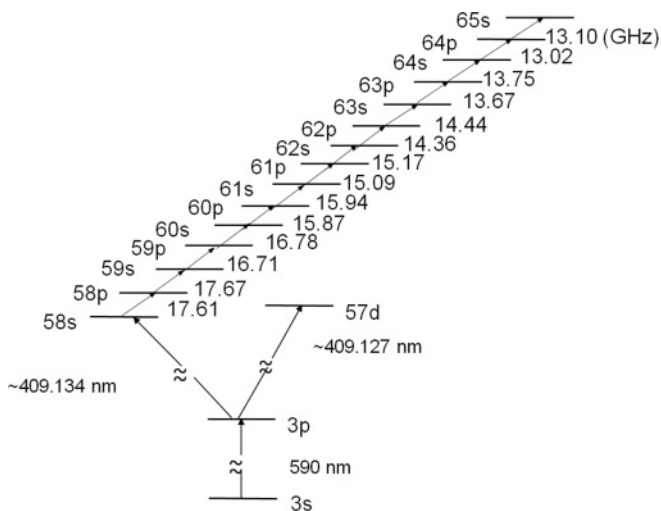
FIG. 2. Laser excitation scheme and energy levels of the Na s-p ladder from $n = 58$ to 65.

of the sequence is unimportant can be easily understood from a Floquet description of the Na ns and np energy levels [8–10]. Figure 3 shows the Floquet energy levels of the Na ns and np states of $58 \leqslant n \leqslant 69$ in 18 to 13 GHz microwave fields of (a) 0 V/cm, (b) 0.1 V/cm, and (c) 0.5 V/cm.

The Floquet energies are obtained from the bare atomic energy levels by adding or subtracting an integral multiple of the microwave frequency ω. Since the integral multiple is arbitrary we are free to choose the zero of the energy scale to be anywhere, and we choose it to be the energy of the 58s state. Accordingly, we define the energies, in atomic units, to be

$$W_{ns} = -1/2(n - \delta_s)^2 + 1/2(58 - \delta_s)^2 - 2(n - 58)\omega \quad (1)$$

and

$$W_{np} = -1/2(n - \delta_p)^2 + 1/2(58 - \delta_s)^2 - 2(n - 58 + 1/2)\omega. \quad (2)$$

We ignore the spin-orbit splitting of the np states, and we consider only the strong ns-np and np-$(n + 1)s$ couplings, which for the microwave field $\hat{z}E \cos \omega t$ are given by $\langle ns|z|np\rangle E = \langle np|z|(n + 1)s\rangle E = n^2 E$. Finally, we make the rotating wave approximation, ignoring far off resonance couplings. The resulting energy levels, obtained by diagonalizing a Floquet Hamiltonian matrix based on the Na ns and np energy levels, are shown in Fig. 3.

As shown in Fig. 2, the transition frequency does not change monotonically along the s-p ladder; the ns-np frequency is slightly lower than the np-$(n + 1)s$ frequency. For this reason in Fig. 3(a) the zero field 58s level crosses the 59s level before it crosses the 58p level. A slow chirp of a weak microwave field, $E < 0.10$ mV/cm, from 18 to 17 GHz would transport 58s atoms to the 59s state via the two photon 58s-59s resonance at 17.64 GHz.

