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LETTER TO THE EDITOR

Landau condensation of the quasi-hydrogenic spectrum of caesium

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Abstract. Landau condensation of the $M = \pm 3$, odd-parity, quasi-hydrogenic caesium spectra is investigated for fields up to 8 T and for energies ranging $\pm 120\ \mathrm{cm}^{-1}$ around the threshold. Continuous single-mode dye laser excitation is used. A dominant series of sharp discrete lines is shown to follow the semiclassical predictions from the Landau to the diamagnetic Coulomb regimes where the lines branch to well defined states of the hydrogenic manifolds. For this highly hydrogenic spectrum, the spacing of the resonances near the threshold is shown to be $1{\cdot}5\ \hbar\omega_c$.

Renewed attention has been paid to the strongly magnetised hydrogen atom problem (Garstang 1977, Gay 1980, Rau 1980) owing to impressive experimental developments in the physics of Rydberg states and to the topical nature of the problem (Fano 1977, 1980).

Experimental investigations on hydrogen atoms would be of obvious interest to stimulate a quantum theoretical understanding but, unfortunately, are not yet possible. For other atoms, the existence of short-range corrections to the Coulomb potential, although they do not alter the basic character of the phenomena (Rau 1977), hinders a detailed analysis of the experimental data. In particular, they remove any generality from a study of the so-called 'fine structure' of the Landau spectrum as is manifest for Ba, Sr (Fonck *et al* 1978, Lu *et al* 1978b) and Rb (Economou *et al* 1979), and mask possible effects of approximate dynamical symmetries (Zimmerman *et al* 1978). Only one experiment has been performed on quasi-hydrogenic states: for Li, with quantum defects smaller than 0·05, the motional Stark field perpendicular to $\boldsymbol{B}$ alters to $\frac{1}{2}\hbar\omega_c$ (where ω_c is the cyclotron frequency) the signature of the quasi-Landau spectrum (Lu *et al* 1978a).

Here, we report an experimental study for magnetic fields up to 8 T and electron energies ranging $\pm 120\ \mathrm{cm}^{-1}$ around the threshold of Landau–Coulomb condensation, of the $M = \pm 3$, odd-parity, quasi-hydrogenic states of caesium. The quantum defects of all the diamagnetically mixed states are less than 0·033 (i.e. the states are closely hydrogenic) and in addition, we have demonstrated experimentally that motional Stark effects are weak.

For the first time, we have traced the complete evolution of the Landau–Coulomb spectrum as a function of the magnetic field. Moreover, in contrast to previous attempts, all our measurements have been performed on fully resolved lines of the

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spectrum and not on broad modulations. The data which are interpreted here and shown to follow the approximate two-dimensional semiclassical theory from the diamagnetic Coulomb to the Landau limit correspond to the dominant lines of the spectrum. They are in the *diamagnetic inter l mixing regime* those which rise fastest in energy with increasing B, namely those which retain the major part of the (n, 1 = 3, $M = \pm 3$) hydrogenic wavefunction at low fields. Our conclusions which are supported both by inferences from numerical calculations and by one other experimental study are of a nature liable to renew our current understanding of the phenomenon, thus opening the way to a full quantum approach to the problem. They have been made possible due to the use of a simple and pure experimental situation and by the use of high-resolution optical techniques.

The optical excitation of the spectrum is performed by using a CW 500 mW power, 10 MHz width, single-mode dye laser (R6G–Ar^+ pumped ring dye laser). Our resolution is at least 100 times greater than in previous pulsed dye laser experiments. We can therefore resolve the so-called 'fine structure' of the resonances and extend the range of investigations to much higher n values. Experiments are performed at pressures of 10^{-2} Torr, 1000 times smaller than in previous ones (Fonck *et al* 1978, Lu *et al* 1978a, b, Economou *et al* 1979).

The studies were performed at a fixed electron energy (discarding obvious paramagnetic terms) by scanning the magnetic field. This aspect of the Landau spectrum has so far attracted only indirect and limited investigations (Fonck *et al* 1980). In principle, scanning B provides an additional experimental check of the correctness of conclusions (Fonck *et al* 1978, 1980, Economou *et al* 1979) based on the two-dimensional semiclassical approximation for the non-separable problem.

