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# A New Method for Producing Circular Rydberg States (*).

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**Abstract.** – We propose a new theoretical scheme for exciting atoms in circular Rydberg states with high efficiency whatever the atomic species. It only requires the use of a weak magnetic field crossed to a weak time-varying electric field. The scheme is based on the symmetry properties of the Coulomb interaction.

Rydberg atoms with extremum values of the magnetic quantum number $|m| = n - 1$ ($n$ the principal quantum number) are called circular states. Electric density plots are peaked along a circle which recalls the familiar Bohr orbit's picture. A feature specific to these states is their very long radiative lifetime which makes them of interest for high-precision spectroscopy and metrology [1, 2]. They have allowed to experimentally demonstrate the inhibition of spontaneous emission in a cavity [3], and have numerous potential applications to basic physics.

However, producing atoms in circular Rydberg states is not straightforward. The techniques previously proposed [1, 3, 4] rely on the conservation of the angular momentum in the interaction of atoms with microwave fields. For instance, in the Hulet-Kleppner scheme [1] presently used in several experiments [1-3], the atoms are laser excited into low-$m$ Rydberg states. Next, the absorption of several microwave photons through a series of adiabatic rapid passages in the Stark energy diagram allows to produce circular states. A time-varying electric field is required in order to match the resonance condition for absorption from the fixed-frequency microwave source. This method works for light quasi-hydrogenic species (*e.g.*, Li [1]), while for heavier species (*e.g.*, Cs [3]), it requires the use of several microwave fields.

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The method we propose here is based on completely different considerations. It fully exploits the $SO(4)$ symmetry properties of the Rydberg shell with principal quantum number $n$ and its breaking in weak crossed electric and magnetic fields.

At the beginning of the process, the hydrogen atom is submitted to weak crossed fields such that the Stark interaction is dominant over the Zeeman one. Efficient laser excitation of the $m=0$ Stark sublevel with maximum parabolic quantum number can be performed. Next, the electric field is adiabatically switched off to zero. Due to the pecularities of the crossed-field energy diagram, the final atomic state is a circular state ($L_z = m = n-1$) at the end of the process.

The key point in the method is the symmetry of the hydrogen atom. A nonrelativistic version, ignoring the role of the spin, is sufficient for dealing with most situations. The Coulomb-Hamiltonian is just (atomic units):

$$H_0 = p^2/2 - \frac{1}{r}. \tag{1}$$

The angular momentum $\boldsymbol{L} = \boldsymbol{r} \times \boldsymbol{p}$ in real space and the Runge-Lenz vector ($E$ the energy)

$$\boldsymbol{A} = \frac{1}{(-2E)^{1/2}} \left\{ \frac{\boldsymbol{p} \times \boldsymbol{L} - \boldsymbol{L} \times \boldsymbol{p}}{2} - \frac{\boldsymbol{r}}{r} \right\} \tag{2}$$

are constants of the motion.

The classical meaning is well-known. The trajectory of the electron is a Kepler ellipse (with focus at the proton). $\boldsymbol{L}$ is the classical angular momentum and $\boldsymbol{A}$ a constant vector along the major axis of the ellipse, proportional to its eccentricity (see fig. 1).

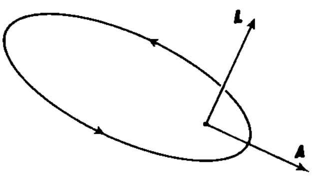


Fig. 1. – Classical trajectory (Kepler ellipse) of the electron in zero field. The angular momentum $\boldsymbol{L}$ and the Runge-Lenz vector $\boldsymbol{A}$ are constants of the motion. $\boldsymbol{L}$ is perpendicular to the plane of the trajectory. $\boldsymbol{A}$ is directed along the major axis of the ellipse and proportional to its eccentricity.