The 58s-58p and 58p-59s level crossings occur at 17.61 and 17.67 GHz, almost the same frequency, and in the field of 0.1 V/cm the result is the double avoided crossing of the 58s, 58p, and 59s states, as shown in Fig. 3(b). A frequency chirp of a 0.1 V/cm microwave field from 18 to 17 GHz transports atoms from the 58s state to the 59s state via the uppermost energy level of Fig. 3(b), that is through the double avoided crossing of the 58s, 58p, and 59s states. If the chirp is continued from 17 to 13 GHz the atoms undergo ARP through a sequence of double avoided crossings, making a sequence of $ns \rightarrow (n + 1)s$ transitions. The first of these, the double avoided crossing of the 59s, 59p, and 60s states, is shown in Fig. 3(b). In population transfer along the upper energy level curve of Fig. 3(b) atoms spend most of the time in ns states, making $ns \rightarrow (n + 1)s$ transitions at isolated double avoided level crossings. When the microwave field is raised to 0.5 V/cm the double avoided crossings overlap, and the upper energy level curves of Fig. 3(c) become smooth. Not shown in Fig. 3 are the Floquet levels of the $\ell \geqslant 2$ states. At 17.6 GHz the Floquet levels from the odd parity $n = 57 \ell \geqslant 2$ levels lie 5 GHz below the double avoided crossing of the 58s, 58p, and 59p states. As we shall see, the onset of significant coupling to these levels limits the range of microwave field strengths over which the Na s-p ladder remains isolated.
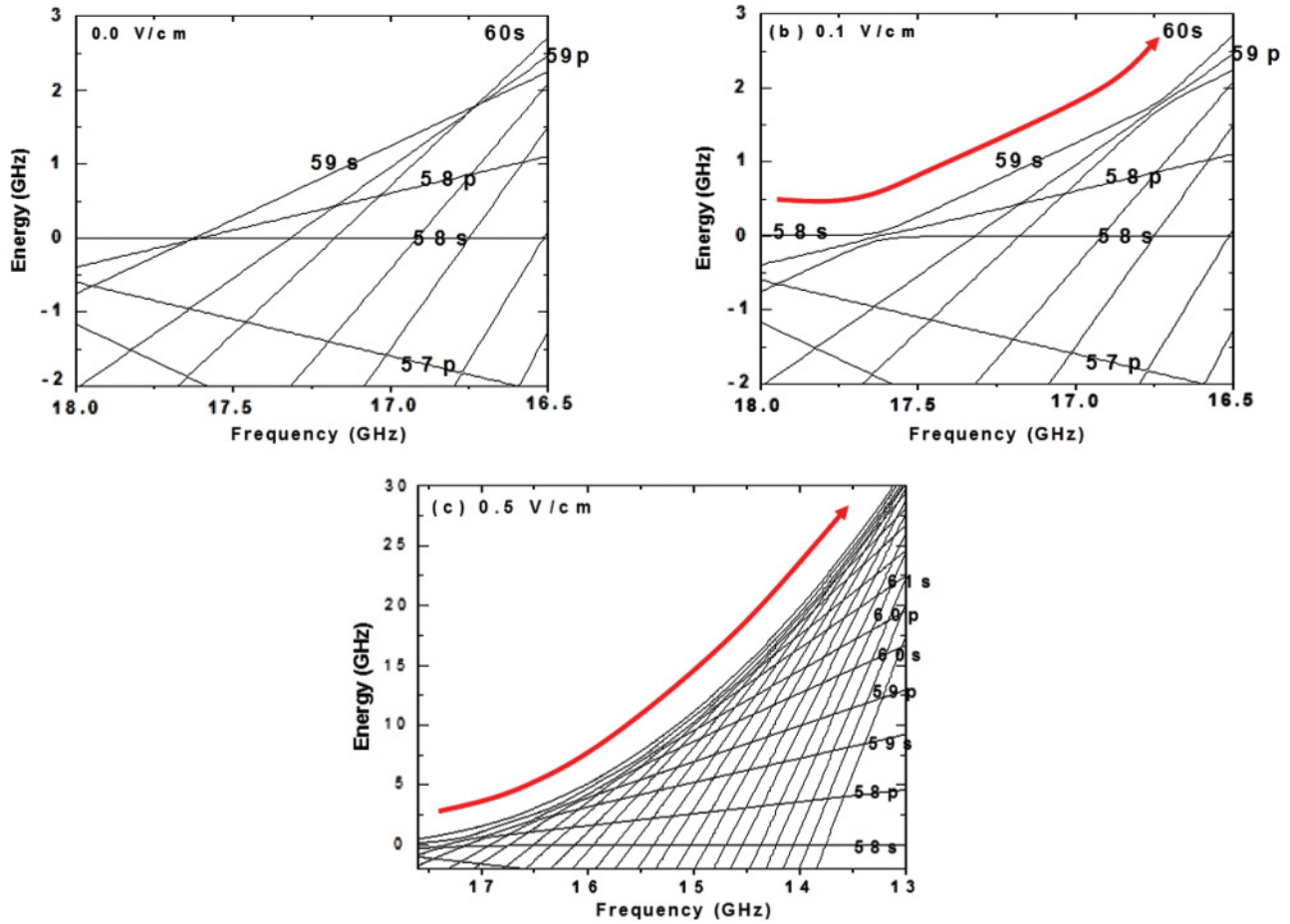
FIG. 3. (Color online) Floquet energy levels of Na ns and np states in microwave fields from 18 GHz to 13 GHz when the microwave field amplitude E is 0, 0.1, and 0.5 V/cm. (a) With $E = 0$ V/cm (i.e., no microwave field) the levels cross at the single and multiphoton resonances. (b) With $E = 0.1$ V/cm the $58s$, $58p$, and $59s$ states exhibit a double avoided crossing at 17.6 GHz, and atoms initially in the $58s$ state pass through it to reach the $59s$ state as the frequency changes from 17.7 to 17.5 GHz. The atoms pass through analogous double avoided crossings to reach higher lying ns states. (c) With $E = 0.5$ V/cm the isolated avoided crossings overlap, generating the smooth curves at the top of the figure. On these curves the energy levels are at all frequencies superpositions of several bare states.