Our apparatus has been previously described by Pendrill *et al* (1979). The laser beam is focused into the equipotential volume of a pyrex electrostatically shielded thermoionic detector (Harvey and Stoïcheff 1977) containing pure Cs vapour at 170 °C (10^{-2} Torr pressure) without any buffer gas. The tungsten wire is heated at about 800 K and the drop voltage 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 a highly linear intensity scan. Field calibration has been previously reported by Gay and Schneider (1976). The laser frequency is locked on an external cavity allowing 100 GHz single-mode pressure scan. Wavenumber calibration to $\pm 0{\cdot}01\ cm^{-1}$ is obtained by comparison with the molecular iodine spectrum (Gerstenkorn and Luc 1976).

P and 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. Although the mechanisms (hybrid resonances, Collins *et al* 1974) are not completely elucidated, efficient production of *weakly perturbed* high-lying Rydberg states occurs at low pressures. Presently, stray electric field effects and collisional perturbations are almost negligible as ascertained by the detection of quasi-unperturbed F states with $n \sim 162$. A typical single mode, 30 GHz frequency scan of the Rydberg spectrum, is shown in figure 1. The absolute red shift of the $n = 140$ state is 200 MHz and the width of the line is about 1 GHz in agreement with the estimated velocity of the atoms issued from the exoenergetic molecular dissociation process.

The use of molecular dissociation as the experimental scheme does not allow any selection of the intermediate Zeeman substate of the 5D atom and may allow simultaneous excitation of various M series in strong fields. Studies of diamagnetism in inter l mixing conditions allows identification and calibration of the field spectra. In

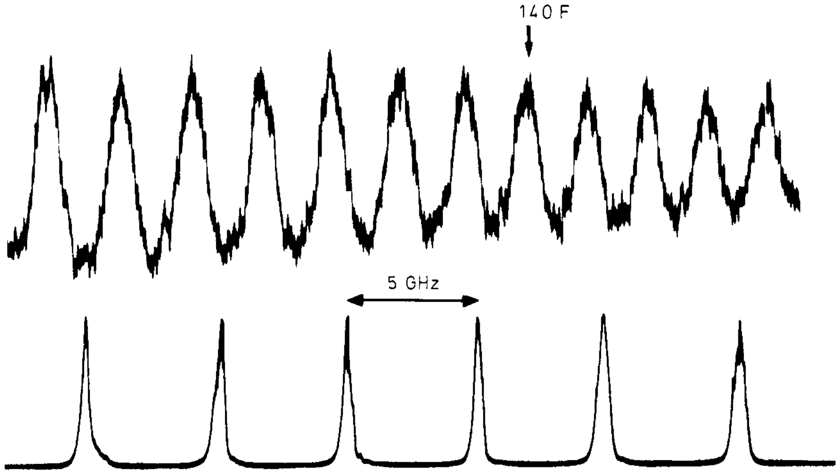

Figure 1. 30 GHz single-mode frequency scan of the nF Rydberg spectrum from $n = 133$ to $n = 144$ against the Airy function of a 5 GHz calibrated PF interferometer.

particular, the $M = \pm 3$ components of the F line are found to be at least one order of magnitude stronger than the others. They provide us with a simple quasi-hydrogenic situation where quantum defects of all the diamagnetically mixed states are smaller than 0·033, which is of interest for studying under its purest form the so-called 'fine structure' (Fonck *et al* 1978, Lu *et al* 1978a, b) of Landau resonances and the quasi-crossing behaviour in the inter n mixing region (Zimmerman *et al* 1978). The diamagnetic line structure of the ($M = \pm 3$, odd) 28F state agrees within 300 MHz on a 4 T range of the magnetic field with the predictions obtained through a straightforward diagonalisation of the diamagnetic Hamiltonian in a hydrogenic basis including the quantum defect for the F state. In the diamagnetic patterns, there is no evidence of lines due to parity breaking (figure 2). Motional Stark effects are then weak but clues to the existence of the $\omega_c/2$ mode seem to exist (Lu and Rau 1980). At the end, LS decoupling effects are negligible in the 5D state.

