Preparation of high-principal-quantum-number "circular" states of rubidium

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We describe a rubidium circular-state-preparation technique suited to high principal quantum numbers. It combines an excellent selectivity and a good efficiency. Preliminary spectroscopic tests on the $n=50 \rightarrow n=51$ circular to circular transition at 51 GHz are reported.

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Rydberg atoms coupled to superconducting cavities have made possible spectacular achievements in the field of cavity quantum electrodynamics (CQED) [1,2]. However, their decay to low-lying states with a relatively short lifetime ($\simeq 150$ μs for $n=50$) places severe limitations on the atom-cavity interaction time. As pointed out recently [3], considerable improvements can be expected with the use of "circular" Rydberg states [4]. These levels, with maximum orbital and magnetic quantum numbers ($l=|m|=n-1$), decay only through a millimeter-wave transition to the nearest less-excited circular state. They have therefore a very long radiative lifetime in free space (20 ms for $n=50$). Combined with atomic-beam laser-cooling techniques, this makes possible long atom-cavity interaction times, in the hundredth of a second range. Moreover, a transition between two circular states can be well approximated by a two-level model, due to the weakness of the fine and hyperfine structures. Finally, these states experience a relatively small quadratic Stark effect in an electric field, as opposed to the huge linear Stark effect of ordinary Rydberg atoms. The circular to circular transitions are therefore quite insensitive to stray electric fields [the $n=50 \rightarrow n=51$ transition shift is only 260 kHz/(V/cm)^2]. Circular states appear thus as an excellent tool for CQED. $n=50$ circular states of rubidium will, for instance, be used in a quantum nondemolition photon-number measurement scheme developed in our laboratory [3].

Circular states, with their high angular momentum, cannot be reached directly from the ground state by laser excitation. Excitation of low $|m|$ states is followed by a "circularization" process. Two experimental methods have been developed so far. The "crossed-field" technique [5,6] makes use of slowly time-varying orthogonal electric and magnetic fields. Though very efficient, it may yield mixtures of high l states if the electric and magnetic fields are not very precisely controlled. The "adiabatic rapid passage" (ARP) technique [4] involves microwave transitions between Stark states whose degeneracy is removed by a slowly varying electric field. It is selective, but prepares both circular states ($m=\pm l$) and can be used only for moderate principal quantum numbers ($n \simeq 25$). Larger quantum numbers ($n \simeq 50$) are requested for CQED experiments. Moreover, the ARP method starts from a level connected by a small dipole matrix element to low-lying states, and produces a weak flux of circular atoms.

We present in this Brief Report a deeply modified ARP technique which can be used around $n=50$ to prepare circular state of well-defined helicity ($m=+l$ only). A Stark switching method [7] is used to reach the initial state of the ARP process, and boosts the overall efficiency.

We describe first the modified ARP method. Figure 1 shows a part of the rubidium $n=50$ manifold of Stark levels in a static electric field $\mathbf{E}_0$ ($|\mathbf{E}_0|=E_R=2.4$ V/cm). These levels are labeled by the magnetic quantum number m (quantization axis parallel to $\mathbf{E}_0$) and by the parabolic quantum number n_1 indicated under each level ($n_1=0,1,\ldots,n-|m|-1$). We consider here only levels with $m>0$. For each m, the lowest-energy level corresponds to $n_1=0$. For $m \geq 3$, the quantum defects are

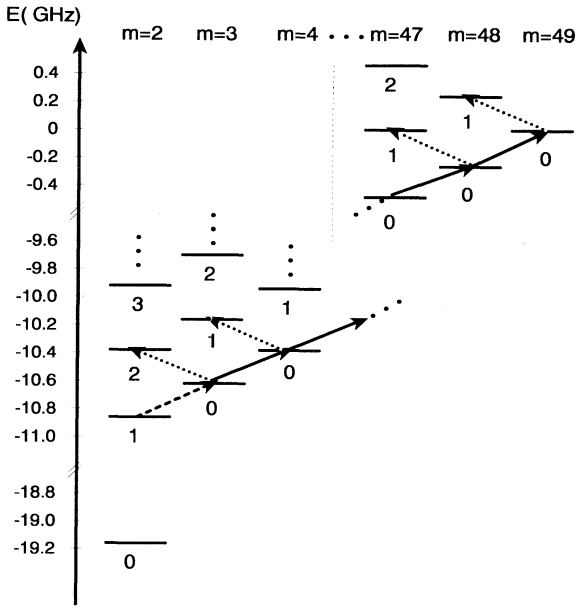

FIG. 1. Partial diagram of the $n=50$ Stark manifold of rubidium in a 2.4-V/cm electric field. 47 nearly degenerate σ^+ transitions at 233 MHz connect the $|m=2,n_1=1\rangle$ state to the circular state (dashed and solid lines). The parasitic σ^- transitions are depicted by dotted arrows. The σ^+ transitions are separated from the σ^- ones by adding a $\mathbf{B}_0=18$-G field along Oz (the σ^+ transitions frequency then increases to 258 MHz).

