Experimental Study and Analysis of the Lithium Atom in the Presence of Parallel Electric and Magnetic Fields

P. Cacciani, E. Luc-Koenig, J. Pinard, C. Thomas, and S. Liberman
Laboratoire Aimé Cotton, Centre National de la Recherche Scientifique, Campus d'Orsay, 91405 Orsay Cedex, France
(Received 26 December 1985)

The structure of the $n=30$, $M=0$ manifold of lithium has been investigated experimentally in the presence of weak parallel electric and magnetic fields. The main properties of the spectra and their evolution with increasing field strengths are interpreted from the analysis of the approximate integral of motion obtained for hydrogen. The lithium spectra exhibit additional alterations which are typical of the short-range core effects.

PACS numbers: 32.60.+i, 31.60.+b

Recent experimental work, performed on lithium atoms in the presence of a magnetic field, has led to the observation of the $M=0$ odd states of a diamagnetic manifold (M being the azimuthal quantum number).[1] The quantum defect δ_p of the p states is small enough for the odd states of lithium to be regarded as quasihydrogenic ones. The quantitative analysis of the experimental spacing between two consecutive components has confirmed the existence of the two types of behavior previously predicted for atomic hydrogen in the framework of Solov'ev's semiclassical theory.[2] In the inter l-mixing regime, each n manifold can be treated separately and the diamagnetic energy shift E_D is expressed to first order in B^2 as

$$E_D=(\gamma^2 n^2/16)[n^2+M^2+n^2\Lambda], \quad (1)$$

where $\gamma=B/B_c$ ($B_c=2.35\times10^5$ T). The approximate constant of the motion Λ is related to the Runge-Lenz vector $\mathbf{A}$ by the expression $\Lambda=A^2(4-5\cos^2\theta)$ [θ being the $(\mathbf{B},\mathbf{A})$ angle.] Since $|\mathbf{A}|$ remains less than 1, the Λ values are restricted to the interval $[-1,+4]$.

For a given diamagnetic energy, the extremity of $\mathbf{A}$ moves on the twofold hyperboloid of revolution $\Lambda=\text{const}$ [Figs. 1(a) and 1(b)]. The value $\Lambda=0$ defines the double cone specified by the B- and n-independent condition $\cos^2\theta_0=\frac{4}{5}$. For the lower-energy part of the hydrogenic manifold ($\Lambda<0$), the hyperboloid consists of two symmetrical sheets: Odd and even states form doublets and their wave functions are stretched along the $\mathbf{B}$ direction. The upper-energy part ($\Lambda>0$) is associated with a single sheet: Even and odd states are no longer degenerate but are approximately equally spaced; they are mainly localized in the plane perpendicular to the $\mathbf{B}$ axis.

When an electric field $\mathbf{F}$ is applied in the $\mathbf{B}$ direction, the symmetry of revolution around the z axis is preserved and M remains an exact quantum number. However, the $\mathbf{F}$ field mixes states with opposite parity so that, since the lowest states of the manifold are degenerate, one can expect a linear Stark effect which removes the degeneracy; in contrast the nondegenerate upper states are expected to be perturbed by a quadratic Stark effect. On the other hand, the spatial localization along the z axis of the lowest states makes them very sensitive to the electric perturbation Fz. In the present paper, the strengths of both fields are supposed to be weak enough so that n can be considered as a good quantum number.

This paper aims to analyze in detail the evolution of the structure of a lithium diamagnetic manifold with increasing electric field. The experiment is performed on an atomic beam of lithium excited with a uv laser source (230 nm). The one-step excitation from the ground state with π-polarized light selects the $M=0$

