Production of Circular Rydberg States with Circularly Polarized Microwave Fields

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(Received 18 July 1994)

We describe an experimental method of producing circular Rydberg states using a circularly polarized microwave field. A Na s state is excited in the circularly polarized microwave field, and as the field is turned off, it makes an adiabatic rapid passage through a multiphoton resonance and evolves adiabatically into the circular state. Minimal optical selectivity is required and the field can be turned off quickly.

PACS numbers: 32.80.Rm

Circular Rydberg atoms, those with $|m| = l = n - 1$, are required for a diverse set of experiments focused on the Rydberg constant [1], collision cross sections [2], cavity quantum electrodynamics [3,4], and stabilization against ionization by strong electromagnetic fields [5]. Here n, l, and m are the principal, angular momentum, and azimuthal orbital angular momentum quantum numbers. Circular states are not optically accessible from low lying atomic states, and, to date, two methods have been used to prepare them. The first is the adiabatic rapid passage method of Hulet and Kleppner [6]. They ramped a static field in the presence of a perpendicular oscillating field to convert an optically accessible Rydberg state of Li to the $n = 19$ circular state by a sequence of adiabatic rapid passages through single photon resonances. Nussenzvieg *et al.* have recently shown how this technique may be extended to $n = 50$ states by adding a small magnetic field [7]. The second approach is the crossed field approach suggested by Delande and Gay [8] and first realized by Hare *et al.* [9]. Atoms are excited to the highest energy Stark state in a strong electric field and pass adiabatically into a region of weak electric field and a perpendicular strong magnetic field. The atoms remain in the highest energy state, which is the circular state in the magnetic field.

Here we report the use of a new method to produce spatially oriented circular states of Na. A Na s state is excited in a circularly polarized microwave field, and as the field is turned off, it makes an adiabatic rapid passage through a single multiphoton resonance and evolves into the circular state. This method differs from the method of Hulet and Kleppner [6] in having a single adiabatic rapid passage rather than a sequence of them, and it is a variant of a proposal to use the circularly polarized fields by Molander *et al.* [10]. The principle of the method is easily understood if we transform the problem to a frame rotating with the microwave field so that the field is static and does not drive transitions. Consider a field which has amplitude E, lies in the horizontal x-y plane, and rotates about the z axis at angular frequency ω in the laboratory frame. The phase of the field is chosen so that it lies along the $+x$ axis in the rotating frame. To find the eigenenergies and states in the rotating frame, we first transform the zero field Na eigenstates to the rotating frame. A zero field Na nlm state with energy W has a laboratory frame wave function given in atomic units by $\Psi_{nlm}(\bar{r}, t) = R(r)\Theta(\theta)e^{im\phi}e^{-iWt}$, where r, θ, and ϕ are the Rydberg electron's polar coordinates relative to the Na^+. Explicitly, r is the distance, θ is the polar angle relative to the z direction, and ϕ is the azimuthal angle. Using the rotating frame coordinates r_R, θ_R, and ϕ_R, where $r = r_R, \theta = \theta_R$, and $\phi = \phi_R + \omega t$, we can rewrite the wave function as $\psi_{nlm}(\bar{r}, t) = R(r_R)\Theta(\theta_R)e^{im\phi_R}e^{-i(W-m\omega)t}$. The spatial wave function is unchanged, but there is an energy shift of $-m\omega$ [11,12]. Using these wave functions, we can diagonalize the Hamiltonian matrix in the rotating frame with an added field term, using the same procedures used for static fields [13]. Unlike a static field, m is not conserved, but $l + m$ is either even or odd.

In Fig. 1 we show the energy levels of the $l + m$ even Na states near $n = 21$ as a function of the amplitude of a field rotating at a frequency of $\omega/2\pi = 8.026$ GHz. As shown by Fig. 1, the zero field levels are displaced by $-m\omega$, so the transformation to the rotating frame has the same effect as adding a B field of ω/μ_0, where μ_0 is the Bohr magneton. As the field is increased from zero, the Stark effect splits the levels emanating from the high l $n = 21$ states, and the lowest energy Stark state, which is the adiabatic continuation of the circular $m = l = 20$ state, apparently crosses the $22s$ state at $E = 252$ V/cm. In fact, there is an avoided crossing, and the separation of the levels is computed to be 210 MHz, as shown by the inset of Fig. 1.