The commutation relations between the $\boldsymbol{L}$ and $\boldsymbol{A}$ operators are [5, 6]:

$$\begin{cases} [A_i, A_j] = i\,\varepsilon_{ijk} \cdot L_k\,, \\ [A_i, L_j] = i\,\varepsilon_{ijk} \cdot A_k\,, \\ [L_i, L_j] = i\,\varepsilon_{ijk} \cdot L_k\,, \end{cases} \tag{3}$$

where $\varepsilon_{ijk}$ is the fully antisymmetric tensor ($i, j, k = 1, 2, 3$). Adding a fourth dimension to space, labelled with the subscript 4, and considering the new operator $\mathscr{L}$ such that

$$\mathscr{L}_{i4} = A_i, \qquad \mathscr{L}_{ij} = \varepsilon_{ijk} \cdot L_k \tag{4}$$

the commutation relations (3) turn out to be those of an angular momentum $\mathscr{L}$ in the four-dimensional space. The symmetry group of the Coulomb problem [5, 6] is $SO(4)$ with the generators $\mathscr{L}_{ij}$.

Consequently, there are several possible choices of eigenbasis. Some are well known as the parabolic ones associated with eigenfunctions of $L_z$ and $A_z$ and the subgroup chain $SO(4) \supset SO(2)_{n_1} \otimes SO(2)_{n_2}$, or the spherical one associated with eigenfunctions of $L^2$ and $L_z$ and the $SO(4) \supset SO(3)_1 \supset SO(2)_m$ subgroup chain. The latter preserves the rotational invariance in the (1, 2, 3) subspace that is «real space», with

$$L^2 = \mathscr{L}_{12}^2 + \mathscr{L}_{23}^2 + \mathscr{L}_{31}^2 .$$

Another choice is important in view of the present applications. From its commutation relations, the operator $\boldsymbol{\lambda} = (\mathscr{L}_{14}, \mathscr{L}_{24}, \mathscr{L}_{12}) = (A_x, A_y, L_z)$ is a 3-dimensional angular momentum. The eigenfunctions of $\boldsymbol{\lambda}^2$ and $\lambda_z$ associated with the subgroup chain $SO(4) \supset$ $\supset SO(3)_\lambda \supset SO(2)_m$ are solutions to the Coulomb problem which have already proven useful for dealing with atomic diamagnetism [7, 8].

In atomic units, the Hamiltonian in crossed electric and magnetic fields can be written down as [9, 10]

$$H = \frac{p^2}{2} - \frac{1}{r} + \frac{\gamma}{2} L_z + F \cdot x + \frac{\gamma^2}{8}(x^2 + y^2), \tag{5}$$

where the electric field (respectively, magnetic field) is parallel to the $x$-axis (respectively, $z$-axis). $\gamma$ and $F$ are the strengths of the fields expressed in atomic units (respectively, $2.35 \cdot 10^5$ T and $5.14 \cdot 10^{11}$ V/m). The only constant of the motion is parity along the $z$-axis.

However, in the weak-field limit, neglecting inter-$n$ mixing and the diamagnetic interaction, the solution is well known [5, 9, 10]. It is obtained by diagonalizing the field perturbation $W = (\gamma/2) \cdot L_z + F \cdot x$ in a given $n$-shell. The operator $x$ in a given $n$-shell is proportional to the component $A_x$ of the Runge-Lenz operator $x = -(3/2) n \cdot A_x$ (this is known as the «Pauli replacement»). Hence, the expression of $W$ in the $n$-shell is

$$(W)_n = \frac{\gamma}{2} L_z - \frac{3}{2} n F A_x = \omega_L L_z + \omega_S A_x , \tag{6}$$

where $\omega_L = \gamma/2$ and $\omega_S = -(3/2) nF$ are the Larmor and Stark frequencies in atomic units ($\boldsymbol{\omega}_L = -(q\boldsymbol{B}/2m)$ and $\boldsymbol{\omega}_S = (3/2)(4\pi\varepsilon_0 \hbar/mq) n\boldsymbol{F}$ in MKSA units). Introducing the unit vector $\boldsymbol{u}(\sin\alpha, 0, \cos\alpha)$ with $\alpha = \mathrm{tg}^{-1}(\omega_S/\omega_L)$, one obtains

$$W = \sqrt{\omega_L^2 + \omega_S^2}\, \boldsymbol{\lambda} \cdot \boldsymbol{u} . \tag{7}$$