negligible and the levels structure is hydrogenic, with nearly equidistant energy levels. The circular state is $|m=49,n_1=0\rangle$, while the three-step laser excitation process prepares $m \leq 3$ states only. Let us recall that laser excitation is performed with three laser diodes tuned to the $5S \rightarrow 5P$, $5P \rightarrow 5D$, and $5D \rightarrow nl$ lines at 780 nm, 776 nm, and 1.26 μm, respectively [8].

A series of σ^+-polarized rf transitions (solid arrows in Fig. 1) connect the $|m=3,n_1=0\rangle$ state to the circular one. All these transitions take place in a 1-MHz range around 233 MHz, and can be excited simultaneously by the same source. The $m=2$ levels are nonhydrogenic. However, a remarkable coincidence makes the $|m=2,n_1=1\rangle \rightarrow |m=3,n_1=0\rangle$ transition frequency very close to 233 MHz also for $|\mathbf{E}_0|=E_R$ (dashed line in Fig. 1). For this field value, the transfer process starts from the $|m=2,n_1=1\rangle$ state. (For a different $|\mathbf{E}_0|$ value, the $|m=2,n_1=1\rangle \rightarrow |m=3,n_1=0\rangle$ transition is not degenerate with the next ones and the process should start from $|m=3,n_1=0\rangle$.)

From the $|m=2,n_1=1\rangle$ state to the circular one, 47 transitions take place. Only adiabatic rapid passage processes can yield a selective and efficient transfer. Let us start with an electric field modulus E_I slightly larger than E_R, and a σ^+-polarized rf field $\mathbf{E}_1$ containing $N+47$ photons ($N \gg 1$). $\mathbf{E}_1$ is resonant with the Stark transitions in a static field equal to E_R. Among the 47 relevant uncoupled atom-field levels ($|m=2,\ n_1=1,\ N+47\rangle$, $|m=3,\ n_1=0,\ N+46\rangle, \ldots, |m=49,\ n_1=0,\ N\rangle$, the first one has the lowest energy. All these levels cross for a static field equal to E_R. Because of the E_1-induced coupling, the dressed atom-field levels anticross instead of crossing. When the static field is slowly decreased from E_I to $E_F < E_R$, the system follows adiabatically the lowest "dressed" energy level. In the final static field, this level coincides with the $|m=49,n_1=0,N\rangle$ circular state. This ARP process is very efficient provided the variation of the Stark transitions frequency is much greater than Ω_1 (typical Rabi frequency in the rf field E_1), and its duration much greater than $1/\Omega_1$.

This simple picture holds only if $\mathbf{E}_1$ is perfectly σ^+ polarized, which cannot be achieved easily. For another polarization, σ^- transitions, which decrease m (depicted by dotted arrows in Fig. 1), become also resonant and the ARP process fails. Let us stress that, in previous experiments on moderate-quantum-number states ($n \simeq 20$) [4], these transitions were far from resonance—due to second-order Stark effect—and did not play any role. In order to avoid these stray transitions, we add a small (18 G) magnetic field $\mathbf{B}_0$ along the quantization axis. All the σ^+ transitions are shifted upwards by the same amount, and are the only ones to come into resonance when the electric field is decreased. The rf frequency is now increased to 258 MHz, so that it lies in between the σ^+ and σ^- transitions frequencies in the field E_I.

The initial state $|m=2,n_1=1\rangle$ could be reached directly by laser excitation. However, the matrix element between this state and the $5D$ state is quite small, and the resulting atom flux is then smaller by a factor of at least 10 than the one obtained with zero-field excitation of the $50F$ Rydberg level. In order to improve the excitation efficiency, we therefore use a Stark switching process. The atoms are prepared in zero electric field in the $|50F,m=+2\rangle$ substate, excited preferentially with σ, σ, and π polarizations for the three laser steps (the magnetic field $\mathbf{B}_0$ splits the excitation lines so that the $m=-2$ states are not excited). Then, the electric field $\mathbf{E}_0$ rises (in about 1 μs) to the value $E_I=2.6$ V/cm. The Stark level originating in $|50F,m=+2\rangle$ evolves into the $|m=2,n_1=1\rangle$ state without crossing any other state. If the field rise is slow enough, all the initial population is then adiabatically transferred to the starting level of the ARP process.