III. EXPERIMENTAL APPROACH

In the experiment a Na beam from an effusive oven passes through a piece of WR-62 rectangular waveguide (1.58 cm × 0.79 cm inside dimensions) through pinholes in the sidewalls of the waveguide, as shown in Fig. 4. At the center of the waveguide it is crossed by two collinear laser beams, which pass through similar holes in the waveguide sidewalls. The laser beams propagate at an angle of $-45°$ to the waveguide and intersect the atomic beam at a right angle. The Na atoms are excited to ns states via the excitation route $3s \rightarrow 3p \rightarrow ns$. We use a pulsed dye laser pumped by the second harmonic of a 20-Hz Nd:YAG laser to drive the first transition from $3s$ to $3p$ at 590 nm. The transition probability from the $3p$ state to the $(n-1)d$ state is six times larger than that to the nearby ns state, and the energy difference between ns and $(n-1)d$ states is quite small. For example, at $n = 58$, the energy difference is only 0.4 cm^{-1}, just a few times larger than the 0.1 cm^{-1} spectral bandwidth of our dye laser. Therefore, we use an amplified blue diode laser for selective excitation of the ns state. the continuous wave (cw) output of the diode laser passes through a 20-mm-long dye cell containing Excalite 411

dye at a concentration of 2×10^{-4} M in p-dioxane. The dye is transversely pumped by the third harmonic of the Nd:YAG laser. The typical output of the diode laser is ∼30 mW, and the typical amplified output from the dye cell is from 20 to 30 μJ/pulse, depending on the pump power of the Nd:YAG laser.

The Na atoms excited to the ns states are exposed to a microwave pulse chirped at -5 MHz/ns. Approximately 50 to 100 ns after the microwave pulse, a negative voltage ramp 1-μs long is applied to a septum in the waveguide to selectively field ionize the Na Rydberg atoms. The field ramp also ejects the electrons from selective field ionization through a 1-mm diameter hole in the top of the waveguide. As Rydberg atoms in different states are field ionized at different fields [11], we can distinguish the final states of the Rydberg atoms subsequent to the microwave pulses by recording the time-resolved electron signal. We detect the electrons with a dual microchannel plate (MCP) detector, capture the signal with a gated integrator, and store the data in a computer for later analysis.

The chirped microwave pulse is produced by a SIVERSIMA V03260P/00 voltage-controlled oscillator
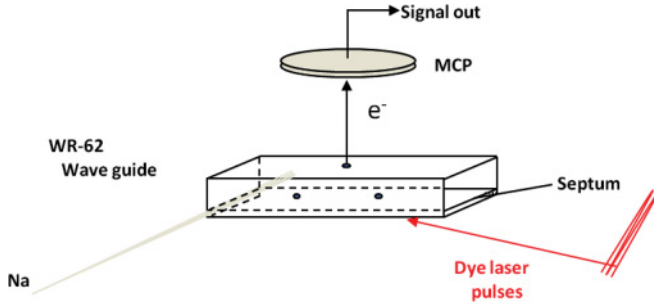
FIG. 4. (Color online) Schematic diagram of the experiment. The Li atomic beam and the exciting laser beams cross at the center of the waveguide, and the chirped microwave pulse is propagated through the waveguide. The septum inside the waveguide allows the application of a field pulse subsequent to the microwave pulse to selectively field ionize the atoms.

(VCO), the frequency of which varies from 13 to 18 GHz as the control voltage is changed from 1 to 12.5 V. To produce a $18 \rightarrow 13$ GHz chirp we use a Hewlett Packard 8112A pulse generator to produce a ramped voltage, which falls from 12.5 V to 1 V in 1 μs. The typical output power of the oscillator is 10 mW. With a HP 11720A pulse modulator we form the output of the VCO into a chirped 1-μs pulse with turn-on and turn-off times of less than 10 ns. The pulse is amplified to powers as high as 300 mW with a MITEQ MPN 4-02001800-23P amplifier and transported to the waveguide inside the vacuum chamber by coaxial cable. The microwave power passes into the waveguide shown in Fig. 4 via a waveguide/coaxial adapter and out of the opposite end of the waveguide by a second waveguide/coaxial adapter. The microwaves pass out of the vacuum system via coaxial cable and are monitored with a crystal detector.