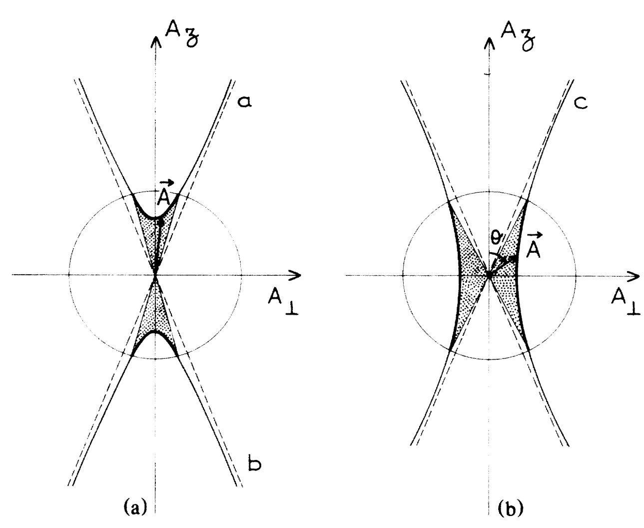

FIG. 1. Secular variations of the Runge-Lenz vector $\mathbf{A}$ under the action of a magnetic field $\mathbf{B}$ parallel to the z direction, valid when $\gamma^2 n^7 \ll 1$ and $M=0$ (from Ref. 3). $\mathbf{A}$ is directed along the major axis of the Kepler ellipse and $|\mathbf{A}|$ is the eccentricity. $|\mathbf{A}|$ is smaller than 1, therefore $\mathbf{A}$ is bounded by the sphere of unit radius centered at the origin. For a given diamagnetic energy, the extremity of $\mathbf{A}$ moves on the twofold hyperboloid of revolution defined by $\Lambda=4A^2-5A_z^2=\text{const}$ and limited by a double cone $\cos^2\theta_0=\frac{4}{5}$. (a) $\Lambda<0$; (b) $\Lambda>0$.

states. A magnetic field **B** and an electric field **F**, both directed along the beam axis, are applied in the interaction region. The evolution with increasing electric field of the structure of the $n=30$, $B=2.33$ T diamagnetic manifold is shown in Fig. 2. As expected, a completely different behavior occurs in the lower ($\Lambda < 0$) and the upper ($\Lambda > 0$) parts of the spectrum. Large and complex modifications appear in the low-energy range where the intensity distribution among the different components strongly differs from the original one. The energies of the lowest components decrease rapidly with increasing F, with a linear variation for sufficiently large electric field strength; simultaneously, additional lines appear at intermediate energy. The upper part of the manifold is only weakly perturbed: Even for the highest value of F, the component spacing and the intensity repartition do not change.

On the other hand, surprisingly, the parity mixing induced by F does not lead to the appearance of additional lines. Actually, this result can be explained by considering the diamagnetic structure of odd and even

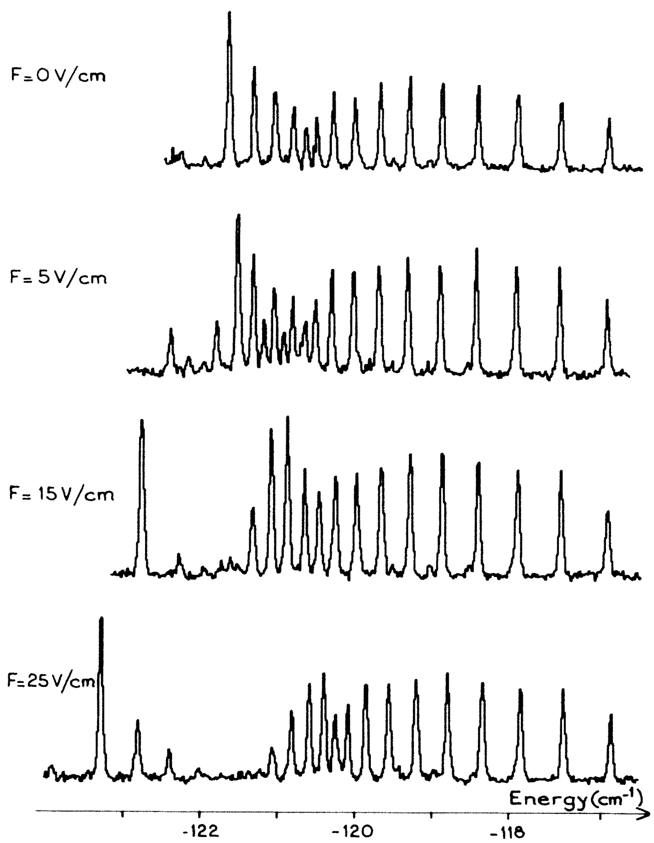

FIG. 2. Experimental recordings of the manifold $n=30$, $M=0$ of Li in the presence of parallel electric and magnetic fields. The energies are referred to the zero-field ionization limit. The spectra correspond to $B=2.33$ T and to increasing F.

