RYDBERG ATOMS IN CROSSED ELECTRIC AND MAGNETIC FIELDS. EXPERIMENTAL EVIDENCE FOR THE PAULI QUANTIZATION

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The experimental evidence for the Pauli quantization obeyed by the Rydberg spectrum of rubidium, in crossed electric and magnetic fields, is reported. When the external field perturbation associated with Zeeman and linear Stark effects are of the same order but small compared to the Coulomb binding energy, the energy levels of the system are given by $E_{n,k} = -R/n^2 + \hbar K(\omega_L^2 + \omega_E^2)^{1/2}$, where K is an integer ω_L, ω_E the Larmor and linear Stark frequencies respectively.

Major progress has been achieved in the last few years in understanding the fundamentals of atoms in external electric or magnetic fields [1]. Solving the problem of atoms in crossed electric and magnetic fields is now one of the most attractive future goals. The topics have already raised numerous conjectures about a possible double well behaviour arising from the combined effects of the Coulomb, electrostatic and diamagnetic interactions. This would lead to trapping of the Rydberg electron at large distance from the nucleus [2–6].

Our present concern is the low crossed $(\boldsymbol{E},\boldsymbol{B})$ fields regime which is established on reliable theoretical grounds [7,8]. Here the electric and magnetic field effects are small compared to those of the Coulomb binding field. Their role is merely to break the Coulomb supersymmetry in a way which has been first established by Pauli [7]. The basic characters of such a "Pauli quantization" for the hydrogenic manifold can be readily obtained from the expression for the perturbing terms in the hamiltonian (neglecting the diamagnetic interaction):

$$W = -(q/2m)\,\boldsymbol{B}\cdot\boldsymbol{L} - q\,\boldsymbol{r}\cdot\boldsymbol{E} \tag{1}$$

by making the usual Pauli replacement $\boldsymbol{r} \to \frac{3}{2} n\boldsymbol{a}$ and introducing the $\boldsymbol{j}_{1,2} = (\boldsymbol{L} \pm \boldsymbol{a})/2$ generators of the O(4) Lie algebra of the Coulomb problem ($\boldsymbol{a}$ is the scaled Lenz vector). Introducing the $\boldsymbol{\Omega}_{1,2}$, angular frequencies such that

$$\boldsymbol{\Omega}_{1,2} = (\boldsymbol{\omega}_L \pm \boldsymbol{\omega}_E),$$

$$\Omega_1 = \Omega_2 = \Omega = (\omega_L^2 + \omega_E^2)^{1/2}, \tag{2}$$

where $\boldsymbol{\omega}_L = |q|B/2m$ and $\boldsymbol{\omega}_E = \frac{3}{2}(4\pi\epsilon_0\hbar/mq)^{1/2} n\,E$ are respectively the Larmor and linear Stark frequencies, one gets the expression of the adiabatic invariant to first order in E and B [7,8]:

$$W = \boldsymbol{\Omega}_1\cdot\boldsymbol{j}_1 + \boldsymbol{\Omega}_2\cdot\boldsymbol{j}_2 = \hbar\Omega(j_{1\Omega_1} + j_{2\Omega_2}),$$

$$-(n-1) \leqslant K = j_{1\Omega_1} + j_{2\Omega_2} \leqslant (n-1). \tag{3}$$

The manifold is split into $(2n-1)$ K labelled components (K is an integer) which do present a residual degeneracy $n - |K|$. The spacing between two adjacent sub-levels is Ω and the wavefunctions are of the $\{j_1^2 j_2^2 j_{1\Omega_1} j_{2\Omega_2}\}$ types. This manifests the existence of a completely new type of quantization and organization in the spectrum in crossed $(\boldsymbol{E}, \boldsymbol{B})$ fields, different from either the linear Stark or the Zeeman ones. Although this is a direct consequence of the supersymmetry of the Coulomb problem, thus applying primarily to the hydrogen atom, the experimental evidence carried out below has been obtained on rubidium atoms.

