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EARLY BUILDING UP OF THE QUASI LANDAU SPECTRUM IN THE DIAMAGNETIC COULOMB REGION

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We report the results of experimental measurements on atomic diamagnetism in inter-l and inter-n mixing conditions. They have been obtained using high resolution techniques on highly hydrogenic $M = \pm 3$ states of cesium, providing a pure experimental situation. Comparison with straightforward hydrogenic calculations and semi-classical predictions allow identification of the dominant lines of the spectrum which are shown to behave as precursors of the quasi Landau spectrum.

In 1969, Garton and Tomkins [1] first demonstrated on barium, in high magnetic fields, the existence near threshold of a system of equally spaced broad resonances. But for the $\frac{3}{2}\hbar\omega_c$ value of the spacing, this was in agreement with earlier experiments on excitons for which quasi Landau condensation of the spectra was well known. The unusual value of the spacing near threshold was explained through semi-classical models assuming that the motion of the electron is confined in the $Z = 0$ plane [2,3].Though the gross features of the spectrum are now well understood, there is a lack of explanation concerning fundamental questions as

– the role of the electron's motion along the B field,

– connections between the Coulomb and Landau limits and finally

– properties of these states which require a quantum understanding of the problem.

The intermediate regime of motion, where the two classical frequencies $2R/n^3$ and $\hbar\omega_c$ are equal, which takes place near threshold, was called the strong field mixing regime and is certainly the prototype of a wide category of phenomena in physics, as pointed out by Rau [4].

Recent experiments on the quasi Landau spectrum near threshold have shown the existence of fine structure of the resonances, with chaotic character [5]. Unfortunately these experiments were done under poor conditions, viz. insufficient resolution, high pressures and on atomic states having strong departures from the hydrogenic behaviour. For various reasons close range corrections to the Coulomb potential are suspected to remove any generality to a study of this so called "fine structure" [6,7].

We present an experimental study performed with high resolution techniques on quasi-hydrogenic states of cesium. We focus here *on studies* of diamagnetism but extensive investigations are reported elsewhere showing that the quasi Landau phenomena affect well-resolved discrete lines we have been able to follow from the Coulomb to the Landau limit [7]. Owing to the highly hydrogenic situation we deal with, progress in the quantum understanding of the inter-l–inter-n mixing regimes of diamagnetism is achieved. We especially show that, in contrast to all previous experimental works, there are dominant lines in the spectrum *in inter-n mixing conditions*, and we identify them through quantum calculations in the low field limit. Their positions also approximately obey the semi-classical predictions and they are those exhibiting the strong field mixing behaviour near threshold. Then we partly give an answer to the fundamental question of the building up of the quantum spectrum.

Experimental set up. The basic features of the set up shown in fig. 1 is the use of a 500 to 1 W power, single mode, c.w. dye laser, 10 MHz width (R6G–Ar^+ pumped). The resolution is about 100 times greater than in previous pulsed dye laser experiments. The

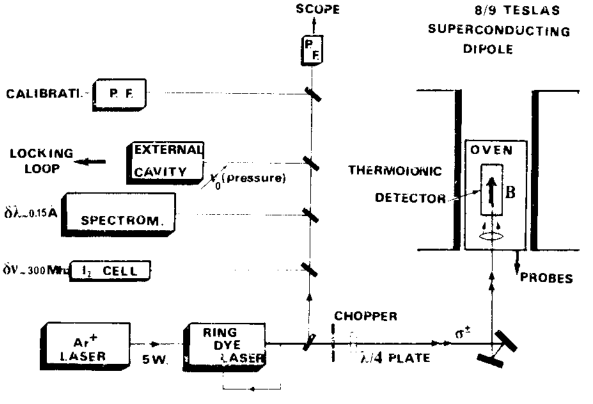

Fig. 1. Schematic view of the experimental set up.

laser frequency is locked on an external cavity allowing 100 GHz single mode pressure scan. Wavenumber calibration to ±0.01 cm^{-1} is obtained through comparison to the molecular iodine spectrum. Experiments are performed at pressures of 10^{-2} Torr (on the average 1000 times smaller than in other attempts). The laser beam is focussed into the equipotential volume of a pyrex electrostatically shielded thermoionic detector containing pure Cs vapour. The tungsten wire is heated to about 800 K and the voltage drop across it is 0.2 V. No additional electric fields are applied. The 8 T superconducting solenoid is built with Nb–Ti windings ensuring negligible hysteresis effects, no flux jumps and highly linear intensity scan.