The basic notion of the experiment is to excite the Na atoms with a pulsed dye laser from the $3p$ state to the $22s$ state at a field between the first two avoided crossings, at 252 and 277 V/cm. The field is then reduced slowly to a field below the first avoided crossing, traversing the avoided crossing in a time long compared to 1/210 MHz, to ensure an adiabatic rapid passage from the $22s$ state to the lowest energy Stark state. Finally, the field is reduced more rapidly to zero, and the Stark state evolves adiabatically into the circular state. The last step can be

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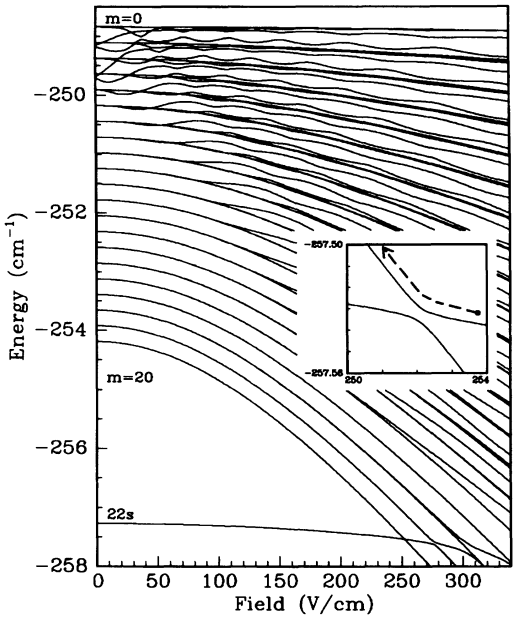

FIG. 1. The Na $22s$ and $n = 21$ $m \geq 0$ energy levels in a frame rotating at 8.026 GHz as a function of the rotating 8.026 GHz field amplitude. The displacement of the levels by $-m\omega$ is quite evident. The lowest Stark state, which is the adiabatic continuation of the zero field circular $n = 21, l = m = 20$ state apparently crosses the $22s$ state at 252 V/cm. As shown in the inset, there is an avoided crossing. To produce the $n = 21$, circular state atoms are excited to the $22s$ state at a field of 255 V/cm; then the field is slowly reduced through the avoided crossing to produce an adiabatic passage to the lowest Stark state; then as the field is more rapidly reduced to zero and the Stark state evolves into the circular state.

done rather quickly since the zero field states are 8 GHz apart. The last step of this method is essentially the proposal of Molander *et al.* [10], and the mathematical similarity of their method to the crossed field method has been pointed out by Chen *et al.* [14]. The major attractions of our method are twofold. First, the laser bandwidth and power requirements are minimal. Only the $22s$ state absorbs the laser light, so even though our laser bandwidth of 0.9 cm^{-1} overlaps several Stark states, it does not matter since they do not absorb the light. Also, very nearly the full $3p$-$22s$ oscillator strength is available. Second, the process can be quick, because of the 8 GHz level separation in the rotating frame. As a result, the atoms do not move far, and very little redistribution due to blackbody radiation occurs, even at room temperature [15]. If the sense of field rotation is reversed, the $m = -l$ circular state is populated instead of the $m = +l$ state.