The eigenfunctions of $H$ can then be chosen as eigenfunctions of $\boldsymbol{\lambda}^2$ (eigenvalue $\lambda(\lambda+1)$) and $\lambda_u$ (eigenvalue $k$). The spectrum is (1)

$$E = -\frac{1}{2n^2} + k\sqrt{\omega_L^2 + \omega_S^2} , \tag{8}$$

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(1) Another description is possible introducing the 3-dimensional angular-momentum operators: $\boldsymbol{j}_{1/2} = \frac{1}{2}(\boldsymbol{L} \pm \boldsymbol{A})$, associated with the decomposition $SO(4) = SO(3) \otimes SO(3)$. This allows to describe the properties in term of a vectorial model involving two vectors $\boldsymbol{j}_1$ and $\boldsymbol{j}_2$ [6, 10].

$k$ being an integer ranging from $-(n-1)$ to $(n-1)$. For a given $k$, values of $\lambda$ are such that $|k| \leq \lambda \leq n-1$, and the degeneracy is $n-|k|$ [7, 8].

Zeeman and linear Stark effects are both contained in (8) as limiting cases, exhibiting similar equally spaced energy level structure (see fig. 2). The eigenfunctions of $(\lambda^2, \lambda_u)$ type continuously evolve from one limit to the other one.

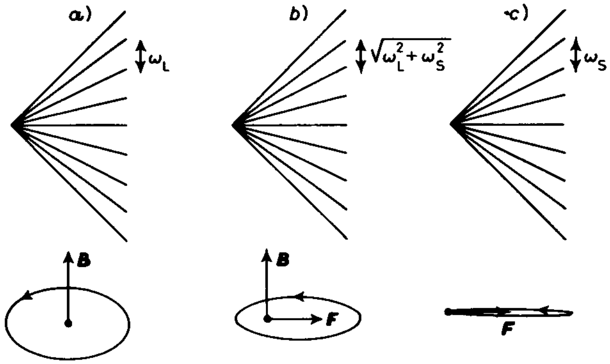


Fig. 2. – Structure of an hydrogenic manifold in the low fields limit and classical trajectories associated with the upper state. *a*) Magnetic field only (along $z$-axis). The upper state is a circular state (circular trajectory). *b*) Crossed fields. The trajectory is an ellipse in the $(x,y)$-plane with major axis along $x$. *c*) Electric field only (along $x$-axis). The upper state is a parabolic state (trajectory is a straight line).

The (nondegenerate) upper state of the manifold with $k = \lambda_u = n-1$ and $\lambda = n-1$, is of special interest. In the Zeeman limit, it coincides with the circular state as $k = n-1 = \lambda_z = L_z$. A classical picture associated to this state is the Bohr circular orbit in the $(x, y)$-plane (see fig. 2). In the Stark limit, the upper state of the manifold coincides with the parabolic state $|n_1 = 0, n_2 = n-1, m = 0\rangle$ as $k = n-1 = \lambda_x = A_x = n_2 - n_1$ (quantization along $x$-axis). Hence $A_x$ takes its maximum value $(n-1)$ and the associated classical trajectory is an ellipse with maximum eccentricity $(=1)$—that is a straight line along the $x$-axis (see fig. 2). Having a low angular momentum, such a state can be efficiently laser excited.

In intermediate (crossed-fields) conditions, the angular momentum $\boldsymbol{\lambda}(A_x, A_y, L_z)$ is along the $\boldsymbol{u}(\sin\alpha, 0, \cos\alpha)$ direction taking its maximum values $\lambda = \lambda_u = n-1$. The associated classical trajectory is such that

$$\begin{cases} L_z = \lambda_z = (n-1)\cos\alpha\,, \\ A_x = \lambda_x, = (n-1)\sin\alpha\,, \end{cases} \tag{9}$$

and it is an ellipse in the $(x, y)$-plane with eccentricity $\sin\alpha$ (see fig. 1 and 2).

The scheme for producing atoms in circular Rydberg states is as follows. Starting with constant electric and magnetic fields such that $\omega_S \gg \omega_L$, the upper state of the manifold $(\lambda = \lambda_x = n-1)$ can be optically excited from low-lying states as it is associated with $L_x = m = 0$ along the electric-field axis. Next the electric field is switched off to zero. Provided the switching is slow enough, the evolution is adiabatic and the atoms are in the circular state $\lambda = \lambda_z = L_z = n-1$ (along $\boldsymbol{B}$ field) at the end of the process.