Excitation scheme. P van F Rydberg states are excited from the 5 $^2D_{3/2,5/2}$ atomic levels which are populated by photodissociation of the Cs_2 molecules. This provides a more efficient way of producing weakly perturbed high lying Rydberg states at low pressures [7]. Stray electric field effects and collisional perturbations are almost negligible as is ascertained by the detection of Rydberg states as high as 162 [7].

Diamagnetism. The use of a molecular dissociation scheme does not allow any selection of the intermediate Zeeman substate of the 5D atom and may authorize simultaneous excitation of various M series at strong fields. Zeeman effects in the process are trivial as the fine structure of the 5D states is 99 cm^{-1} while it is negligible for nF states.

Studies of diamagnetism in inter-l mixing conditions *primarily* allow accurate identification and calibration of the spectrum. They show that the $M = \pm 3$ components of the lines are dominant whatever the field and electron's energy are, by at least one order of magnitude (see fig. 2). It is then a matter of fact to recognize the special suitability of the situation for studying the anticrossing behaviour and fine structure problem as the ($M = \pm 3$, odd-parity) states are almost hydrogenic ones with quantum defects smaller than 0.033. Theoretical agreement with a simple hydrogenic model must occur.

The diamagnetic line structure of the $5D_{5/2} \to$ $\to 28F_{5/2,7/2}$ transition has been studied in field and energy from the inter-l mixing regime to conditions where partial merging with $n = 29$ states occurs. Fig. 2 allows a display of the main features of the spectrum in a magnetic field, at fixed laser frequency. The dominant series in fig. 2 corresponds to $M = 3$ states of the hydrogenic manifold, excited from two different Zeeman sublevels of the $5D_{5/2}$ state. One of them ($M = 3, K$) extends over more than 40 kG. The value of K refers to previous classifications [6] and roughly corresponds to perturbed $l = 3, 5, \ldots$ odd-parity hydrogenic states of the $n = 28$ manifold. In addition, weaker lines exist which are classified in the same (M, K) scheme and are due to excitation of other series in the hybrid resonance process. In fig. 2, we only see $M = 1$ and $M = 2$ (odd-parity) series. The positions of all the components of the series agree fairly well with hydrogenic calculations including proper quantum defects for the implied states. The diamagnetic structure of the P state which is directly

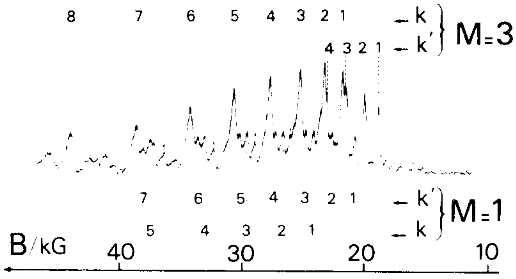

Fig. 2. Diamagnetic structure of the $5D_{5/2} \to n = 28$ (M, odd) transitions as recorded at fixed laser energy using the hybrid resonance pumping scheme. Two dominant series of $M = \pm 3$ states exist corresponding to excitation through the $m = 3/2$ and $m = 5/2$ Zeeman substates of $5D_{5/2}$. Two $M = +1$ series of lines of somewhat smaller intensities are also resolved. The arrows indicate the components of a series which corresponds to $M = +2$ states. The positions of these lines agree with hydrogenic calculations including quantum defects.

populated through the hybrid resonance schemes is too weak to be seen on the record. The efficiency of optical excitation of the P states from the 5D levels is about 40 times smaller than that of the F state. Obviously, the excitation of $M = \pm 3$ lines is about one order of magnitude more efficient than that of other lines. In addition, there is no evidence in the pattern of lines corresponding to parity breaking associated with motional Stark field effects. They are then, at least, one order of magnitude smaller than the ($M = +3$) main ones. Our conditions are then suitable for a study of diamagnetism on $M = +3$ and -3 quasihydrogenic states, the relative positions of which just differ through obvious paramagnetic terms.

In fig. 3, we have plotted the results of systematic measurements performed on the ($M = \pm 3$, K, odd) diamagnetic series as a function of the field B, for $n = 28$. For $B \leqslant 10$ kG, the phenomena are well known and the positions of the lines vary as expected, with departures from the B^2 law due to the small quantum defects ($\delta = 0.033$) of the F state. Fairly good agreement exists with the hydrogenic calculations in the $n = 28$ manifold (including the quantum defect of the F state) for intermediate field values. At higher fields ($B \approx 35$ kG) the results curve again. This is due to the mixing with the components of the other manifolds, the lower components of the $n = 29$ being close to the $K = 1$ component of the $n = 28$ manifold. Inclusion of 6 manifolds around $n = 28$ in the calculations allows one to restore very good agreement with the experimental results, although the accuracy of the predictions cannot be perfect (due to the neglect of an infinity of configurations).