IV. OBSERVATION OF POPULATION TRANSFER IN THE *s-p* LADDER

In Figs. 5(a) (red) and 5(b) (black) we show traces of the field ionization signals of Na 58s and 64s atoms in zero field. In Fig. 5(c) (blue) we show the signal from 58s atoms after exposure to a 0.3 V/cm microwave field chirped from 18 to 13 GHz. As shown, the resulting field ionization signal is similar to the zero field 64s signal of Fig. 5(b). Applying the Landau-Zener criterion for adiabatic passage with our chirp rate of 5 MHz/ns implies that a field of 0.05 V/cm is required, consistent with our observations [12]. In Fig. 5(d) (green) we show the result obtained when 58s atoms are exposed to a $\sim$2 V/cm microwave field chirped from 18 to 13 GHz (green dotted curve). In this case we observe a field ionization signal peaked at a later time than either of the signals of 58s and 64s, and we interpret it as population transfer from the 58s state to the energetically nearby $\ell \geqslant 2$ states of $n = 57$.

We believe the higher ℓ states to be the source of the signal at 800 ns in Fig. 5(d) for two reasons. First, it has been previously observed that the higher ℓ states of high n states ionize at higher fields than do the optically accessible states [13], an observation counter to early descriptions of pulsed field ionization [14]. Second, as stated earlier, the Floquet levels of the odd parity $n = 57$ $\ell \geqslant 2$ states lie 5 GHz below the double
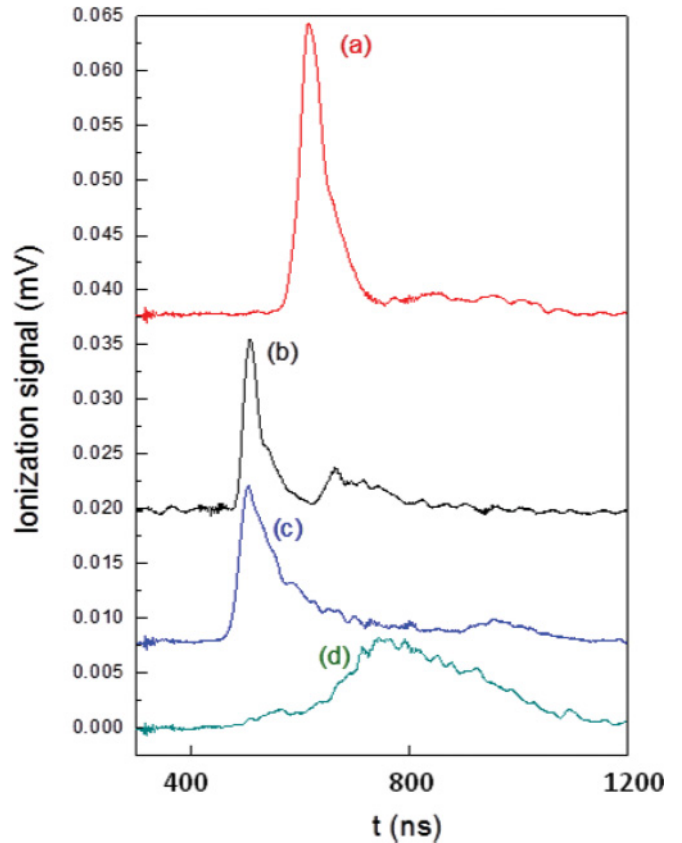

FIG. 5. (Color online) Field ionization signals from (a) (red) 58s and (b) (black) 64s atoms with no microwave field. (c) (blue) Field ionization signal from 58s atoms after exposure to an 18–13 GHz chirped pulse of amplitude $E = 0.2$ V/cm, showing population transfer to the 64s state. (d) (green) Field ionization signal from 58s atoms after exposure to an 18–13 GHz chirped pulse of amplitude $E = 2$ V/cm, showing population transfer to the energetically close $\ell \geqslant 2$ states of $n = 57$.

avoided crossing of the 58s, 58p, and 59s states. If we ignore the effect of the 17.6 GHz field on the $\ell \geqslant 2$ states we expect the states in the *s-p* ladder to overlap with them in a microwave field of 3 V/cm. We observe this feature in Fig. 5 at 2 V/cm, a field slightly lower than expected, in reasonable agreement with our simple estimate. The important point is that the *s-p* ladder is no longer isolated at a microwave field strength of 2 V/cm.

