Circular Atoms Prepared by a New Method of Crossed Electric and Magnetic Fields

J. Hare, M. Gross, and P. Goy
Département de Physique de l'Ecole Normale Supérieure, Laboratoire de Spectroscopie Hertzienne, 24 rue Lhomond, 75231 Paris CEDEX 05, France
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Circular atoms, i.e., Rydberg atoms with maximum angular momentum, are semiclassical atoms of great interest for Rydberg constant metrology and QED tests. Following a suggestion by Delande and Gay, we have prepared circular atoms by a new method, involving the adiabatic switching of crossed electric and magnetic fields. The method, which is demonstrated for Rydberg states of lithium (principal quantum number $n=20$ to 25), is applicable over a wide variety of experiment conditions.

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Circular atoms are Rydberg atoms whose angular momentum is maximum ($l=|m|=n-1$, where n, l, and m are the principal, orbital, and magnetic quantum numbers, respectively). Such atoms are ideal semiclassical objects at the frontier of classical and quantum physics: The excited electron distribution has the shape of a thin torus along the Bohr circular orbit of radius n^2a_0 (a_0 is the Bohr radius).

As the electron distribution does not penetrate the atomic core, any atom in a circular state is a quasihydrogenic system. Moreover, circular atoms radiatively decay only through $\Delta n=-1$, $\Delta m=-1$ circularly polarized radio-frequency transitions and have thus very long lifetimes (2 ms for $n=30$). These features allow QED basic tests such as the inhibition of spontaneous emission in a cavity,[1] and make the millimeter microwave spectroscopy of the circular-circular transitions highly interesting for a frequency measurement of the Rydberg constant.[2,3]

Circular atoms were prepared first in 1983 by Hulet and Kleppner[4] using an adiabatic microwave transfer method (AMTM). Delande and Gay,[5] and independently Liang[6] have proposed preparing them by a scheme involving crossed electric (E) and magnetic (B) fields, these fields being adiabatically switched off and on, respectively. The crossed-fields method (CFM) requires few adjustments, has no resonance condition, and works in principle for any atomic species. This Letter presents the first CFM experimental evidence.

CFM is related to the specific symmetry of the Coulomb interaction, making the levels of a given n principal-quantum-number manifold nearly degenerate. External fields, electric or magnetic, remove the degeneracy and provide a "natural" eigenstate basis. In a low static **E** field, the eigenstates are parabolic states ($n,m,n1$ quantum numbers), in a low static **B** field both parabolic and spherical (n,l,m) bases may be used. Without external fields, spurious **E** fields varying along the atom path have a dominant effect and make the atomic state evolve randomly inside the n manifold: Any state becomes experimentally unstable.[7]

The CFM process is simple: Atoms are prepared in an electric **E** field in the state whose electric dipole is maximum. A crossed magnetic **B** field is then adiabatically switched on, while the **E** field is switched off. Let us consider the n-manifold energy-versus-time diagram during this process [see Figs. 1(a)–1(c)]. Figure 1(a) corresponds to the initial situation with $\mathbf{E}\neq 0$ and $\mathbf{B}=0$. Figure 1(b) corresponds to the intermediate one, where the **E** field is continuously decreased as the **B** field is increased. Figure 1(c) corresponds to the final one with $\mathbf{E}=0$ and $\mathbf{B}\neq 0$. In orthogonal fields, the symmetry makes the n manifold partly degenerate with $2n-1$

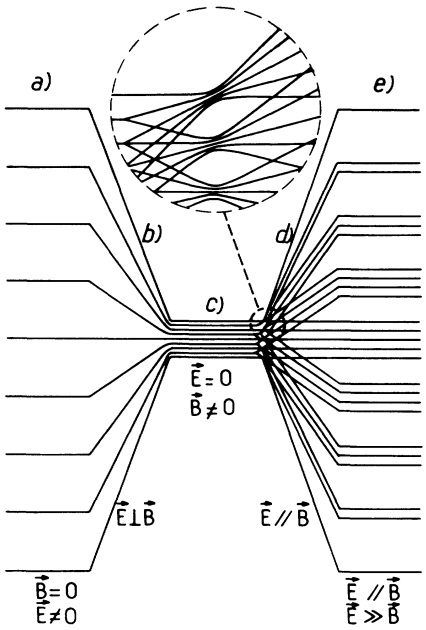

FIG. 1. Energy vs time diagram of the n manifold: (a) at the very beginning of the experiment, with **E** field alone; (b) during the switching process (orthogonal **E** and **B** fields are switched off and on, respectively); (c) at the end of the switching process, with **B** field alone; (d) with nearly parallel fields, **E** increasing from zero with constant **B**; (e) with nearly parallel fields, with **E** dominant.