To produce a circular state and verify its identity requires that we be able to produce and turn off a circularly polarized field in the x-y plane and then apply a pulsed ionizing field in the z direction. While an open Fabry-Pérot cavity should meet the microwave requirements [12], it is awkward for field ionization, and we have used a cylindrical cavity instead. It is constructed of two identical brass halves with the cavity symmetry axis vertical, in the z direction, as shown in Fig. 2. The inside length and diameter of the cavity are 1.98 and 6.40 cm, and the separation between the two halves is 0.51 cm. We operate the cavity on two degenerate TE_{111} modes at a frequency of 8.026 GHz. On the axis of the cavity one has its E field in the x direction and the other in the y direction. By driving the x and y polarized modes to produce fields with the same amplitude and a 90° relative phase shift we produce a circularly polarized field in the x-y plane. The orthogonally polarized fields are coupled into the cavity through 6.68 mm diameter irises in the top and bottom of the cavity from orthogonally polarized waveguides. Each waveguide is cut off to the other polarization, minimizing the coupling between the cavity modes. When the cavity is aligned there is a 0.3 MHz difference in the center frequencies of the modes, and their Q's are 3519 and 3535. The Q's must be very nearly the same, or the field in the cavity rapidly becomes elliptically polarized as the modes decay.

To allow the application of an ionizing field in the z direction we insulate the feed waveguide from the bottom half of the cavity with a 0.5 mm thick teflon sheet. It allows us to put up to 11 kV on the lower half of the cavity leading to ionizing fields as high as 5.5 kV/cm, enough to ionize circular states of $n \geq 20$. Electrons ejected from atoms at the center of the cavity pass through the iris in the upper cavity half and through a 2 mm diameter hole in the upper feed waveguide, as shown in Fig. 2.

The microwave power originates in a Hewlett-Packard (HP) 83620A synthesized sweep oscillator which is kept at 8.026 GHz. The power level is controlled externally. The output of the oscillator is amplified in a Litton 624 gated traveling wave tube amplifier which can produce up to 100 W. The output of the amplifier is split into two

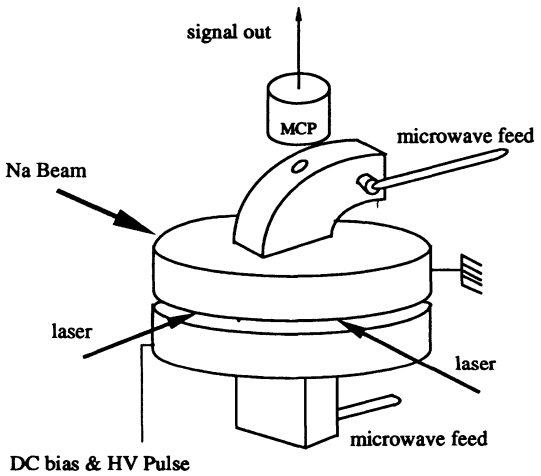

FIG. 2. Schematic diagram of the apparatus.

arms to feed the x and y polarized cavity modes. One arm contains an HP X885A phase shifter, an HP 8495 0–70 dB step attenuator, and a 20 dB directional coupler with an HP 432A power meter. The second arm contains a Waveline 622 precision rotary vane attenuator. There is a circulator in each arm just before it enters the vacuum chamber. With this arrangement we are able to produce circularly polarized fields in the cavity with an amplitude of 1100 V/cm and measure them with an accuracy of 7.3%.

In the experiment a Na beam passes between the two halves of the cavity, as shown by Fig. 2. The atoms are excited by two pulsed dye laser beams, which also pass between the two halves of the cavity. One beam, tuned to the $3s$-$3p$ transition at 589 nm, crosses the Na beam at a right angle 1.6 mm upstream from the center of the cavity. The second beam, tuned to the $3p$-$22s$ transition at 414 nm, is perpendicular to the first and counterpropagating to the atomic beam. The timing sequence of the experiment is as follows. The lasers fire at time $t = 0$ μs. The oscillator is turned on at $t = -8$ μs, and the amplifier is gated on for 4.6 μs starting at $t = -4$ μs, so the field in the cavity is at a steady state value of 255 V/cm when the lasers fire. The power of the oscillator is reduced linearly between $t = 10$ ns and $t = 600$ ns, and the field in the cavity drops from 255 to 173 V/cm during this time. This time is long enough that more than 98% of the atoms should pass adiabatically from the $22s$ state to the lowest Stark state. At $t = 600$ ns the oscillator is turned off, and the field in the cavity decays with the cavity time constant of 70 ns. The field ionization pulse begins at $t = 1.6$ μs and has a 700 ns rise time. We detect electrons from atoms ionized by the field pulse with the microchannel plate detector shown in Fig. 2. The arrival time of the electrons implies the ionizing field, and the calibration is checked using the known ionization fields of the Na ns states. The signal from the microchannel plates goes to a digital oscilloscope and a gated integrator. The whole procedure is repeated at the 10 Hz repetition rate of the laser. Typically, a small static field of 2.5 V/cm in the z direction is present, and the magnetic field is less than 10 mG.