In the panels of Fig. 6 we present grayscale renditions of the time-resolved field ionization signals of Na 58s atoms subjected to pulses with chirps starting at 18 GHz but ending at different frequencies. In each of the panels the microwave field strength E ranges from $\sim$0 to $\sim$12 V/cm. In Fig. 6(a) we show the results obtained for the same 18–13 GHz chirp used to obtain the traces shown in Fig. 5. As the field is increased from 0.08 V/cm to 0.1 V/cm the field ionization signal remains peaked at 600 ns and does not change appreciably. At $E = 0.1$ V/cm the peak in the field ionization signal shifts to an earlier time, 500 ns, the location of the 65s field ionization signal. The field ionization signal remains at this time for $0.1 < E < 2$ V/cm. For $E > 2$ V/cm the signal moves to a later time, exhibiting a broad feature peaked at 700 ns. We attribute this signal to population transfer to the $n = 57$ $\ell \geqslant 2$
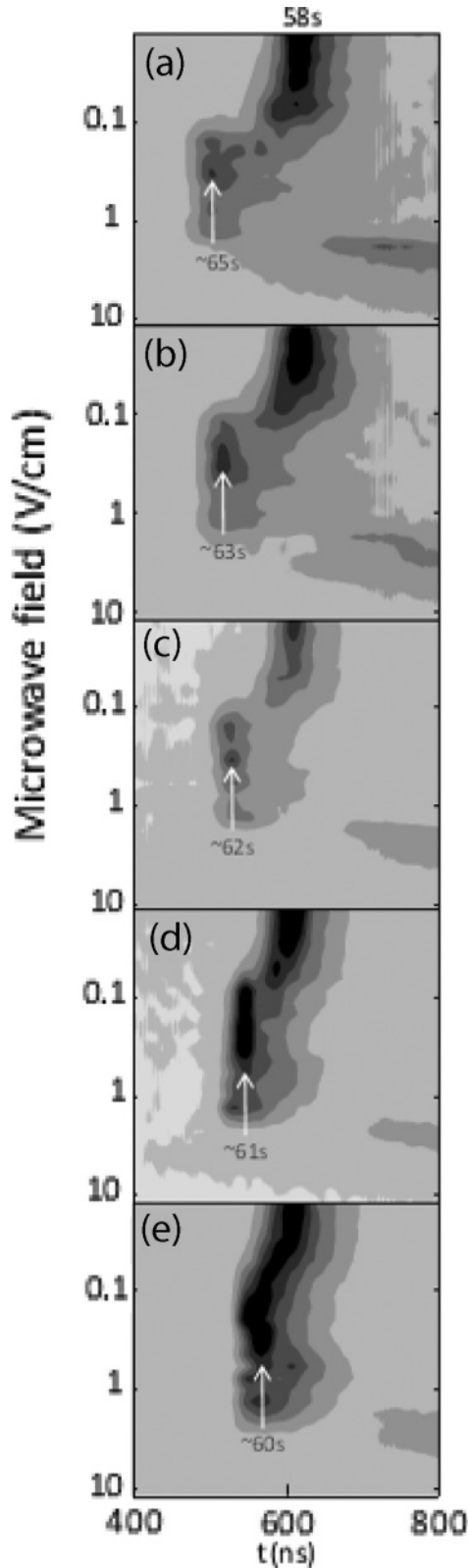
FIG. 6. Field ionization signals of Na 58*s* atoms exposed to chirped microwave pulses with field strengths changing from 0 V/cm to 12 V/cm. The microwave pulses are chirped from 18 GHz to (a) 13 GHz, (b) 14.3 GHz, (c) 15 GHz, (d) 15.8 GHz, and (e) 16.6 GHz. Efficient transfer to a higher *n* state occurs for $E \cong 0.1$ V/cm in all cases. Similarly, the later field ionization signal, attributed to $\ell \geqslant 2$ states of $n = 57$, occurs at $E \cong \sim 2$ V/cm in all cases.

states, as in Fig. 5. At fields approaching ∼10 V/cm the signal disappears due to microwave ionization. Figure 6 underscores the point that over a factor of ten in microwave field strength, a factor of one hundred in power, it is possible to transfer population through the isolated *s-p* ladder.

Figures 6(b)–6(e) show analogous sets of field ionization signals obtained when 58*s* atoms are exposed to chirps ending at frequencies higher than 13 GHz, which transfer population from the 58*s* state to *ns* states of $60 \leqslant n \leqslant 63$. The final states are those expected from Fig. 3. Efficient population transfer occurs at $E \approx 0.1$ V/cm in all cases, which is to be expected since the $n = 58$ dipole matrix elements are the smallest of those that play a role in the population transfer. Finally, note that the onset of the later field ionization signal always occurs at $E \sim 2$ V/cm, suggesting that it is in all cases due to transfer from the 58*s* state to the $n = 57 \ \ell \geqslant 2$ states.