states of lithium. Indeed, for s states the quantum defect is so large ($\delta_s = 0.4$) that in the present experimental situation ($n=30$, $B \lesssim 2.5$ T) the diamagnetic shift remains of the same order of magnitude as the non-Coulombic part of the energy. This induces a complete modification of the pure diamagnetic structure ($F=0$): Now, odd and even states alternate in the lower part of the manifold while pairs of quasidegenerate odd and even states are present in the upper part. This anomalous structure is not typical of lithium but could be observed in other alkali-metal atoms, as has been pointed out analytically by Braun.[4] This has been confirmed by numerical diagonalization after the introduction of the non-Coulomb character of lithium.[5]

In order to analyze the different recorded spectra (Fig. 2), we have generalized the semiclassical approach of Solov'ev[2] to the case where an electric field F is applied.

In the hydrogen atom, the operator relation $\mathbf{r} = -\frac{3}{2}n^2\mathbf{A}$ is valid within a given subspace.[6] Consequently, in the ℓ-mixing regime the Hamiltonian describing the perturbation due to weak enough parallel magnetic and electric fields can be written in a form similar to Eq. (1) by changing Λ into Λ_β:

$$\Lambda_\beta = 4A^2 - 5(A_z - \beta)^2 + 5\beta^2. \qquad (2)$$

In atomic units $\beta = \frac{12}{5}n^2 f/n^4\gamma^2$ and $f = F/F_c$ with $F_c = 5.142 \times 10^9$ V/cm. The parameter β ($0 \leqslant \beta < \infty$) measures the relative strength of the linear Stark effect with respect to the diamagnetic interaction. The generalized constant of the motion Λ_β is valid to first order in B^2 and F. Quantitative information on the low-field Stark structure of a hydrogenic diamagnetic manifold can be deduced from the analysis of the secular variations of the Runge-Lenz vector **A** under the simultaneous actions of the electric and magnetic fields.[7] From Eq. (2), the extremity of **A** moves on a hyperboloid of revolution. The limiting cone is defined by the unchanged θ_0 angle but the top is shifted in the z direction by 5β (Fig. 3).

The sphere that bounds the modulus of **A** is not modified by the field F. Consequently, the geometrical symmetry of the problem with respect to the $z=0$ plane disappears. The intersections between the hyperboloid and the sphere depend on the β value. For $M=0$, three cases, limited respectively by $\beta = 0$, $\frac{1}{5}$, and 1, must be considered. Figures 3(a) and 3(b) correspond to the case where $0 < \beta < \frac{1}{5}$. For a given energy state, the extremity of the vector **A** moves on one single sheet of the hyperboloid. The z projection of **A** is limited by either the extremal points of the hyperboloid A_1^- and A_2^-, or the intersection points of the two surfaces A_1^+ and A_2^+. As a consequence, three classes of states, I, II, and III, can be defined depending on the extremal A_z values according to Table I.

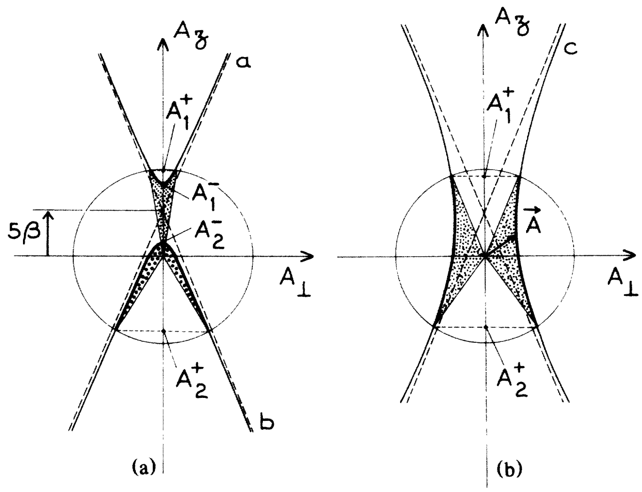

FIG. 3. Secular variations of the Runge-Lenz vector **A** under the joint actions of magnetic **B** and electric **F** fields parallel to the z direction, valid when $\gamma^2 n^7 << 1$ and $fn^5 << 1$, and $M=0$. The extremity of **A** moves on the hyperboloid of revolution $\Lambda_\beta=\text{const}$ [Eq. (2)] with $\beta=\frac{12}{5} f/n^2\gamma^2$. Compared to Fig. 1 the limiting double cone is shifted in the z direction by the quantity 5β. This figure corresponds to $\beta<\frac{1}{5}$. A_1^-, A_2^-, z projections of the extrema of the hyperboloid; A_1^+, A_2^+, z projections of the intersections between the sphere and the hyperboloid. (a) When $\Lambda_\beta \leq 25\beta^2$ the hyperboloid consists of the two no longer equivalent sheets a and b, respectively, corresponding to the I and II classes of states. (b) When $\Lambda_\beta \geq 25\beta^2$ the single sheet c corresponds to the class III.