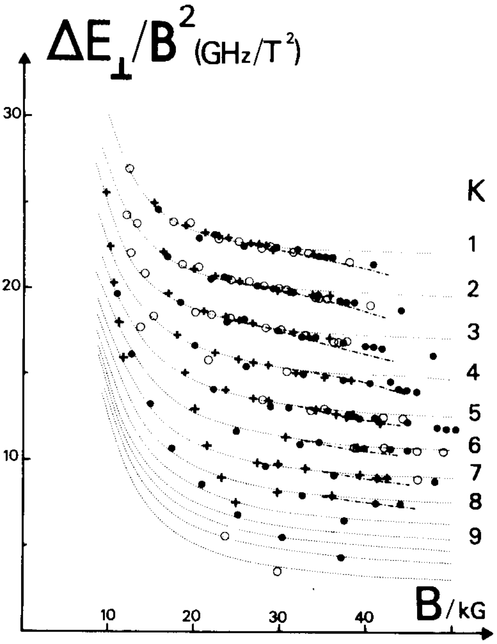

Fig. 3. Diamagnetic structure of the $n = 28$ ($M = \pm 3$, odd-parity) states as a function of B. $\Delta E_\perp$ is the energy of the components reduced to the $n = 28$ F position in zero field and corrected for paramagnetic terms. $\Delta E_\perp$ just represents the diamagnetic shifts on the components and $\Delta E_\perp / B^2$ would be constant in the perturbation regime for 0 quantum defects. Experimental points (crosses, circles and points) are in agreement with inter-l mixing calculations (dotted lines) including quantum defects of the F state. The agreement is even better with calculations including 6 hydrogenic manifolds around $n = 28$ for fixed values between 35 and 50 kG, illustrating the role of the other manifolds (dash–dotted lines).

The presence of two ($M = \pm 3$) dominant series will be a source of strong complications *in inter-n mixing* conditions. We have then used the $5D_{3/2} \rightarrow n$F ($M = \pm 3$, odd) series for studying accurately the formation of the quasi Landau spectrum though the efficiency of production is 10 times smaller. A typical spectrum, in inter-n mixing conditions, is shown in fig. 4, at fixed laser energy corresponding to the Coulomb regime with $n \approx 50$, in a 7 T range of the field. At low fields, in inter-l mixing conditions, the characteristic diamagnetic structure of the states is easily recognized exhibiting in some situations [7] more than 14 components which corresponds to perturbed hydrogenic states with $l = 29$. At higher fields, in fig. 4, components be-

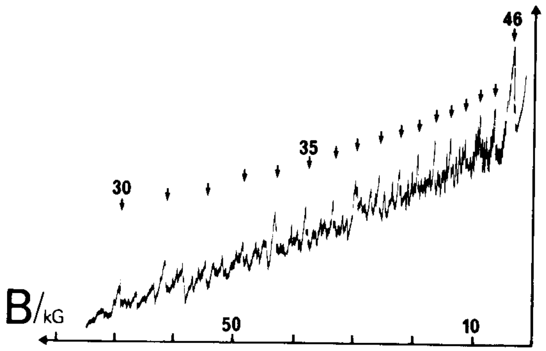

Fig. 4. Inter-n mixing regime as a function of the field B. The laser frequency is fixed to a value corresponding to the $n \approx 50$ Coulomb region. Though merging of the various manifolds is almost complete, dominant lines associated with $K = 1$ states at low fields still exist in the spectrum and are indicated by arrows.

come entangled but dominant lines still exist, corresponding to $K = 1$ states of the successive manifolds, the positions of which agree both with predictions of two-dimensional semi-classical calculations [7] and with quantum inter-n mixing calculations for the lower field values. This is not surprising in the light of the discussions in ref. [6] where it is shown that $K = 1$ states are almost localized in the (xy) plane. But the main problem is that experiments are far more accurate than quantum calculations. As an example fig. 5 shows a comparison of measurements for $n = 50$ and $n = 49$ of the positions of the $(n, K = 1$ to $14)$ lines with quantum calculations including several manifolds. Discrepancies of several hundredths Gauss exist between the measurements and predictions near the crossings but these are lying inside the *probable uncertainty due to the neglect of other configurations* in the calculations. The accuracy of the experiments is better than 1 GHz which is also confirmed by low field measurements of the positions of the lines in inter-l mixing conditions. Of course, measurement of the size of the anticrossing of $(49, K = 1)$ with $(50, K)$ is not possible as the predicted size is less than 1 MHz, and essentially due to the small quantum defects [8] of F states.