Using the timing sequence outlined above, we have excited the Na $22s$ state at three different peak microwave fields, below, at, and above the avoided crossing with the lowest $n = 21$ Stark state. Experimentally, we find the avoided crossing at 247 V/cm, not at 252 V/cm as shown in Fig. 1, but well within the uncertainty of the field measurement. To minimize confusion in comparisons to Fig. 1 we give all experimental microwave fields after multiplying them by 252/247. In Fig. 3 we show the resulting field ionization signals. With $E = 249$ V/cm, below the avoided crossing, the field ionization signal, shown in Fig. 3(a), is identical to that of the $22s$ state in zero field. At $E = 252$ V/cm, the field of the avoided crossing, we excite both states with the laser and there are

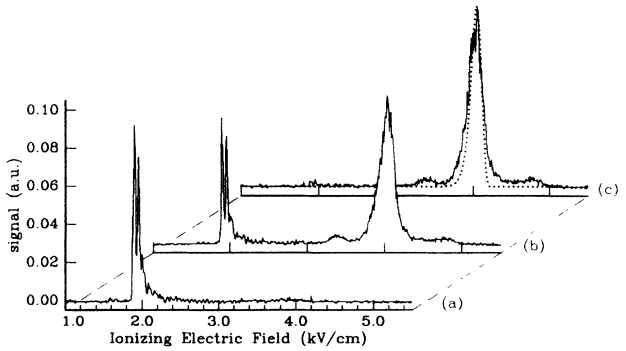

FIG. 3. The field ionization signals when the $22s$ state is excited in three different circularly polarized microwave field amplitudes. (a) For $E = 249$ V/cm, below the avoided crossing, only the $22s$ field ionization signal at $E_I = 1.92$ kV/cm is visible. (b) For $E = 252$ V/cm, at the avoided crossing, half the signal is in the $22s$ state and half is in the $n = 21$ circular state, at $E_I = 4.06$ kV/cm. (c) $E = 255$ V/cm, above the avoided crossing, nearly all the signal matches the calculated ionization signal for the $n = 21$ circular state shown by the dotted line. The small peaks at $E_I = 3.4$ and 4.8 kV/cm are attributed to the $n = 20$ and 22 circular states. The signals of (b) and (c) are multiplied by three relative to the signal in (a); the number of atoms detected is the same in all three cases.

two features in the field ionization signal, one due to the $22s$ state and one due to the $n = 21$ circular state. Finally, at $E = 255$ V/cm, a field above the avoided crossing with the lowest Stark state but below the next one, is, by far, the largest feature visible in the $n = 21$ circular state, as shown in Fig. 3(c). It is in excellent agreement with the signal calculated using the hydrogenic ionization rates of Damburg and Kolosov [16] and the 8×10^9 V/cm s slew rate of our ionizing pulse. In Fig. 3(c), the $22s$ signal has almost vanished, but there are small signals peaked at 3.4 and 4.8 kV/cm, the ionization fields of the $n = 20$ and 22 circular states, which are populated from the $n = 21$ circular state by blackbody radiation [15]. When the field is raised beyond $E = 273$ V/cm, transfer to lower l and m states of $n = 21$ begins to occur. Comparing the areas under the signals of Fig. 3(c) we find that 1% of the atoms remain in the $22s$ state and 90% are left in the $n = 21$ circular state. Of the remaining 9%, 4% and 5% are transferred to the $n = 20$ and 22 circular states, respectively, by blackbody radiation [15]. If the apparatus were cooled, 99% of the atoms would presumably be left in the $n = 21$ circular state.