The objective of examining the Na *s-p* ladder is to have a one-dimensional system, and it is thus of interest to show that the nearly hydrogenic $\ell \geqslant 2$ states are not coupled to the *s-p* ladder. Accordingly, we have exposed Na 57*d* atoms to the same 18 to 13 GHz chirped pulse as the 58*s* atoms. The 57*d* state is energetically separated from the 58*s* state by 0.4 cm^{-1}.
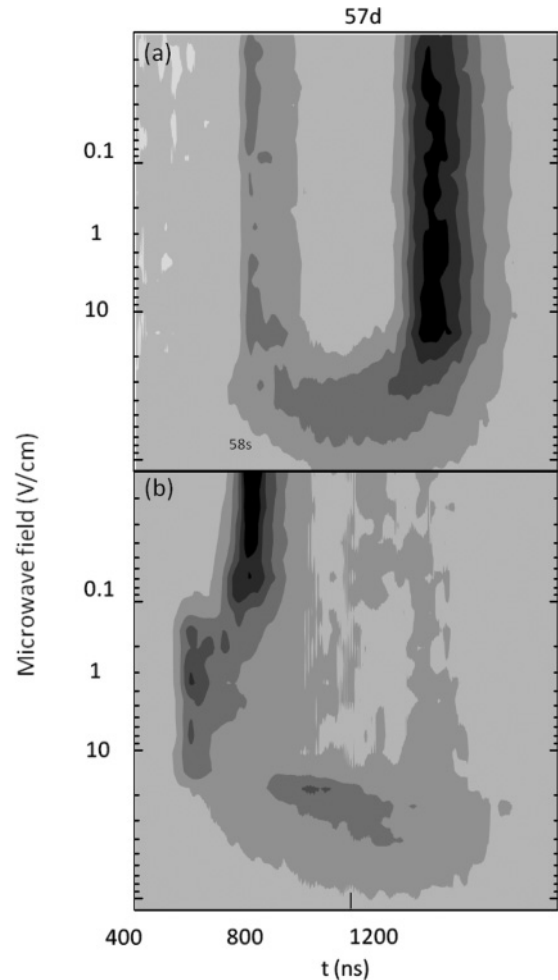