Furthermore, in this classical approximation, the wave functions of states belonging to the same class can be characterized by a specific spatial localization.

For $\beta=0$ (i.e., for a pure diamagnetic manifold), classes I and II possess the same energy extension $-1 \leq \Lambda_\beta \leq 0$, while class III corresponds to $0 \leq \Lambda_\beta \leq 4$, in agreement with a previous study.[8] As soon as β is nonzero, classes I and II are no longer equivalent and the corresponding states are no longer degenerate. The lower bounds of Λ_β vary linearly with β, which corresponds to the linear Stark effect. With regard to the class-III states the modification is β^2 dependent, which corresponds to the quadratic Stark effect. Furthermore, as soon as $\beta=\frac{1}{5}$, class II disappears, and for β larger than 1, only class I remains.

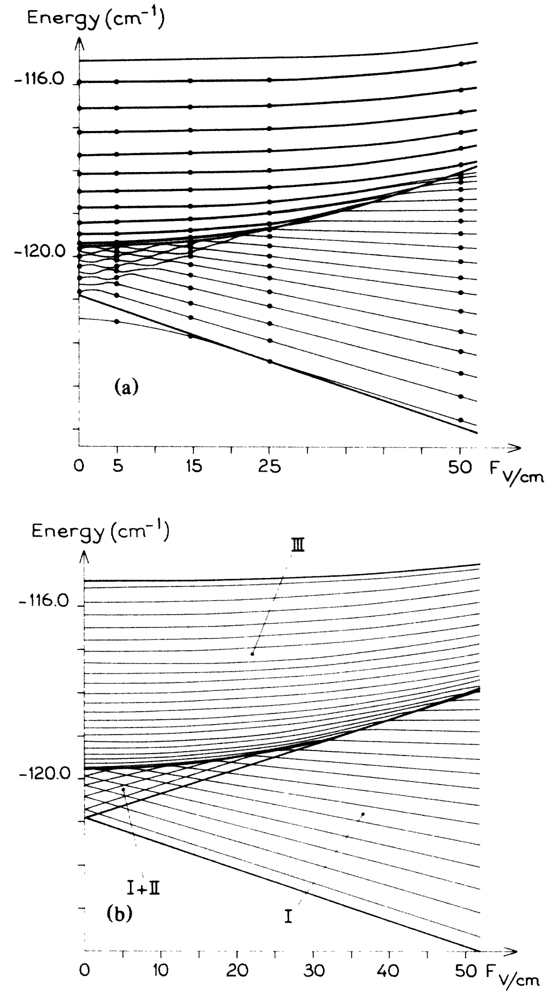

FIG. 4. Evolution with increasing field F of the structure of the $n=30$, $M=0$, $B=2.33$ T manifold (**F**‖**B**). (a) Lithium atom; (b) hydrogen atom. Closed circles, experimental values; solid lines, calculated values, in both parts. The three classes of states I, II, and III appear in the domains limited by the heavy curves and defined in Table I.

TABLE I. Classification of the $M=0$ states of a hydrogenic n manifold in the presence of parallel electric and magnetic fields. The three different types of classes are defined from the conditions limiting the A_z projection of the Runge-Lenz vector ($A_1^+, A_2^+, A_1^-, A_2^-$ are defined in Fig. 3). The energy extension of the different classes depends on the β value and is shown in columns 3–5. Λ_β is related to the energy shift E_β by $E_\beta=(\gamma^2 n^4/16)(1+\Lambda_\beta)$.