Under the action of external fields, most of the states in alkali atoms should follow the basic hydrogenic

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laws [9–11]. The only requirement is that the external field perturbations are dominant over the role of quantum defects. States with $l > 3$ fulfill such a condition at the lowest field values and consequently will behave as an incomplete (some components may be missing) hydrogenic manifold.

While the experimental technique is a variant of Doppler free two-photons spectroscopy on Rb atoms, the experimental method is based on the detection of the anticrossings (AC) of the nS series with the components of the $(n-3)$ quasi-hydrogenic manifold. Owing to their large quantum defects ($\delta(S) = 3.1312$) the nS states do have strong non-hydrogenic behaviours at low fields which results in a very weak field dependence of their diabatic energy curve. The tracking of the positions of the AC as a function of the fields strengths will provide us with a direct test of the quantization law obeyed by the incomplete hydrogenic manifold *at nearly constant electron energy.* This is schematized on fig 1.

The experimental set-up is based on a 800 mW R6G Ar^+ pumped cw ring dye laser with complete servo control of the 1 MHz linewidth [12]. The frequency is measured to within 3 MHz accuracy by means of a 75 MHz free spectral range PF interferometer. The laser beam is focussed into a thermoionic detector working in the space charge limited regime, with silica Brewster angle windows and filled with Rb vapour (4×10^{-3} Torr pressure). The electrode arrangement allows us to apply a small electric field (in the range 0–20 V/cm) between a mesh and a plane electrode. The magnetic field is produced by means of air coils in Helmoltz position giving up to 700 Gauss (RMN calibrated – 10^{-5} inhomogeneity). The Rayleigh length is of about 2 mm which limits the effects of the electric field's inhomogeneity in the interaction region. The E and B fields are crossed to within 2°.

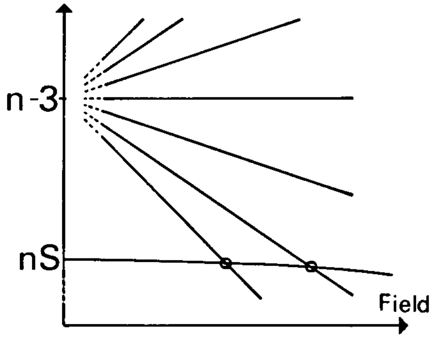

Fig. 1. Sketch of the experimental situation. The anticrossings of the nS diabatic curve with the components of the $(n-3)$ hydrogenic manifold are used for checking the quantization at nearly constant energy.

One fundamental improvement is the use of FM modulation techniques of the laser frequency (around 230 Hz). The recorded two-photons signal (fig. 2) is the derivative of the usual lorentzian-shaped one while the signal to noise ratio is only limited by the specific FM noise. The detection of nS series up to 150 in zero field has been achieved this way.

The study of the Stark map of the nS, nP and nD series allows us to check the experimental method and to deduce an accurate calibration of the electric field strength in the interaction region. While the nS diabatic energy curve presents a small quadratic Stark shift, the $M = 0$ states of the $(n-3)$ incomplete manifold do behave linearly with the field strength [‡1] (fig. 1). As a result of the weak coupling, scaling as $n^{23/2} E^3$, the transfer of oscillator strengths takes place in the very vicinity of the AC with the nS curve, while the sizes of AC are in the range 40 to 200 MHz, thus en-

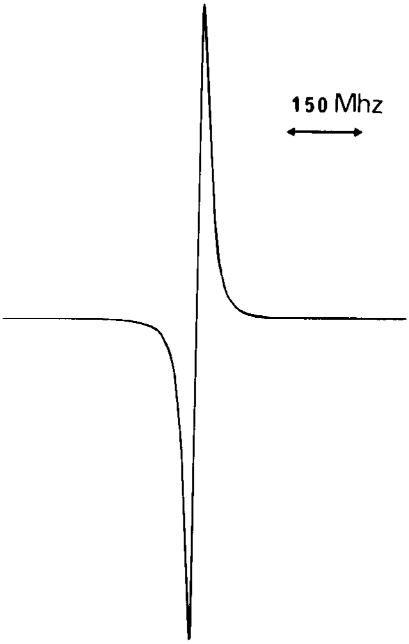

Fig. 2. FM Doppler-free two photons lineshape (^{85}Rb 5S → 37S transition).