The experimental apparatus is sketched in Fig. 2(a). All the relevant parts are inside a 1.4-K enclosure (not shown) screening the blackbody microwave radiation from outside. The three excitation laser beams intersect the collimated rubidium atomic beam at right angles near the center of the circularization zone, where the fields are applied. The magnetic field $\mathbf{B}_0$ is produced by superconducting Helmholtz coils (section shown by black squares in the figure). As the other parts of the experiment require a good control of stray magnetic fields (at the mG level), the circularization zone is shielded by a superconducting niobium enclosure. The static electric field $\mathbf{E}_0$ is produced by two-plane copper plates, each made of three separate strips (plate separation and strip size are about 1 cm). The three strips in a plate are inductively coupled to the same dc bias source. A careful compensation of the copper-niobium contact potentials allows us to control the fields at the mV/cm level, and to obtain a good spatial homogeneity. The rf tunable generator is connected symmetrically between the first and last strips of each plate. It produces an $|\mathbf{E}_1|=5$ mV/cm field, roughly linearly polarized, parallel to the atomic beam.

The timing of the experiment is shown in Fig. 2(b). The whole cycle lasts 150 μs, and is repeated at 6.6 kHz. From 0 to 9 μs, the dc electric field is zero, and the rf power is off. A few $|50F,m=2\rangle$ atoms are prepared. Between 10 and 11 μs, the electric field rises to $E_I=2.6$ V/cm. From this time on, the lasers are no longer resonant with the Stark shifted levels, and no more atoms are produced. The rf power at 258 MHz is then switched on. As the initial detuning between the Stark transitions and the rf is half the Zeeman splitting in field $\mathbf{B}_0$ ($\simeq 50$ MHz), it is not much larger than the rf-induced Rabi frequency $\Omega_1/2\pi=5$ MHz. The rf amplitude has to be switched on adiabatically. It rises linearly between 13 and 16 μs, and decreases between 16.2 and 18 μs. At the same time, the static field decreases to $E_F=2.1$ V/cm, crossing the resonant value $E_R=2.4$ V/cm at $t=15$ μs.

At $t=18$ μs, the circularization process is completed. The field remains at 2.1 V/cm until 30 μs, and is then raised to 3.1 V/cm. This latter value is chosen to provide the weakest residual excitation by the lasers. Let us stress that the electric field cannot be canceled, since the circular states are not stable in vanishing fields [9]. Between 20 and 30 μs, a millimeter wave pulse (40–150 GHz) can be applied to probe the atomic state. Downstream, the atoms exit the circularization zone supercon-

