

## LETTER TO THE EDITOR

# Excitation of high angular momentum Rydberg states

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**Abstract.** We describe and analyse an RF-optical technique for exciting alkali atoms in an atomic beam to a specific high angular momentum Rydberg state. In the procedure the Rydberg states are dressed by an RF field and two optical photons are used to excite the dressed state that goes adiabatically into the desired Rydberg state as the RF field is turned off. We describe in detail the excitation of the aligned-circular-orbit states. These states have the maximum angular momentum quantum numbers for a given principal quantum number, and thus are the most nearly classical of all eigenstates. It is shown that in excess of 90% of the excited state population can be put into the aligned-circular-orbit state.

Rydberg atomic states have a number of striking properties, including nearly macroscopic dimensions, large polarisabilities, long radiative lifetimes and nearly classical behaviour (Haroche and Raimond 1985, Gallas *et al* 1985). Within this class of states exists a set with unique properties, the aligned-circular-orbit states. These states have maximum angular momentum ( $l$ ) and Zeeman ( $m$ ) quantum numbers for a given principal quantum number ( $n$ ). Their properties include the largest magnetic moments, the smallest sensitivity to static electric fields, the greatest collisional anisotropy and the longest radiative lifetimes of all states within a given  $n$  manifold. Several papers have suggested that these states are ideally suited to the study of various phenomena; for example, the modified spectra of a two-level atom in a cavity (Kleppner 1981) and the role of orientation in the collision process (de Prunele 1985).

There are two difficulties in efficient preparation of aligned-circular-orbit states. Firstly, the atom must be supplied with many units of angular momentum. It is not practical to supply approximately 20 units of angular momentum by means of an optical field, so it must be done with an RF or microwave field. Secondly, the high angular momentum Rydberg states are almost exactly degenerate so that it is difficult to populate selectively only one state. The production of high angular momentum Rydberg states was discussed by Richards (1984). Hulet and Kleppner (1983; see also Hulet *et al* 1985) have developed a method of exciting the high angular momentum Stark states of an atom using optical and microwave fields and a linearly ramped DC field. In particular, they have populated 'circular states' which are the Stark states with the parabolic quantum numbers  $n_1$  and  $n_2$  equal to zero and  $|m| = n - 1$ .

We propose here a method that excites the aligned-circular-orbit state selectively. The method uses RF and optical fields. The RF field interacts strongly with the states in the Rydberg manifold, preparing or 'dressing' the atom for the optical excitation. The resulting dressed states are linear combinations of many different angular momentum states, but each dressed state will go continuously into a particular angular

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momentum state as the RF field is turned off adiabatically. While the atom is dressed, the dressed state that is connected to the aligned-circular-orbit state is excited optically. The method allows either pulsed or cw optical excitation of the dressed state. An adiabatic turn-off of the RF field leaves the population in the aligned-circular-orbit state.

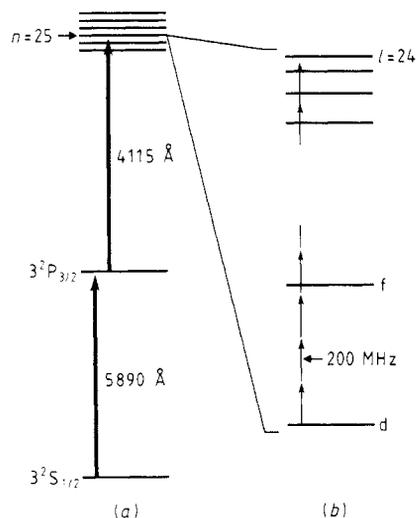
We first examine the dressed-state analysis which describes this method and then present the results of a numerical model of a system in which it is implemented.

The Hamiltonian for the atom-field system is

$$H = H_a + H_f + H_I \quad (1)$$

where  $H_a$  is the atomic Hamiltonian in which the atom's ionic core has been characterised by a dipole and a quadrupole polarisability,  $H_f$  is the field Hamiltonian and the interaction Hamiltonian,  $H_I$ , is  $-d \cdot E(t)$ .

A few assumptions can simplify the representation of this Hamiltonian and make it more applicable to our problem. Firstly, assume that the ground state is in an s state and that the optical excitation is by two-photon absorption of a circularly polarised field (figure 1(a)), which will populate a d state with  $m = 2$ . Also the RF field that supplies the necessary  $n - 3$  units of angular momentum (figure 1(b)) is circularly polarised. The frequency of the RF field is chosen near the  $n - 3$  multiphoton resonance between the d state and the aligned-circular-orbit state so that the mixing of these states is strong. These assumptions reduce the number of states that need to be included in our analysis. The s and p Rydberg states may be ignored since their energies are quite different from the other states of the  $n$  manifold. This energy difference is sufficiently large to keep the RF field from mixing these states with the rest of the manifold. The circular polarisation of the fields eliminates the need to consider states that would be excited by  $\Delta m = 0$  transitions. The transitions between aligned states are favoured over those from aligned to unaligned states. The squares of the dipole



