RYDBERG ATOMS IN MAGNETIC FIELDS – THE LANDAU LIMIT OF THE ATOMIC SPECTRUM

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We present some of the more striking experimental aspects of the quasi-Landau atomic spectrum in a magnetic field, observed near and above the zero field ionization threshold. The Landau limit is shown to be almost reached in our experiments on caesium, led at fixed electron energies scanning the magnetic field.

In spite of early attempts [1,2] and of numerous experiments on the scaled problem of excitons [3,4], the spectrum of hydrogen atoms in magnetic fields is far from being completely understood. Especially, no quantum theory presently allows a total interpretation of the experiments which reveal quite surprising and unexpected features of the problem of *diamagnetism in the atomic spectrum.* Experiments led in 1969 [5] to the discovery of a new signature of the atomic spectrum near the zero field atomic ionization threshold where resonances exist with a spacing of $\frac{3}{2}\hbar\omega_c$ (ω_c the cyclotron frequency). This is now well understood from semi-classical quantization conditions [6,7]. Indeed, the fundamental mechanisms of this strong field mixing [7] behaviour turn out to be general ones in the physics of non separable hamiltonian systems [8].

The basic features of the strong field mixing regime can be understood from arguments of classical physics. The atomic spectrum will be completely altered when the classical frequencies associated with the motion of the electron in the Coulomb and magnetic fields considered separately are of the same order of magnitude [9]. That is:

$$2R/n^3 \sim \hbar\omega_c \tag{1}$$

(R the Rydberg constant, $\omega_c = qB/m$ the cyclotron frequency) meaning that a new signature of the spectrum will exist for $n \sim (B/B_c)^{-1/3}$ where B_c is the critical field for which $\hbar\omega_c = 2R$. Far into the continuum, a Landau signature of the atomic spectrum must exist.

Such a very simple analysis, agreeing with experimental results for a part of the spectrum [10], has been recently more steadily established due to numerical studies of the classical motion of the electron [11–13]. They show that between the quasi pure Coulomb and Landau regimes, a third one takes place in which the classical motion of the electron may be chaotic or quasi-periodic following the initial conditions. This is, partly, a confirmation of the existence (in a restricted sense) of a dynamical symmetry in this problem [14,15]. Basically, the strong mixing regime is equivalent to what is called an Hill's region in the problem of two coupled anharmonic oscillators [16].

We here present some of the more striking aspects of a high resolution optical study of the atomic Landau spectrum, performed on highly hydrogenic odd parity $M = \pm 3$ states of caesium. Other aspects of this study, concerning the Coulomb limit [17] and Landau–Coulomb condensation of the atomic spectrum [10] have been previously reported. The optical excitation scheme uses the hybrid resonance phenomena [18], namely excitation of high lying atomic Rydberg states through molecular repulsive states of Cs_2 molecules. This is an efficient process at low pressures where Cs_2 densities are really negli-

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gible which produces lines having the Doppler width. Owing to the collisional mixing during the interaction, one photon excitation of the $5D_{3/2,5/2}$ states of caesium occurs. The absorption of a second photon allows the production of nF Rydberg states. The laser is a 10 MHz width, 800 mW power, c.w. dye laser in single mode operation and frequency controlled. Detection of Rydberg states or Landau resonances is achieved through the use of a shielded thermoionic detector [19] placed in the clear bore of a 8 T superconducting solenoid. This scheme has allowed the obtention in zero field of almost unperturbed Rydberg states up to 160 [10]. Furthermore, these F states are highly hydrogenic with quantum defects $\delta = 0.033$ which is of special interest in the magnetic field problem, as it provides with a good approximation to the ideal situation of the hydrogen. Additional details have been reported in refs. [10,17]. We will focuss hereafter on the *spectrum of resonances* connected with states of positive energies lying above the zero field atomic threshold. The intensity aspects will be discussed in a forthcoming paper.

The experiments have been done at fixed electron energies scanning the B field. This is nearly the experimental analog of making a group theoretical approach at the spectrum on the Fock hypersphere [20] under which condition structure of the theoretical problem seems simpler.