[‡1] The red shift of the 37S is 1200 MHz at maximum while states of the $n = 34$ manifold have experienced a 22 GHz red shift at the AC.

suring the accuracy of the method. Both positions and sizes of the AC are in excellent agreement with the theory.

As to the B field action, its main result is a splitting of the two-photon line into several components associated with the ground state hyperfine structure. The hyperfine and fine structures are negligibly small in the manifold while the spin effects are trivial.

The main requirements of the experimental method are thus fulfilled. In crossed (E, B) fields, the AC are detected through the strong decrease in the two photons signal which is induced by the locally resonant coupling with one state of the manifold. As shown on fig. 3, the sensitivity of the method is typically 5×10^{-2} V/cm hence permitting an accurate determination of the positions of AC.

The results of the tracking of the AC of the 37S with the components of the $n = 34$ manifold in (E, B) fields are displayed in fig. 4, at nearly constant energy in the spectrum [‡1]. Such a (E^2, B^2) plot [‡2] supplies us with the striking evidence that the sublevels in the manifold are quantized according to a completely new law in crossed (E, B) fields which indeed is a remnant

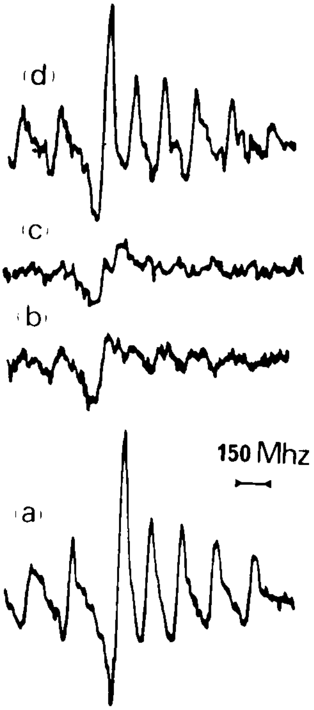

Fig. 3. Modulated two photons signal in crossed (E, B) fields (5S → 37S transition). The B field is fixed at 350 G. The electric field values are from (a) to (d) 16.03, 16.57, 16.61, and 17.08 V/cm, respectively. All the plots are on the same scale. A strong decrease of the signal occurs at anticrossings.

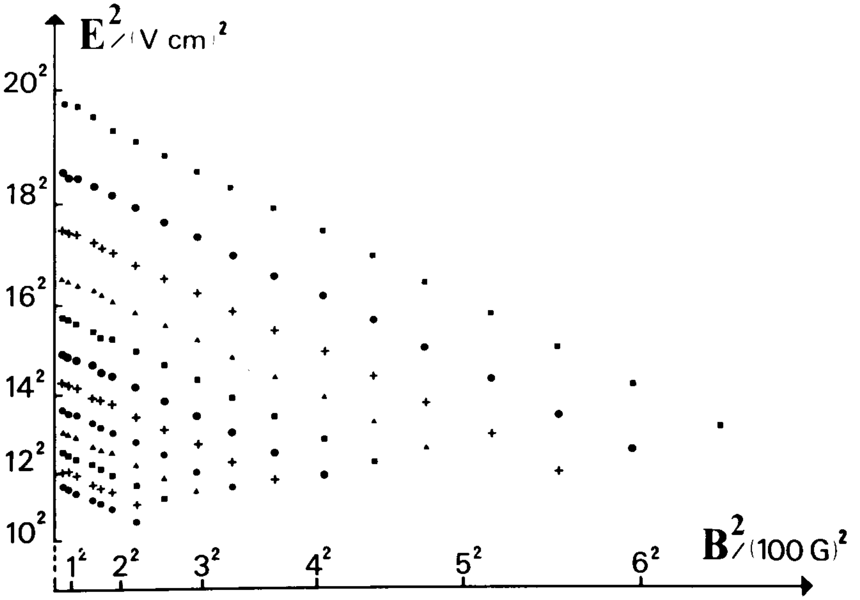

Fig. 4. Plot of the AC of 37S with states of the $n = 34$ manifold (at nearly constant electron's energy) displaying the new quantization regime in the manifold.