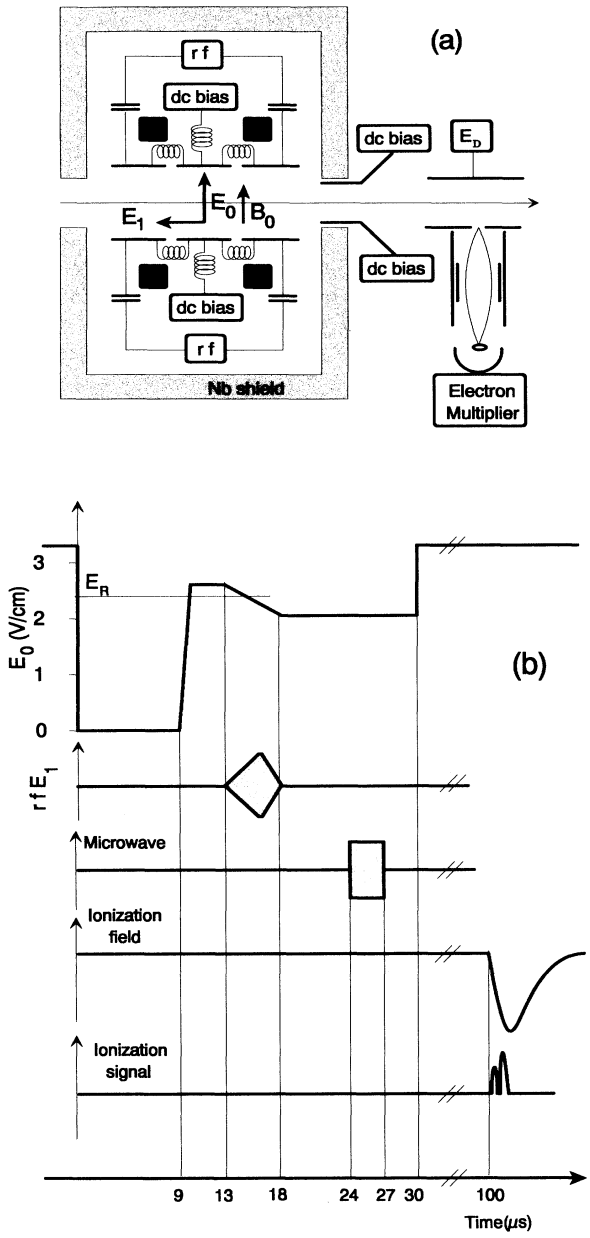

FIG. 2. (a) Sketch of the experimental setup. (b) The timing of the circularization sequence.

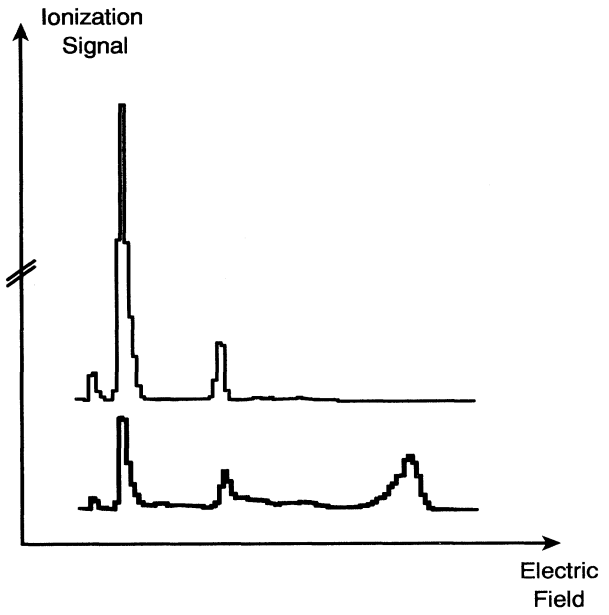

FIG. 3. Typical ionization signals vs field in the detection condenser. Upper trace: initial $|50F,m=2\rangle$ level. Lower trace: evidence of circular-state ionization near 148 V/cm.

ducting shield. Two small electrodes in the exit hole (diameter 4 mm, length 5 mm) provide a roughly homogeneous electric field to stabilize them. The atoms enter then inside a condenser, where an electric field ramp (E_D) is fired at $t=100$ μs (rise time 20 μs, maximum value 200 V/cm). The electrons resulting from Rydberg-state ionization are electrostatically focused on a particle counter. The ionization signal is recorded as a function of E_D and averaged.

Typical ionization signals are shown in Fig. 3. The upper trace corresponds to the $50F$ initial state. The double-peak structure is due to the complicated ionization process for this low m state. When the circularization sequence is performed, the lower trace is observed. The highest-field feature is attributed to the $n=50$ circular state. The corresponding ionization field value (148 V/cm) agrees quite well with theoretical predictions [10].

We have observed, with the probe microwave pulse, the one- and two-photon transitions to the $n=51$, 52, 49, and 48 circular states. All frequencies are in excellent agreement with the predicted values (taking into account the applied electric and magnetic fields). The Stark shifts of these transitions are found to be quadratic, with the expected slope. Finally, we have checked that no clear evidence of a σ^+ transition could be obtained on the $n=50\rightarrow n=49$ line. This is a good indication that the obtained level is a maximum magnetic number one. Taking into account the SNR of the detection, we infer that more than 95% of the high-field peak corresponds to a pure circular state.

The efficiency is quite good. The remaining low-field features in the lower trace of Fig. 3 correspond mainly to atoms initially in an $m=3$ state, parasitically populated due to imperfections in the laser polarizations and hyperfine coupling effects. More than 70% of the initial $m=2$ atoms have been promoted to the circular state. The resulting flux is about 30 000 atoms/s, and could be increased by optimization of the third laser step.