equally spaced levels. The state with the largest energy is nondegenerate. It coincides for $\mathbf{B}=0$ with the $m=0$, $n1=n-1$ parabolic state, whose electric dipole, oriented along the $\mathbf{E}$ field, has the largest amplitude. For $\mathbf{E}=0$, it coincides with the circular state whose magnetic dipole, oriented along the $\mathbf{B}$ field, has the largest amplitude. For an adiabatic switching, the atomic system remains, at any time, in the state of the diagram, which is related to the initial one by continuity. By this way, the atoms initially prepared in the $m=0,n1=n-1$ state become circular. The condition of adiabaticity is[5]

$$dE/dt \ll B^2/(6n) \quad (1)$$

(where $\mathbf{E}$ and $\mathbf{B}$ are expressed in atomic units). For $n=25$ and $B=100$ G, Eq. (1) yields $dE/dt \ll 400$ (V/cm)/μs.

In nonorthogonal fields and/or with nonhydrogenic atoms, CFM acts similarly. Whatever the exact shape of the diagram is, the state with the largest energy is nondegenerate and possible crossings are avoided. The atoms initially prepared with maximum energy are at the end in the state whose energy is maximum in the $\mathbf{B}$ field, i.e., in the circular state.

For a classical hydrogenic system, external fields remove the $1/r$ symmetry, and make the valence-electron elliptical orbit precess slowly, with a constant major axis size. For low orthogonal $\mathbf{E}$ and $\mathbf{B}$ fields, there is a very peculiar orbit which remains unperturbed: the electric and magnetic force effects balance each other over one period. This ellipse is orthogonal to the $\mathbf{B}$ field, and its major axis is oriented along the $\mathbf{E}$ field. Its eccentricity is[5]

$$e=\tfrac{3}{2}nE[(B^2/4)+(g/4)n^2E^2]^{-1/2} \quad (2)$$

(where $\mathbf{E}$ and $\mathbf{B}$ are expressed in atomic units). This condition implies an extremum of dipole-field energy $-\mathbf{E}\cdot\mathbf{D}-\mathbf{B}\cdot\mathbf{M}$ (where $\mathbf{D}$ and $\mathbf{M}$ are the electric and magnetic dipoles). Any state involved in CFM (initial, intermediate, or final), having the largest energy for a given set of $\mathbf{E}$ and $\mathbf{B}$ fields [upper level in Figs. 1(a), 1(b), or 1(c)], corresponds to such an elliptical orbit. The initial state $m=0,n1=n-1$ corresponds to a linear degenerate ellipse ($e=1$); the circular state corresponds to a circle ($e=0$); and the intermediate states correspond to ellipses whose eccentricity decreases adiabatically to zero during the CFM process. These states are the quasiclassical Bohr-Sommerfeld states: The valence-electron distribution is a thin elliptical torus. Each of them is stable in crossed fields obeying Eq. (2).

The experimental setup is shown in Fig. 2 (bottom). Lithium atoms of a thermal horizontal beam (with a mean atomic velocity of 1500 m/s) are prepared in the initial level, interact with the fields, and are detected in three separate zones. Preparation is performed by yttrium-aluminum-garnet pumped-pulsed-laser excitation inside a stack of equally spaced metallic plates, which provides a homogeneous electric field parallel to