Obviously, the scheme does not involve a series of adiabatic rapid passages as the energy levels are *always equally spaced*. At any stage of the evolution from the Stark to the Zeeman limits, the electron is in a well-defined $|\lambda = n-1, \lambda_u = n-1\rangle$ state. This is actually a coherent state of the $SO(3)_\lambda$ subgroup, which means that the classical pictures previously discussed are valid. The decomposition of such a state on the usual $(n l m)$ spherical basis is quite complicated. But the whole process is nothing but a $\pi/2$ «adiabatic» rotation of the angular momentum $\boldsymbol{\lambda}$ in the $(x, z)$-plane. The generator is $\lambda_y = A_y$ and, consequently, it is *not* a rotation in real space. It transforms the parabolic state into the circular state with completely different electronic densities (and classical representations).

The condition of adiabaticity is

$$\frac{d\alpha}{dt} \ll (\omega_L^2 + \omega_S^2)^{1/2} \qquad \text{or} \qquad \frac{d\omega_S}{dt} \ll \frac{(\omega_L^2 + \omega_S^2)^{3/2}}{\omega_L}, \tag{10}$$

which is easily fulfilled in most situations. For example, typical orders of magnitude for $n = 25$ are $B = 10$ G and $E = 3$ V/cm. This means $\omega_L = 14$ MHz and $\omega_S \simeq 10\omega_L \gg \omega_L$ at the beginning of the process. The electric field can be switched off to 0 within 5 μs which complies with inequality (8). Moreover 5 μs is smaller than any radiative lifetime in the $n = 25$ manifold. Hence 100% efficiency in the excitation of circular states can be achieved. Geometrical imperfections of the set-up (*e.g.* polarization of the laser or the crossed character of $\boldsymbol{E}$ and $\boldsymbol{B}$) and fine or hyperfine structure have no practical consequences. What matters is that the upper state of the manifold be isolated from the other ones and be efficiently excited in the Stark limit.

With minor amendments, this scheme based on the $SO(4)$ symmetry applies to nonhydrogenic species, *e.g.* alkali Rydberg atoms. The $n^2$ degeneracy of the $n$-manifold is broken by core corrections (quantum defects). However, most of the states have a negligible quantum defect ($l \geqslant 1$ for lithium and $l \geqslant 3$ for caesium) and build a quasi-hydrogenic manifold. The «nonhydrogenic states» (low-$l$ values) are isolated from the incomplete manifold. In the Zeeman limit, the circular state ($L_z = n-1$) is the upper state of the manifold and *unaffected by core corrections*. In the Stark limit, the upper state of the quasi-hydrogenic manifold is a $L_x = 0$ state, but not of the type $A_x = n-1$ as the low-$l$ components are lacking. The previous method using adiabatic passing from the Stark limit to the Zeeman limit then allows efficient excitation of circular states, provided the upper $L_x = 0$ state can be optically excited in a selective way.

Quantum defects in lithium are $\delta(S) = 0.40$, $\delta(P) = 0.05$ and $\delta(D) \simeq 0.002$. Selective optical excitation of the $n = 25$ quasi-hydrogenic manifold has been realized by Liang [2] in a field of 180 V/cm. With such a field, the $D$ state is mixed with the quasi-hydrogenic manifold, and the upper $L_x = 0$ one can be efficiently optically populated using stepwise excitation [2]. Even lower electric field could be used. With the laser linewidth of ref. [2] (2 GHz) and $n = 25$, the minimum field value to *selectively* excite the state is around 90 V/cm. With a line width 10 times smaller, $E = 3$ V/cm and $B = 10$ G would be sufficient to ensure 100% efficiency in the production of circular states.