[‡2] The passing from the E to the crossed (E, B) fields regime presents a finite width in B field (about 50 G) before the behaviour of AC becomes regular while their number is twice the number in the E field alone. AC will transform into crossings at vanishing E (Zeeman regime).

Table 1
Comparison of the experimental results with the hydrogenic predictions (eq. (3)). The experimental values are corrected for second order Stark effect and diamagnetism (n^* and K^* are the experimentally deduced values)

n = 34/37S			n = 39/42S		
n^*	$n^*/34$	$\lvert K^*\rvert$	n^*	$n^*/39$	$\lvert K^*\rvert$
34.04	1.001	29.44	39.48	1.012	25.11
33.94	0.998	28.47	39.44	1.011	24.18
33.93	0.998	27.43	39.25	1.006	23.18
34.00	1.000	26.35	39.38	1.010	22.06
33.95	0.999	25.34	39.30	1.008	21.15
33.90	0.997	24.32	38.78	0.994	20.22
33.76	0.993	23.41	38.59	0.989	19.31
33.79	0.994	22.34	38.35	0.983	18.33
33.73	0.992	21.33	37.75	0.968	17.40
33.41	0.983	20.41	38.27	0.981	16.28
33.49	0.985	19.36	37.84	0.970	15.28
33.51	0.986	18.30			

of the underlying supersymmetry of the Coulomb problem.

A full comparison with the predictions from the hydrogenic theory (eq. (3)) can be made from table 1, in which n^* and K^* are the experimentally deduced values. The roles of the second order Stark effect and diamagnetism are fully accounted for here. The dominant correction comes from the E field dependence of the nS diabatic curve [‡1] while its diamagnetic blue shift represents 3.3×10^{-4} MHz/G^2 for n = 37S. As far as the states of the manifold are concerned, the second order (hydrogenic) Stark effect represents -0.38 MHz/(V/cm)^2 and the diamagnetic shifts are 4×10^{-4} MHz/G^2 on the average (for n = 34). The latter have been calculated exactly assuming a purely hydrogenic behaviour of the wavefunctions.

The experimental results are then completely consistent with the hydrogenic formulation. The absolute n^* value agrees with the hydrogenic one to within 2% while the successive K^* values are equally separated by one. This establishes the basic n^2 and K dependences in (3) leading to a $h(\omega_E^2 + \omega_L^2)^{1/2}$ spacing of the energy levels in the manifold. However slight departures from (3) are noticeable. The non integer character of K^* cetainly manifests a departure from a pure Coulomb behaviour in Rb and the incomplete character of the manifold. A detailed analysis is a matter of computational work not considered here.

Notice that the present experiments are an extremely severe test of equation (3). Indeed, the wavefunctions depend on $\boldsymbol{E}$ and $\boldsymbol{B}$, and for a given K^* value in the plot of fig. 3, their character should completely change from one point to another. This makes the agreement even more striking.

A preliminary study of the crossed $(\boldsymbol{E}, \boldsymbol{B})$ fields spectrum has been reported in ref. [13] with similar goals but using different techniques. Unfortunately, the maximum value (0.1) of the ratio ω_L/ω_E was too small for the Pauli quantization regime be established [‡2]. The patterns in ref. [13] are consequently not regular.

Therefore this work is the first experimental demonstration of the existence of a highly organized regime with new quantization laws, taking place in the atomic spectra in crossed $(\boldsymbol{E}, \boldsymbol{B})$ fields. The Zeeman and linear Stark behaviours are contained as limiting situations. The experimental results also highlight the fact that the dynamics of most atomic states for any atom in external fields is similar to that of the hydrogen atom, even though such states are not seen in optical spectra.

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