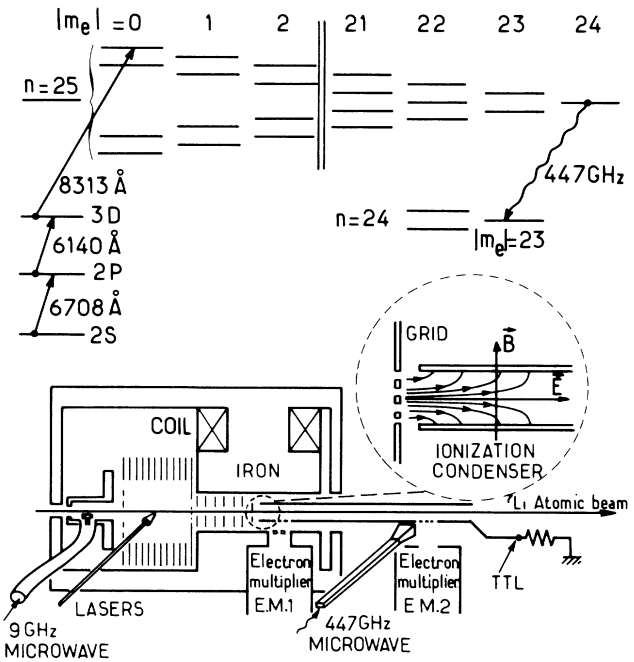

FIG. 2. (Top) Energy levels of lithium relevant for the experiment. Some of the Stark states of the $n=25$ are represented on the right, the various m manifolds being displayed as vertical energy scales. (Bottom) Cross section of the experimental setup in the atomic beam plane. The place where the switching process occurs is enlarged.

the atomic beam axis. The field amplitude (−200 V/cm) provides the energy splitting necessary to prepare selectively the initial $n=20$ to 25, $m=0,n1=n-1$ level. Lasers are π polarized with respect to the field and are in resonance with the $2s-2p$, $2p-3d$, and $3d-n,m=0$, $n1=0$ transitions, at the wavelengths indicated in Fig. 2 (top). The atoms pass through a second stack of plates, interacting with a smaller field (−10 V/cm). Then they enter between two condenser plates where they are detected by the field ionization method[8]: A ramp of electric field is applied across the condenser and ionization takes place at different times for the various $n,m,n1$ states. The whole setup is placed inside a liquid-nitrogen-cooled iron shield, which also acts as the frame of an electromagnet producing a $\mathbf{B}$ vertical field (15 to 100 G). The ionization condenser overlaps the electromagnet gap on the exit side of the iron shield. Atoms are ionized and detected either in the electromagnet gap, or outside the shield with two separate electron multipliers.

The atoms experience the crossed fields when entering the ionization condenser. This part of the setup is enlarged in the inset of Fig. 2. The atoms leave the second stack of plates passing through a thin metal mesh located 3 mm before the 3.4-mm-spaced condenser plates. The mesh potential is −3 V and the plates are close to the ground. The higher plate potential is adjusted to lower

the vertical component of **E**. The plates act as an electric shield, and the **E** field seen by the atoms decreases to zero at the condenser entrance. The atoms pass adiabatically from the vicinity of the metal mesh, where the electric coupling is dominant, to the inside of the condenser, where they interact with the **B** field alone. As dE/dt is typically 3 (V/cm)/μs, the adiabatic condition [Eq. (1)] is fulfilled, and the atoms become circular with their angular momentum oriented along **B**.

A 5 ns, rising, 0 to 3 V transistor-transistor-logic (TTL) pulse is then applied to the lower electrode of the ionizing condenser, which is grounded by a 50-Ω terminating impedance. The circular atoms thus pass suddenly through parallel fields, from a dominant **B** field back to a dominant **E** field, keeping their circular character. Figures 1(c)–1(e) show the energy diagram before, during, and just after this back process, respectively. Diagram 1(c) is a Zeeman diagram, 1(e) is a Stark diagram where the Zeeman energy removes the degeneracy, and 1(d) corresponds to intermediate coupling. This last diagram exhibits many level crossings. As the fields are not perfectly aligned, many of these crossings are avoided (see inset of Fig. 1). As the electric field rises fast [2000 (V/cm)/μs], the atoms pass diabatically through the crossings and remain circular. The condition of diabaticity is related to the frequency width $\Delta\nu$ of the largest avoided crossing, and to the time taken by the atoms to pass through it. For $n=25$, $B=100$ G, and for a typical 2° angle between **E** and **B**, $\Delta\nu$ is about 100 MHz, and the diabatic condition is $dE/dt \gg 15$ (V/cm)/μs. With a slower rising field, the atoms follow adiabatically or partly adiabatically the avoided crossings, and evolve towards lower angular-momentum states.