**Figure 1.** (a) An energy level diagram for sodium is depicted indicating the optical excitation of the  $n = 25$  Rydberg manifold. The excitation is accomplished by a resonantly enhanced two-photon absorption using the  $3^2P_{3/2}$  as the intermediate state. (b) A blow-up of the  $n = 25$  manifold is shown. The circularly polarised RF field is tuned near the  $n - 3$  multiphoton resonance between the d state and the aligned-circular-orbit state.

moments for the aligned-to-aligned transitions are larger by a factor of  $(2l+1)(l+1)$  (Bethe and Salpeter 1957). The s and p states have been dropped, so this factor is greater than or equal to 15 ( $l \geq 2$ ). Since the optical excitation has been designed to produce an aligned state ( $l = m = 2$ ), only the aligned states of the Rydberg manifold will be included in the analysis. It should be noted that all the magnetic moments remain aligned in transitions between these states and the fine and hyperfine structure is therefore eliminated for these transitions.

With these assumptions, the Hamiltonian can be written in terms of a reduced basis set. The states of this basis are denoted  $|n, l, m = l\rangle \equiv |l\rangle$ . The dressed-state Hamiltonian is obtained by a transformation to a rotating frame. This transformation is accomplished by the unitary operator

$$U(t) = \exp\left(-i\omega t \sum_{l=2}^{n-1} |l\rangle\langle l|\right). \quad (2)$$

The Hamiltonian in the rotating frame is

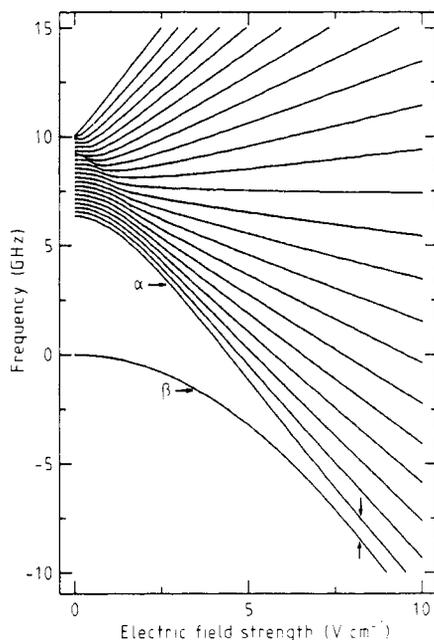
$$H' = \sum_{l=2}^{n-1} \hbar[(l-2)\omega - \omega_{l,2}]|l\rangle\langle l| - E_0 \sum_{l=3}^{n-1} d_{l,l-1}(|l\rangle\langle l-1| + |l-1\rangle\langle l|) \quad (3)$$

where  $\omega$  is the frequency of the RF field. The transition frequency from the d state to the state  $l$  is  $\omega_{l,2}$ . The amplitude of the RF field is  $E_0$  and the dipole moment between aligned states is  $d_{l,l-1}$ .

The eigenvectors of  $H'$  are the dressed states of the system. The eigenvalues of the manifold of dressed states, as a function of the electric field strength, are found by diagonalising this Hamiltonian at a fixed frequency and allowing the field strength to vary (figure 2). Note that the dressed states linked to the zero-field d state and the aligned-circular-orbit state undergo a weakly avoided crossing, at which the projection of the latter on the zero-field d state is at its maximum.

The RF field strength is chosen so that the avoided crossing is reached. The frequency of the RF field is determined by three factors. It must be near the  $n-3$  multiphoton resonance so that the dressed state connected to the aligned-circular-orbit state has a large projection on the zero-field d state. The frequency must, however, be far enough from resonance that the separation of the dressed states is sufficient to allow the optical excitation of only the desired dressed state. The frequency is also constrained by the need for an adiabatic turn-off of the RF field. The adiabatic turn-off time depends inversely on the energy separation of the desired dressed state from its neighbours. The smallest separation occurs at zero RF field. In the limit, as the RF field strength approaches zero, the dressed states are separated by the energy of an RF photon. An RF field with a suitable frequency may thus be turned off adiabatically in a time short compared with the decay time of the dressed state.