FIG. 7. Final-state distributions of Na (a) 57*d* and (b) 58*s* atoms exposed to chirped microwave pulses with field strengths changing from 0 to 12 V/cm. The microwave pulses are chirped from 18 to 13 GHz. The plot in (b) is the same plot given in Fig. 6(a) except the scale of the horizontal time axis extends to later times.
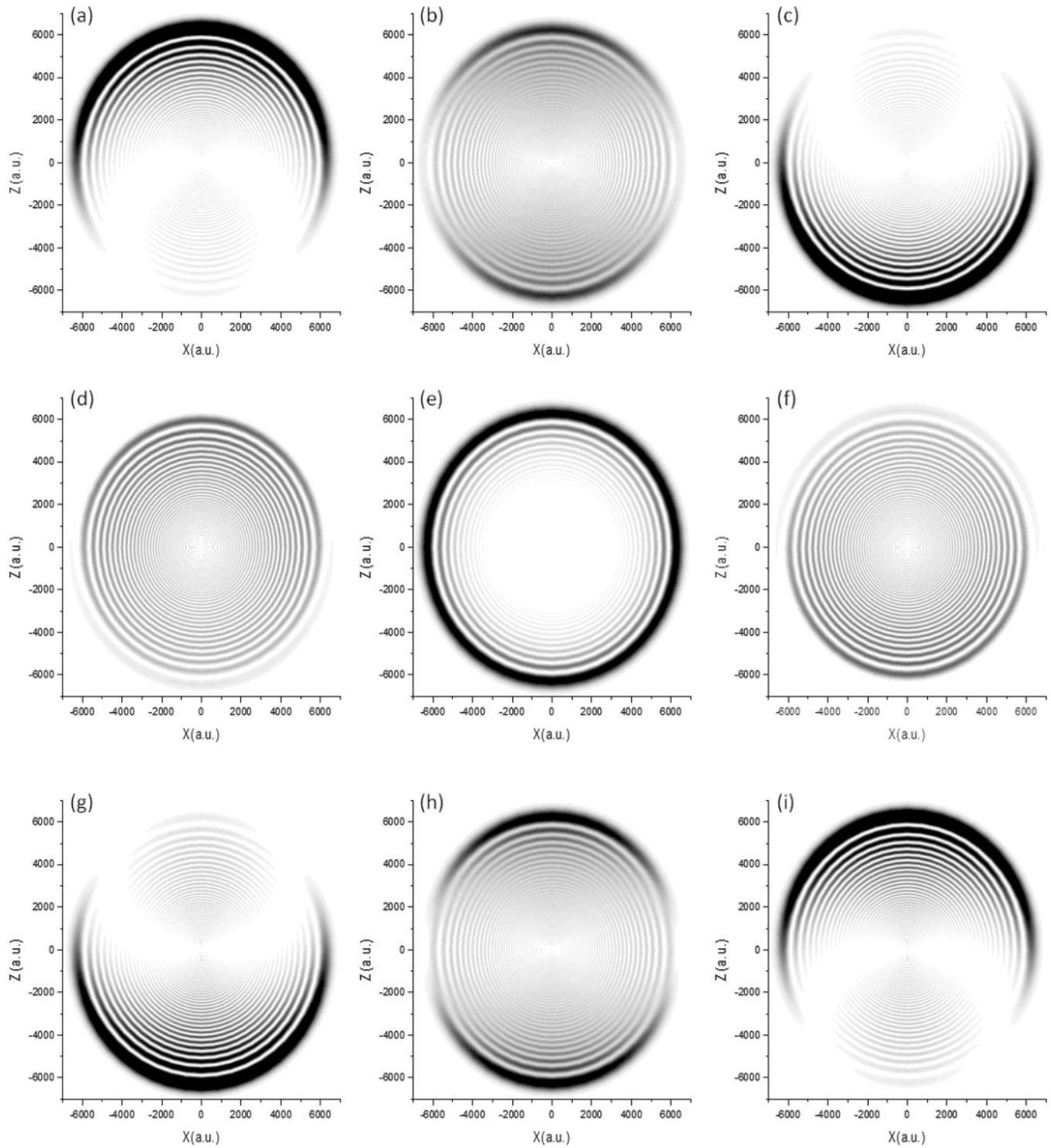
FIG. 8. Electron probability density oscillations for the Floquet eigenstates at the double avoided crossing of the $58s$, $58p$, and $59s$ states in a 17.61 GHz microwave field of 0.1 V/cm amplitude. (a)–(c) The upper energy state of the double avoided crossing at phases of the microwave field $\omega t = 0$, $\pi/2$, and π, respectively. The maximum of the probability density moves from the $+z$ direction to the $-z$ direction; the dipole oscillates π out of phase with the microwave field. (d)–(f) The electron probability density of the center state at the same three phases. There is only a radial oscillation, with no dipole moment, at twice the microwave frequency, the Kepler frequency. (g)–(i) With the same three phases the electron probability density for the lowest energy state moves from lying along the $-z$ axis to lying along the $+z$ axis. The oscillating dipole is in phase with the microwave field.

In Fig. 7(a) we show the field ionization signals obtained when $57d$ atoms are exposed to 18 to 13 GHz chirped pulses with amplitudes from 0 to 12 V/cm. The horizontal time scale of Fig. 7(a) is expanded relative to that of Fig. 6(a), and, for comparison, in Fig. 7(b) we replot the data shown in Fig. 6(a) with the same horizontal scale as Fig. 7(a). With our field ramp, atoms in the $57d$ state are field ionized mainly via two distinct paths, corresponding to adiabatic and diabatic field ionization. The two paths lead to ionization at two different fields and thus at two different times in the field ramp, as clearly seen in the trace of $E = 0$ V/cm in Fig. 7(a). The second peak, which we attribute to diabatic ionization of $|m| = 1$ atoms [11], is not present in the $E = 0$ field ionization signal from the $57s$ state, as shown in Fig. 7(b). Figure 7(a) clearly shows that there is no population transfer from the $57d$ state for $E < 1$ V/cm. Thus chirped pulses with $E < 1$ V/cm can not transfer population into the $57d$ state. Since population transfer from the $58s$ state occurs for $E > 0.1$ V/cm, for $0.1 < E < 1$ V/cm the population transfer is confined to the *s*-*p* ladder and is, in this sense, one-dimensional.

As the microwave field amplitude E is increased to ~ 2 V/cm, the two peaks of Fig. 7(a) disappear and are replaced by a single broad peak at $t = 800$ ns, which we attribute to the $\ell \geqslant 2$ states of $n = 57$. This peak is similar to the broad peak observed when the $58s$ state is exposed to a chirped pulse of amplitude 2 V/cm. It is for this reason that we attribute the field ionization signals shown in Fig. 5 obtained with microwave fields of $E \approx 3$ V/cm to the $\ell \geqslant 2$ states of $n = 57$. Once population enters the $\ell \geqslant 2$ states it remains there unless the atoms are field ionized.