Ionization occurs 30 μs after the laser excitation with a flight length of 6.5 cm. The ionizing field rises from 0 to 3.75 kV/cm in 10 μs. Time-resolved ionization signals, detected in the electromagnet gap, are shown in Figs. 3(a) to 3(d). Figure 3(a) shows the initial $(n,m=0,n1=n-1)$ state signal with a short time peak. It corresponds to a low ionization threshold, about twice less than for the $n,m=0,n1=n-1$ state of hydrogen. In lithium the $m=0$ and $m=1$ states are perturbed by the atomic core, and have nonhydrogenic properties, such as lower ionization thresholds.[9] Figure 3(b) shows the CFM circular-state signal when the TTL pulse is applied just before (about 1 μs) the beginning of the ionizing pulse. This signal exhibits a large $n=25$ circular peak which takes place in the characteristic circular ionizing **E** field (2.0 kV/cm for $n=25$), and two smaller $n=24$ and $n=26$ side peaks related to the thermal transfer from the $n=25$ circular state.[1] Figure 3(c) shows the same signal without TTL pulse. It has a different shape with two peaks. The first, which occurs in the same field as in Fig. 3(a), corresponds to $m=0$ and 1 states. The second peak is broader than the Fig. 3(b) circular peak, with a small shift towards high ionizing field: It reveals (with still circular atoms) noncircular atoms in lower angular-momentum states $(n-1 > |m| > 1)$. Both classes of noncircular states are populated during the ionizing ramp by atoms which pass partly adiabatically through the Fig. 1(d) avoided crossings: The very beginning of the ramp rises parabolically and does not fulfill the diabatic condition.

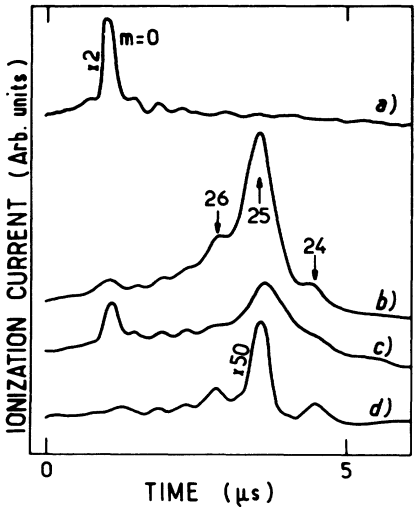

FIG. 3. Time-resolved ionization signals detected in the electromagnet gap. (a) Initial $m=0,n2=0$ state signature. (b) and (c) CFM circular state signal with and without the TTL pulse, respectively. (d) AMTM circular-state signal.

A first test experiment is performed by preparation of circular atoms by AMTM in the same setup. The microwave horn (at left in Fig. 2 bottom) in front of the first stack of plates radiates the needed 9-GHz microwave. The AMTM signal [Fig. 3(d)] is very similar to the CFM one, with a large $n=25$ circular peak, and two smaller side peaks. Although both CFM and AMTM methods are 100% efficient, the CFM signal is typically 50 times larger than the AMTM one. This is due to a more efficient laser excitation of the CFM initial level. The CFM circular peak is broader than the AMTM one. Because of the nonhydrogenic character of the lithium S states, the $m=0,n1=n-1$ CFM initial level is quasidegenerate with the $m=1,n1=n-2$ level (about 3-GHz energy splitting for $n=25$) and both levels are prepared by the third-step laser whose linewidth is 3 GHz. Atoms initially prepared in the $m=1,n1=n-2$ level become quasicircular ($|m|=n-2$) and broaden the AMTM ionization signal. A narrower linewidth laser would suppress this unwanted effect.

A second test control is made by observation of the $n=24$ to $n=25$ circular-circular transition at 447 GHz.[2] The experiment is performed out of the iron shield, where the **B** field is low (about 1 G). The observed transition, which has a typical 5-MHz full width, is within 3

MHz at the expected frequency. As the TTL field (10 V/cm) shifts the other resonances (noncircular-to-circular transitions) by at least 20 MHz, this proves that circular atoms have been actually prepared.

This experiment demonstrates the efficiency of the crossed-fields method for preparing circular atoms. This method has many advantages.

(i) CFM is a nonresonant process which is very insensitive to experimental adjustments: Changing them is not required when changing n.

(ii) CFM selectively prepares one of the circular helicities (which may be reversed by changing the **B** field sign).

(iii) Laser excitation of the initial level is generally more efficient in the CFM case. This yields larger signals than the AMTM ones.

(iv) CFM requires a low electric field which does not quench the metastable $2S$ state of hydrogen. Circular states of hydrogen might thus be prepared.

(v) CFM allows us to prepare circular states of any atomic species. It would be interesting to obtain circular states of two-electron atoms which are good candidates for preparing stable (low autoionizing rate), doubly excited Rydberg atoms.[10]

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