We will now consider the excitation of sodium atoms in an atomic beam to the  $n = 25$  manifold. The beam of sodium atoms enters the interaction region, where it is dressed by the circularly polarised RF field. The latter has a frequency of 200 MHz and a maximum field strength of  $8 \text{ V cm}^{-1}$ . The desired dressed state is excited by a resonant two-photon process. Population is transferred from the ground state ( $3^2\text{S}_{1/2}$ ) to the  $3^2\text{P}_{3/2}$  state by a laser tuned to resonance ( $5890 \text{ \AA}$ ). A second laser ( $4115 \text{ \AA}$ ) excites the desired dressed state. The population of this dressed state follows adiabatically into the aligned-circular-orbit state after the turning-off of the RF field. The adiabatic turn-off is accomplished by the departure of the atoms from the interaction



**Figure 2.** The eigenvalues of the dressed Rydberg states are shown as a function of the RF field strength. The frequency of the RF field is 200 MHz.  $\alpha$  is the dressed state linked to the aligned-circular-orbit state.  $\beta$  is the dressed state linked to the d state. At the weakly avoided crossing,  $\alpha$  and  $\beta$  are separated by 1 GHz.

region. A typical length for such an interaction region is 2–3 mm. Since the velocity of the atoms is of the order of  $1000 \text{ m s}^{-1}$ , the atoms leave the interaction region in approximately  $3 \mu\text{s}$ . This time is short compared with the  $15 \mu\text{s}$  lifetime of the dressed state. The following analysis demonstrates that the departure time is long compared with the adiabatic turn-off time.

A model of this system is easily analysed numerically to find the population excited to the sodium  $n = 25$  aligned-circular-orbit state. The necessary sodium dipole moments are found by integrating the radial integral numerically using a Numerov algorithm (Froese 1963, Blatt 1967).

The dressed-state Hamiltonian is diagonalised numerically to find the eigenvector associated with the desired dressed state at the avoided crossing. Using this eigenvector as the initial state, we integrate the time-dependent Schrödinger wave equation. This integration gives the distribution of the population following the atom's departure from the interaction region. Approximately 99% of the population in the dressed state reaches the aligned-circular-orbit state.

Calculations similar to those described above can be performed for various degrees of circular polarisation of the RF field. The basis set is expanded to include states that are excited by  $\Delta m = 0$  transitions. As the polarisation becomes more elliptical, two processes are affected: (i) the selective excitation of the desired dressed state and (ii) the adiabatic turn-off of the RF field. Table 1 summarises the effects of changing the RF polarisation. For example, consider an RF field which is elliptically polarised, with 80% of the intensity as right circular polarisation and 20% as left circular polarisation. The nearest state with a comparable projection of the d state is then only 600 MHz away. This state previously had no projection on the d state since  $\Delta m = 0$  transitions

are not allowed in a pure circularly polarised field. The result is that an elliptically polarised RF field tightens the constraints on the bandwidth of the lasers. The adiabatic turn-off of such an RF field leaves 94% of the excited-state population in the aligned-circular-orbit state. The next most populous state (5%) is the  $|n = 25, l = 24, m = 23\rangle$  state. Thus the requirements for an adiabatic turn-off are relatively insensitive to the polarisation of the RF field.

**Table 1.** Summary of the effects of changing the polarisation of the dressing field.  $\nu_1$  and  $\nu_2$  are the separations (in MHz), at the avoided crossing, of the desired dressed state from the nearest and next-nearest dressed states respectively.  $p_1$  and  $p_2$  are the projections of these states onto the d state.  $p_{dd}$  is the projection of the desired dressed state onto the d state.  $P_{ACO}$  and  $P_N$  are the percentage populations of the aligned-circular-orbit state and the next most populous state, after the adiabatic turn-off of the RF field.

|           | Right circular polarisation of the RF field (%) |       |       |       |       |
|-----------|---|-------|-------|-------|-------|
|           | 100   | 90    | 80    | 70    | 60    |
| $\nu_1$   | 456   | 585   | 596   | 614   | 578   |
| $\nu_2$   | 1512  | 1035  | 875   | 796   | 720   |
| $p_1$     | 0.0   | 0.039 | 0.072 | 0.067 | 0.002 |
| $p_2$     | $9 \times 10^{-15}$                             | 0.257 | 0.218 | 0.168 | 0.125 |
| $p_{dd}$  | 0.332   | 0.331 | 0.309 | 0.223 | 0.107 |
| $P_{ACO}$ | 99.24   | 97.25 | 94.32 | 87.95 | 80.95 |
| $P_N$     | 0.75  | 2.23  | 5.15  | 11.36 | 18.43 |

We have described a method for exciting aligned-circular-orbit states based on the dressing of the atom by an RF field. A possible experimental system was discussed and shown to be feasible. The system allows pulsed or cw excitation of the aligned-circular-orbit states of sodium atoms in a thermal beam. The numerical analysis of this system has shown that the method is efficient, being capable of leaving 99% of the Rydberg population in the aligned-circular-orbit state. These results were obtained for a circularly polarised RF field. The main effect of an elliptically rather than circularly polarised RF field is the stiffening of the laser bandwidth requirements. The percentage of the population reaching the aligned-circular-orbit state remains high, in excess of 90% for reasonable experimental conditions.

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