V. DISCUSSION

Population transfer via the *s*-*p* ladder in Na is one dimensional in that the sequence of states through which it passes is unique. Only s and p states are involved; there is never any branching into states of $\ell \geqslant 2$. However, this system is certainly not one dimensional in a spatial sense. Considering the motion in the coherent superposition states formed in a wave packet or at the avoided level crossings in adiabatic rapid passage illustrates this point. In a radial wave packet, consisting of np states for example, the motion of the electron is a purely radial oscillation at the Kepler frequency. In the chirped population transfer to higher n of extreme Stark states the motion is a linear oscillation of growing amplitude, at the decreasing Kepler frequency.

In contrast, the motion in the *s*-*p* ladder is a combination of radial and angular motion. Consider for a moment the coherent superposition of states at the double avoided crossing of the $58s$, $58p$, and $59s$ Floquet states at 17.61 GHz, assuming there to be a small microwave coupling field of 0.1 V/cm, as shown in Fig. 3(b). There are three superposition states, or Floquet eigenstates. The highest energy eigenstate shifts up in

energy with increasing microwave field, and the lowest energy state shifts down in energy. The central eigenstate exhibits only a negligible shift to higher energy. In each of the three states there is an oscillation in the electron probability density, which is shown for half a microwave field cycle in Fig. 8. As expected from the energy shifts, the lower and upper energy states have dipole moments which oscillate in and π out of phase with the microwave field. In the upper state, shown in Figs. 8(a)–8(c), the probability density oscillates from lying in the $+z$ direction to lying in the $-z$ direction, while in the lower energy state the probability density oscillation is reversed, as shown in Figs. 8(g)–8(i). These two states have significant components of all of the $58s$, $58p$, and $59s$ states. Although the motion may appear to be largely angular, the motion is due to the difference in energies, which comes from the radial functions. As pointed out by Yeazell and Stroud, angular wave packets in alkali atoms move because of the differing energies of different ℓ states, which has its origin in the non-Coulombic differences in the radial potentials of different ℓ [15]. Coherent superpositions of degenerate hydrogen states do not move, as exemplified by hydrogenic Stark states.

As shown by Figs. 8(d)–8(f), the oscillation of the electron probability density in the center state is a radial breathing motion, which has no dipole moment, as expected from its lack of an energy shift. Furthermore, the oscillation is at twice the microwave frequency, which is very close to the Kepler frequency. The center state is composed of the $58s$ and $59s$ states with only a very small $58p$ admixture. It is essentially a radial wave packet. The very different eigenstates at these double avoided crossings show clearly that the *s*-*p* ladder is not spatially a one-dimensional system.

VI. CONCLUSION

We have demonstrated that it is possible to coherently transfer population via the *s*-*p* ladder in Na, a system in which a unique sequence of states is responsible for the population transfer, unlike other systems that have been explored experimentally. In this sense it is equivalent to the one-dimensional problem considered by Meerson and Freidland [2]. While the process is one dimensional in the above sense, it is clearly not one dimensional spatially. Nonetheless, since the pathway is unique, this system should be of interest as a testing ground for such processes.

[1] S. Andresen *et al.*, Phys. Rev. Lett. **106**, 025002 (2011).

[2] B. Meerson and L. Friedland, Phys. Rev. A **41**, 5233 (1990).

[3] J. Lambert, M. W. Noel, and T. F. Gallagher, Phys. Rev. A **66**, 053413 (2002).

[4] H. Maeda, D. V. L. Norum, and T. F. Gallagher, Science **307**, 1760 (2005).

[5] H. Maeda, J. Nunkaew, and T. F. Gallagher, Phys. Rev. A **75**, 053417 (2007).

[6] 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).

[7] A. Buchleitner, D. Delande, and J. Zakrzewski, Phys. Rept. **368**, 409 (2002).

[8] H. Maeda and T. F. Gallagher, Phys. Rev. Lett. **92**, 133004 (2004).

[9] H. Maeda and T. F. Gallagher, Phys. Rev. A **75**, 033410 (2007).

[10] J. H. Shirley, Phys. Rev. **138**, B979 (1965).

[11] T. F. Gallagher, *Rydberg Atoms*, (Cambridge University Press, Cambridge, 1994).

[12] J. R. Rubbmark, M. M. Kash, M. G. Littman, and D. Kleppner, Phys. Rev. A **23**, 3107 (1981).

[13] K. M. Dietrick, M.S. thesis, University of Virginia, 1990 (unpublished).

[14] T. F. Gallagher, L. M. Humphrey, W. E. Cooke, R. M. Hill, and S. A. Edelstein, Phys. Rev. A **16**, 1098 (1977).

[15] J. A. Yeazell and C. R. Stroud Jr., Phys. Rev. A **35**, 2806 (1987).