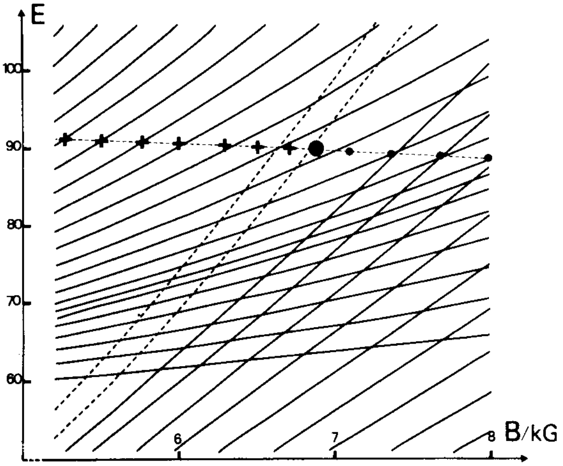

Fig. 5. Energy diagram as a function of the field for the anticrossing region between the $n = 49$ and $n = 50$ manifolds. Theoretical curves have been obtained through diagonalization (including quantum defects of the F state) of the diamagnetic hamiltonian on basis sets including all the $l \geqslant 3$, odd states for various numbers of manifolds. Full curves are from calculations including the $n = 49$ to 51 manifolds. Dashed lines represent calculations for $(n = 49; K = 1, 2)$ with larger $n = 47$ to 52 basis set. Crosses and points are experimental results. Crosses are the $K = 8$ to $K = 14$ components of $n = 50$, respectively. The large point represents the $(n = 49, K = 1)$ position and the smaller points the components $(n = 49, K = 2 \rightarrow 5)$. The thin dashed line corresponds to fixed laser energy in the energy diagram. Discrepancies exist between the experimental positions of the $(n = 49, K = 1, 2, ...)$ states and the predictions. The relative spacing of the K components in the field is in agreement with simple predictions.

Approximate semi-classical calculations for $Z \approx 0$ suffer from various shortcomings [7] but are still of interest in view of the lack of more refined techniques. They are in rough agreement for $(n = 49, K = 1)$ with experimental results. Whatever the electron's energy, the position of the dominant lines agrees well with the semi-classical predictions as is manifest in fig. 6. Discrepancies as pointed out in ref. [7] cannot be considered as meaningful since the "two-dimensional" quasi-classical solution is really a crude approach neglecting motion along the B field and effects due to non-separability. The systematic underestimation of the positions of lines at high fields, equivalently overestimation of the energies at fixed fields illustrates the limitations of this model. n_r is just the radial quantum number associated with semi-classical quantization of the radial

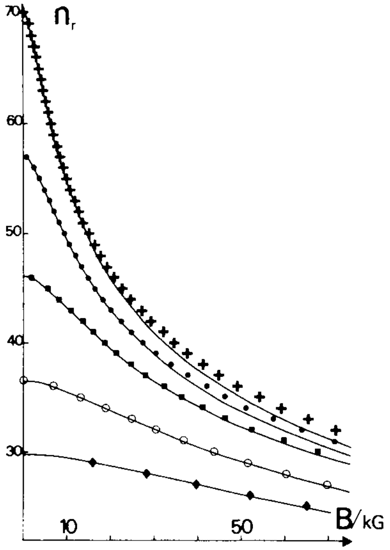

Fig. 6. Plots of the radial quantum number $(n_r = n - |M| - 1$ in the Coulomb $B = 0$ region) as a function of B at fixed laser energy (below threshold). Full curves are from semi-classical calculations while points refer to the dominant lines of the spectrum. The assignment of the Coulomb quantum number is absolute. Good overall agreement exists in spite of systematic deviations which may be a manifestation of the limitations of the two-dimensional semi-classical approach [7].

momentum in cylindrical coordinates for $Z = 0$, and just $n - |M| - 1$ in the Coulomb limit [7].

Those lines which coincide at low fields with $K = 1$ lines are in the Coulomb region the earlier manifestation of the quasi Landau phenomena which can be followed up to more than 3000 GHz above the threshold [7].

The present interpretation involves only a part of the dominant discrete spectrum, associated with states having the strongest oscillator strengths in the optical excitation process. Other series (weakly interacting with the previous one if a hidden dynamical symmetry exists), connected with other K values in the low field limit will contribute to the spectrum near threshold, with chaotic character. Though there are strong inferences from experimental results and from three-dimensional semi-classical predictions, this is not clearly established. But it is now clear that the *strong field mixing phenomenon* involves series of discrete lines, when experimental conditions are convenient, and that a better understanding of the problem must be sought in the inter-n mixing regime where, really, things are building up.

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