We limit the present report to the characteristics of the spectrum near and above threshold. Previous ones [10,17,21] have proved the discrete nature of the quasi Landau spectrum and shown the existence of a serie of dominant lines followed from the Coulomb to the Landau limits. These lines are associated with what has been called $K = 1$ states [14,15] with a wavefunction concentrated near the $Z = 0$ plane (perpendicular to the field). At low field these states are eigenvectors of the Coulomb problem – but not of the usual types – and their spectrum in field and energy are obeying the predictions of a two dimensional approximate WKB formula [11]:

$$\int_{\rho_1}^{\rho_2} [E_\perp - \gamma^2\rho^2/4 + 2/\rho - (|M| + \tfrac{1}{2})^2/\rho^2]^{1/2}\, d\rho = (N_r + \tfrac{1}{2})\pi, \qquad (2)$$

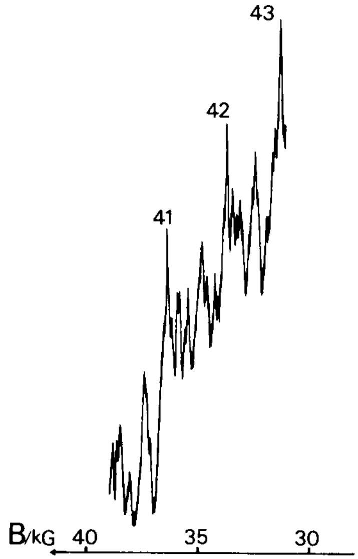

Fig. 1. Aspects of the quasi Landau atomic spectrum below threshold, at constant energy of the electron ($E = -12.6$ cm^{-1}), scanning the magnetic field. The dominant lines are associated with $K = 1$ states [15] with a wavefunction concentrated near the plane $Z \approx 0$ (perpendicular to the field), and their positions are agreeing approximately with predictions of formula (2).

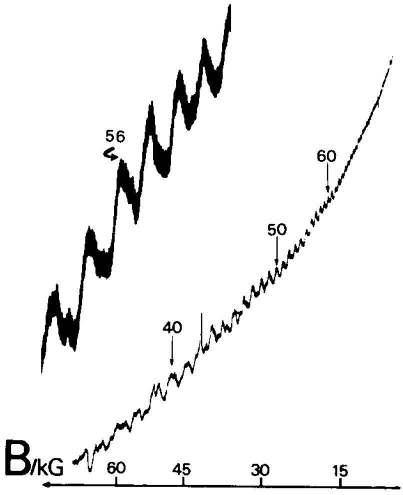

Fig. 2. Atomic Landau spectrum above threshold seen at constant energy of the electron ($E = +10$ cm^{-1}) as a function of the B field. The positions of the lines agree with predictions of formula (2). Some small substructures are still existing.

where N_r is the radial quantum number, $E_\perp$ is the transverse energy of the motion, $\gamma^2 \rho^2/4$ is the diamagnetic interaction in units of R, and M is the value of L_z.

Two plots of the spectrum in field, for energies below and above threshold are shown on figs. 1 and 2. Below threshold, lines are sharp with numerous "fine structure" components. Above threshold, broad but well defined lines still exist together with indications of the existence of a "fine structure". The width which is probably due to the autoionizing character of these resonances along the B field increases with the energy.

Fig. 3 plots the radial quantum number N_r as a function of $1/B$, for various values of the energy ranging from $E = 0$ to $E = 120$ cm^{-1}. At threshold, the experimental data follows a $N_r^3 B$ = const. quantization law in agreement with predictions from formula (1) or (2). This is associated with the strong field mixing regime [7,10]. The spacing in energy at a fixed field is then 1.52 $\hbar\omega_c$. Far above threshold, however, the experimental data follows a $N_r B$ = const. quantization law which is characteristic of the Landau regime. For $B \sim 20$ kG, $N_r \sim 90$ and $E = 100$ cm^{-1}, the spacing in energy is 1.1 $\hbar\omega_c$ indicating that the Landau regime is almost reached. In effect, it will be reached as an asymptotic limit, only, when $N_r \to \infty$. This seems paradoxical in that, for constant and positive values of the energy, the increase of the field will result in a departure from the $N_r B$ = const. quantization law obeyed in the Landau limit. Results, as shown on fig. 3, will always curve and tend towards the $N_r^3 B$ = const. quantization law obeyed at the threshold, whatever the energy value. This reveals the extreme conceptual importance of the strong field mixing regime and of the $N_r^3 B$ = const. quantization law.