In their study of inhibited spontaneous emission on caesium circular states, Hulet *et al.* [3] made use of two different microwave fields. Again, a crossed-field scheme seems to provide us with a simpler way of achieving the production of circular states. As shown on fig. 3, the upper state of the incomplete manifold at low fields is a $L_x = 0$ state, which cannot be optically excited through two-step excitation, being a mixture of states with $l \geqslant 3$. However, at higher fields, it interacts with the $(n+3)\,D$ state and can be efficiently excited. We checked this on the $n = 22$ manifold of caesium. The energy levels are plotted on fig. 3 (calculations incorporate the $n = 22$ manifold and the nearest nonhydrogenic states, fine

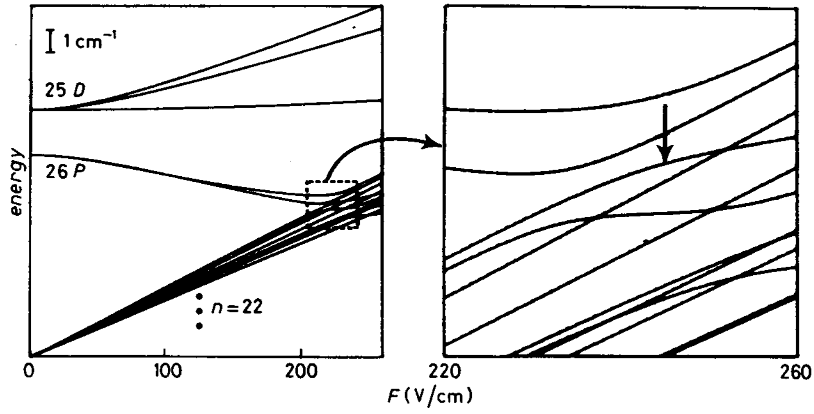


Fig. 3. – Energy diagram of caesium as a function of electric field. Upper states of the $n = 22$ manifold and nonhydrogenic $25D$ and $26P$ states are represented. In the vicinity of 245 V/cm, because of the strong interaction with the $26P$ and $25D$ states, the upper state of the manifold (indicated with an arrow) carries 15% of the ($25D$, $M = 0$) state and can be efficiently excited.

structure is neglected). For $B = 10$ G and $E = 245$ V/cm, the upper state of the manifold (indicated with an arrow) is well isolated from the others (distance greater than 3 GHz) and carries 15% of the $23D$ oscillator strength. Consequently, efficient stepwise excitation can be achieved. After switching off the electric field, $n = 22$ circular states will be produced.

In summary, the method we propose for producing Rydberg atoms in circular states has several advantages. Conditions for adiabaticity are less stringent than in the microwave-type experiments. A weak magnetic field is needed ($\simeq 10$ G) instead of one or several microwave sources. The same set-up can be used for several $n$ values and various atomic species, by only changing the field strengths. Hence this is far more flexible. Finally, the method fully exploits the symmetry property of the $n^2$ degenerated Rydberg shell and illustrates in a special case how to control, with electric and magnetic fields, the structure of quasi-hydrogenic atomic species.

## REFERENCES

[1] HULET R. G. and KLEPPNER D., *Phys. Rev. Lett.*, **51** (1983) 1430.
[2] LIANG J., Thèse de Doctorat d'Université, Paris (1986); GROSS M. and LIANG J., *Phys. Rev. Lett.*, **57** (1986) 3160.
[3] HULET R. G., HILPER E. S. and KLEPPNER D., *Phys. Rev. Lett.*, **55** (1985) 2137.
[4] MOLANDER W. A., STROUD jr. C. R. and YEAZELL J. A., *J. Phys B*, **19** (1986) L-461.
[5] PAULI W., *Z. Phys.*, **36** (1926) 339.
[6] GAY J. C. and DELANDE D., in *Atomic Excitation and Recombination in External Fields*, edited by M. H. NAYFEH and C. W. CLARK (Gordon and Breach, New York, N.Y.) 1985.
[7] HERRICK D. R., *Phys. Rev. A*, **26** (1982) 323.
[8] LABARTHE J. J., *J. Phys. B*, **14** (1981) L-467.
[9] DEMKOV Y. N., MONOZON B. S. and OSTROVSKII V. N., *JETP Lett.*, **30** (1970) 775.
[10] PENENT F., Thèse 3e Cycle (Paris, 1984); PENENT F., DELANDE D., BIRABEN F. and GAY J. C., *Opt. Commun.*, **49** (1984) 184.