Class	Limiting A_z motion	Energy extension $\beta<\frac{1}{5}$	$\frac{1}{5}<\beta<1$	$\beta<1$
I	$A_2^+ < A_z < A_2^-$	$-1-10\beta \leq \Lambda_\beta \leq 25\beta^2$	$-1-10\beta \leq \Lambda_\beta \leq -1+10\beta$	$-1-10\beta \leq \Lambda_\beta \leq -1+10\beta$
II	$A_1^- < A_z < A_1^+$	$-1+10\beta \leq \Lambda_\beta \leq 25\beta^2$		
III	$A_2^+ < A_z < A_1^+$	$25\beta^2 \leq \Lambda_\beta \leq 4+5\beta^2$	$-1+10\beta \leq \Lambda_\beta \leq 4+5\beta^2$	

The above results have already been obtained by Braun and Solov'ev[9] from a completely different approach where, in a given hydrogenic n manifold, the diagonalization of the total perturbation (**F** and **B**) is performed by use of a discrete analog of the Wentzel-Kramers-Brillouin analytical method.

The present experiment is performed on lithium with β values restricted to the range $[0,\frac{1}{5}]$ (Fig. 2). The three classes of states exist and their observed energy extensions agree with the corresponding ones calculated in hydrogen.

For the lithium atom, all of the spectra recorded for the same value of the field B but for different F strengths can be analyzed together: The evolution with increasing β of the structure of the $n=30$, $B=2.33$ T manifold is presented in Fig. 4(a). This experimental map agrees completely with the calculated one obtained by diagonalization of the total Hamiltonian.[5] Figure 4(b) reports the corresponding theoretical map obtained for atomic hydrogen. Similar global behavior is observed in the two cases, especially with regard to the properties of the three classes of states, namely, energy extension, evolution of components, and disappearance of classes II and III. Nevertheless, significant differences arising from the nonhydrogenic character of lithium are observed. The upper states remain quasidegenerate even for $\beta > 0.14$, that is, when the $30s$ component merges into the manifold. Furthermore, as can be seen in the lower-energy part of the map [see Fig. 4(a)], states of classes I and II exhibit large anticrossings.

In summary, this paper reports, for the first time, a detailed experimental analysis of the evolution of the structure of a lithium diamagnetic manifold under the influence of an electric field. The experimental evidence for the existence of different classes of states has been interpreted by the introduction of a new constant of motion Λ_β, valid for atomic hydrogen perturbed by parallel fields **F** and **B** in the inter l-mixing regime. The global features of the recorded structures are similar to the corresponding ones of atomic hydrogen, but in the lithium atom the ionic core induces significant alterations in the spectra, which persist even at high field strengths: quasidegeneracy, anticrossing, and intensity distribution. As previously pointed out in the analysis of the Stark effect for nonhydrogenic atoms,[10] the hydrogenlike properites are linked to the long-range interactions (Coulomb, Stark, and diamagnetic in the present case). However, the short-range core effects modify the wave functions near the nucleus, so that the spectrum takes on an additional character, typical of the studied atom.

The Laboratoire Aimé Cotton is associated with the University Paris-Sud.

[1]P. Cacciani, E. Luc-Koenig, J. Pinard, C. Thomas, and S. Liberman, Phys. Rev. Lett. **56**, 1124 (1986).

[2]E. A. Solov'ev, Pis'ma Zh. Eksp. Teor. Fiz. **34**, 278 (1981) [JETP Lett. **34**, 265 (1981)], and Zh. Eksp. Teor. Fiz. **82**, 1762 (1982) [Sov. Phys. JETP **55**, 1017 (1982)].

[3]J. C. Gay and D. Delande, Comments At. Mol. Phys. **13**, 275 (1983).

[4]P. A. Braun, J. Phys. B **16**, 4323 (1983).

[5]E. Luc-Koenig, S. Liberman, and J. Pinard, Phys. Rev. A **20**, 519 (1979).

[6]W. Pauli, Z. Phys. **36**, 336 (1926).

[7]P. Cacciani, E. Luc-Koenig, J. Pinard, C. Thomas, and S. Liberman, to be published.

[8]P. A. Braun, Zh. Eksp. Teor. Fiz. **84**, 850 (1983) [Sov. Phys. JETP **57**, 492 (1983)].

[9]P. A. Braun and E. A. Solov'ev, Zh. Eksp. Teor. Fiz. **86**, 68 (1984) [Sov. Phys. JETP **59**, 38 (1984)].

[10]D. A. Harmin, Phys. Rev. A **26**, 2656 (1982), and **30**, 2413 (1984).