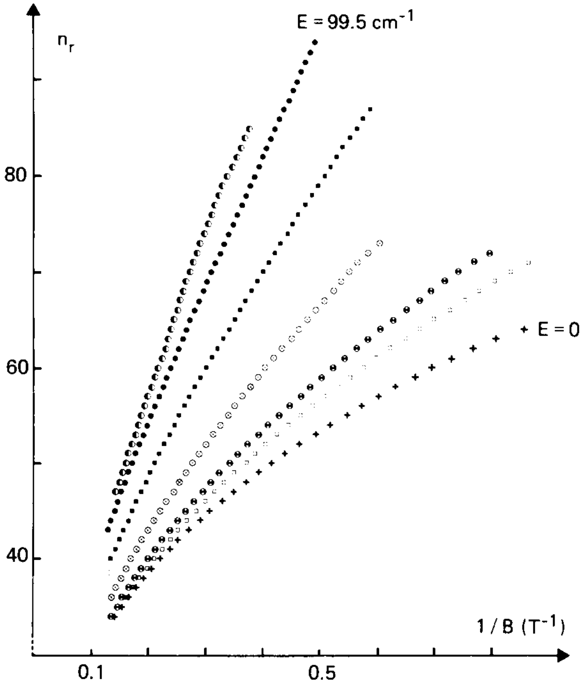

Fig. 3. Experimental plot of the radial quantum number as a function of $(1/B)$ for states with positive energies (in the atomic continuum). Each curve corresponds to a fixed energy value of the electron (E = 0, 11.5, 17.2, 37.3, 65.7, 99.5, 121.5 cm^{-1}). The results at $E = 0$ are obeying a $N_r^3 B$ = const. quantization law [10], while for $E = 99.5$ cm^{-1} and small field values they are aligned on a $N_r B$ = const. curve indicating that the Landau limit is reached.

Results in fig. 3 show the *first experimental observation of the Landau regime in the atomic spectra.* There is good agreement with predictions of formula (2) [10]. But this formula does not account for the spectrum in its whole complexity due to its two dimensional approximation of the real three dimensional non separable problem.

Let us stress upon the fact that the results prove that we really observe the spectrum of *electrons which are tightly bound to the ionic core.* If not, for free electrons, we would observe Landau levels of these free electrons. Then, no departure from the $N_r B$ = const. Landau quantization law would appear in a plot like that of fig. 3. Instead, the important curvature towards the $N_r^3 B$ = const. law at $E = 0$ is manifesting the increasing importance of the coulombic contribution to the total energy of the electron when the field is increased.

References

[1] F.A. Jenkins and E. Segre, Phys. Rev. 55 (1939) 52.
[2] L.I. Schiff and H. Snyder, Phys. Rev. 55 (1939) 59.
[3] J.G. Mavroides, Optical properties of solids, ed. F. Abelès (North Holland, 1972) p. 351.
[4] H. Hasegawa, Physics of solids in intense magnetic fields ed. E.D. Haidemenakis (Plenum, N.Y., 1969) p. 246.
[5] W.R.S. Garton and F.S. Tomkins, Astrophys. J. 158 (1969) 839.

[6] A.F. Starace, J. Phys. B 6 (1973) 585.
[7] A.R.P. Rau, Phys. Rev. A 16 (1977) 613.
[8] V.I. Arnold, Méthodes mathématiques de la mécanique classique (MIR, Moscou, 1976).
[9] J.C. Gay, Com. At. and Mol. Phys. 9 (1980) 97.
[10] J.C. Gay, D. Delande and F. Biraben, J. Phys. B 13 (1980) L729.
[11] M. Robnik, J. Phys. A 14 (1981) 3195.
[12] R.A. Pullen and A.R. Edmonds, J. Phys. A 14 (1981) L319.
[13] J.C. Gay and D. Delande, unpublished.
[14] M.L. Zimmerman, M.M. Kash and D. Kleppner, Phys. Rev. Lett. 45 (1980) 1092.
[15] D. Delande and J.C. Gay, Physics Letters 82A (1981) 399.
[16] E.J. Heller, E.B. Stechel and M.J. Davis, J. Chem. Phys. 73 (1980) 4720.
[17] D. Delande and J.C. Gay, Phys. Lett. 82A (1981) 399.
[18] C.B. Collins, B.W. Johnson, M.Y. Mirza, D. Popescu and I. Popescu, Phys. Rev. A 10 (1974) 813.
[19] L.R. Pendrill, D. Delande and J.C. Gay, J. Phys. B 12 (1979) L603.
[20] For example, B.R. Judd, Angular momentum theory for diatomic molecules (Academic Press, 1975).
[21] J.C. Castro, M.L. Zimmerman, R.G. Hulet and D. Kleppner, Phys. Rev. Lett. 45 (1980) 1780.