The tolerances of the method constitute an important factor in determining its usefulness. From the level diagram of Fig. 1 it is apparent that the field amplitude must be between the first and second anticrossings. The microwave field must be controlled to $\pm 9\%$ and the power to $\pm 18\%$ (0.7 dB), not a terribly stringent requirement. On the other hand, ellipticity in the polarization due to imperfect phase and amplitude adjustments can be a problem. Ellipticity adds a field oscillating at 2ω in the

rotating frame, and this field can drive resonant transitions between the states of Fig. 1. In Fig. 4 we show the field ionization signals with the correct amplitudes of both modes, $E = 260$ V/cm, but with phase errors of $\Phi = 3°$, 5.5°, and 7.5° from circular polarization. A phase error of $-\Phi$ leads to the same field ionization signal as $+\Phi$. The circular state signal is unchanged with a $\pm 3°$ phase error and is reduced by half with $\pm 5.5°$ of phase error, which corresponds to an oscillating field with 5% of the amplitude of the rotating field. With a phase error of $\pm 7.5°$, the fraction of atoms left in the circular state is markedly diminished. Keeping the phase error to $\pm 3\%$ is not particularly difficult. Ellipticity caused by amplitude error has comparable effects. When the power into one of the arms is changed by ± 0.5 dB from circular polarization, the signal from the circular state is diminished by half. A 0.5 dB error corresponds to an oscillating field amplitude which is 6% of the rotating field amplitude. We attribute the deleterious effects of ellipticity to processes occurring at high fields near the avoided crossings. While we have little direct experimental evidence, we presume that when the Stark shifts are less than the frequency, ellipticity is not a severe problem. This notion is supported by the calculations of Molander *et al.* [10], which indicate that ellipticity is tolerable in the regime well below the avoided crossing with the s state. A static field of ± 2.5 V/cm in the z direction is always adequate to overcome stray fields. With field it is not necessary to cancel the Earth's magnetic field.

With an operating frequency of 8.026 GHz and rotating fields of 255 and 366 V/cm we have observed the production of $n = 21$ and $n = 20$ circular states. We cannot produce $n \geq 22$ circular states since their avoided crossings with the $(n + 1)s$ states occur at such low fields that they are too small to be usable. A lower frequency would alleviate this problem. We have observed the complete depopulation of the $18s$–$21s$ states at fields within 3% of the calculated fields of their avoided crossings with the lowest Stark states which are the adiabatic continuations of the $n = 17$–20 circular states. We think we are producing these circular states, but we cannot explicitly observe them because we cannot field ionize them. With our maximum microwave field of 1100 V/cm at 8.026 GHz we cannot depopulate the $17s$ state. However, the same field at 9.4 GHz would be adequate to depopulate it.

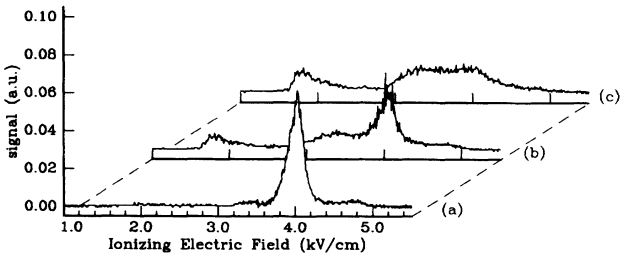

FIG. 4. Field ionization signals with a field amplitude of 255 V/cm and three phase detunings Φ from circular polarization (a) $\Phi = 3°$; the circular state production is unchanged from Fig. 3(c). (b) $\Phi = 5.5°$; the circular state production is reduced by half. (c) $\Phi = 7.5°$; most atoms are left in states other than the circular state.

In conclusion, the technique described here is an efficient and rapid way of producing circular states. It is quite general and can be optimized for a range of n by the choice of microwave frequency. With minor modifications it should also be possible to produce elliptic states using this approach, although it is less natural than using the crossed field method [17].

It is a pleasure to acknowledge M. A. Gatzke for a critical reading of the manuscript. This work has been supported by the Air Force Office of Scientific Research.

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