In a further step, we have propagated the atoms along the whole available experimental space (about 20-cm path). A set of electrodes provides on this path a carefully controlled electric field. A low-Q brass confocal microwave cavity has been added for spectroscopic tests. It

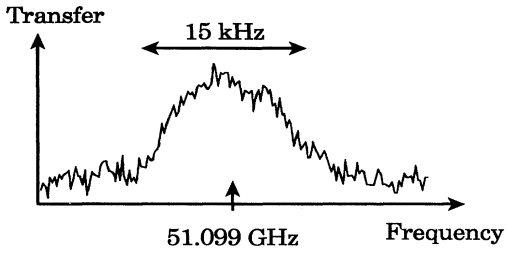

FIG. 4. $n=50$ to $n=51$ circular-to-circular transition induced by a 51.099-GHz microwave in a confocal cavity during a 20-cm flight between the circularization and the detection zone. The linewidth (12 kHz) is transit-time limited (waist of the cavity mode 6 mm).

sustains a 6-mm-waist mode close to the $n=50\rightarrow n=51$ transition frequency. The mode width ($\simeq 20$ MHz) is quite large, so that cavity length can be preset to the desired value without need of mechanical tuning. Downstream, two detection condensers produce inhomogeneous dc electric fields. The first ionizes the $n=51$ state, while the second is sensitive to the $n=50$ state. We have first checked that all the circular states survive the 600-μs flight across the apparatus. We have then performed the $n=50\rightarrow n=51$ transition in the cavity. A typical resonance is shown in Fig. 4. The observed width (12 kHz) is transit-time limited. Such a narrow line is a good indication of the feasibility of precise spectroscopic investigations on these levels [3].

In conclusion, we have described a circular-state-preparation technique for $n=50$ combining an excellent selectivity with a good efficiency. The circular-state-atom flux is comparable to the one obtained for ordinary Rydberg states. We have checked the purity of the circular state and performed encouraging spectroscopic investigations on a circular to circular transition. This method can be extended to other n values in rubidium, and adapted for other alkalis.

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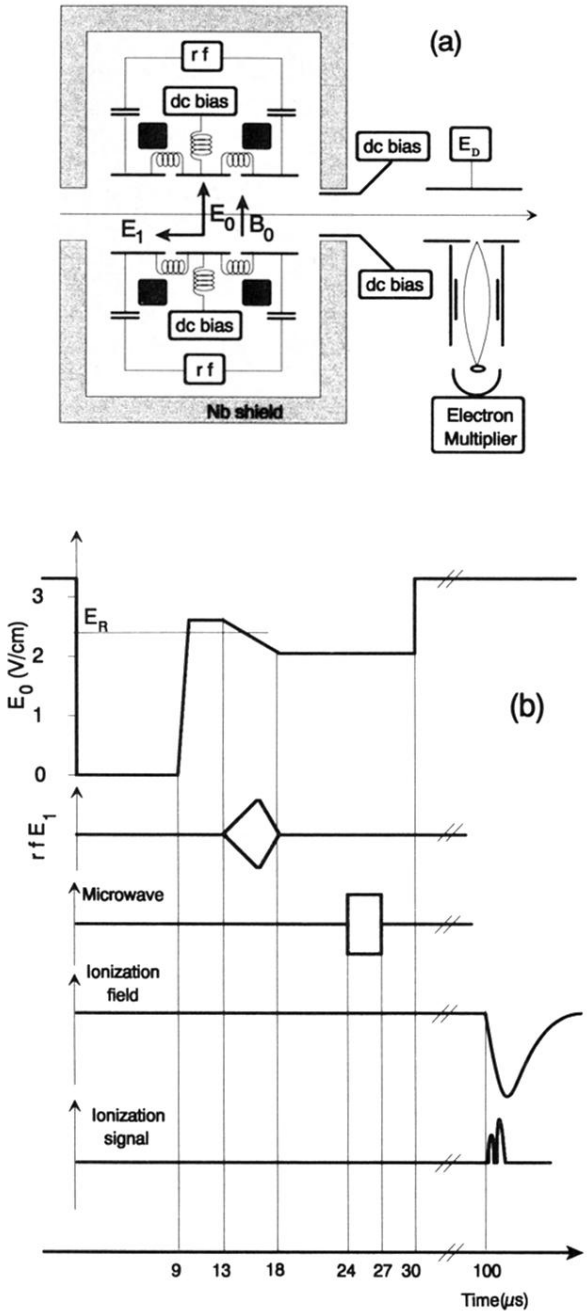

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