Quasi-Landau condensation of Cs quasi-hydrogenic spectra has been studied for magnetic fields between 0 and 8 T, at fixed energies ranging from the $n = 40$ Coulomb region to 120 cm^{-1} above the ionisation threshold. Below the threshold, the Landau–Coulomb spectrum is composed of numerous sharp lines. While scanning the field at a fixed laser energy, it presents three successive regimes. At low fields, the well understood inter l diamagnetic regime occurs where the wavefunctions $|n, K, M, \pi\rangle$ are eigenstates of the restriction of the diamagnetic Hamiltonian to the n hydrogenic manifold. K is just a label. At low fields, $K = 1$ retains the major part of the $l = 3$ hydrogenic wavefunction and the associated energy exhibits the fastest increase with the magnetic field B inside the manifold. At higher fields, the inter n mixing regime occurs with an apparently total merging of the various manifolds. In contrast to all previous experimental situations (Garton and Tomkins 1969, Fonck *et al* 1978, Economou *et al* 1979), *dominant lines exist in our data* the positions of which are correlated to the $K = 1$ lines of the inter l mixing regime at low fields. We believe that this aspect does not emerge from other experiments because of spurious effects of the short-range corrections to the potential on the efficiency of optical excitation. At higher fields and near threshold (figure 3), pronounced sharp edges appear which correlate

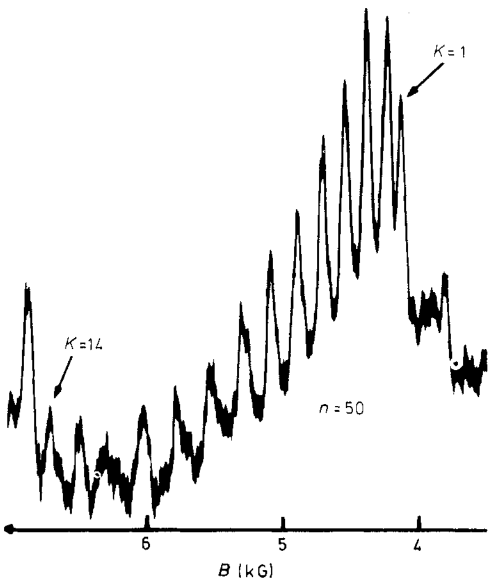

Figure 2. Typical record against magnetic field of the diamagnetic structure of the (n = 50, M = +3, odd-parity) states of caesium, exhibiting 14 (odd-parity) components of the (perturbed) hydrogenic manifold.

with the dominant lines mentioned above. In addition, there is a large number of sharp lines which simulate a 'fine structure' but may be the result of the superposition of several weakly interacting 'Landau series' with slightly different characteristics. As an indication, additional edges appear at fields greater than 50 kG. The lines and edges have very small widths in the magnetic field as expected from semiclassical arguments since dE/dB is proportional to the radial quantum number n_r. The width increases with B with a maximum value of about 1 kG. All the measurements reported have been done on the dominant discrete lines of the spectra (labelled $K = 1$).

Above threshold, the Landau spectrum is somewhat simpler and free from sharp resonances. Indications of the existence of more than one series exist. The lines become broader and broader for increasing energies of the electron and, at the highest energies, are a smooth modulation of the diode current (cf figure 3).

In the absence of a more reliable theoretical approach, which requires the use of EBK quantisation (Maslov 1972, Percival 1977) or a realistic three-dimensional model (Fano 1980), approximate predictions for this non-separable problem are obtained through adapted Bohr–Sommerfeld quantisation, namely

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

where $E_\perp$ is the electronic energy including paramagnetic terms and $\gamma = \hbar\omega_c/2R = B/B_c$ ($B_c = 2{\cdot}35 \times 10^5$ T). The rotational energy expression is chosen for (1) having the right low-field diamagnetic Coulomb behaviour for the $K = 1$ line, with $\alpha = \frac{1}{2}$. But in fact, α is likely to vary with E and B as it must be $\frac{1}{4}$ in the Landau limit. Equation (1) does not allow any absolute calibration of the spectrum. Experimental assignments of the n_r value are absolute in the Coulomb limit for energies lower than $-20\ \mathrm{cm}^{-1}$. Otherwise they are performed at low fields through (1). Usually the agreement with

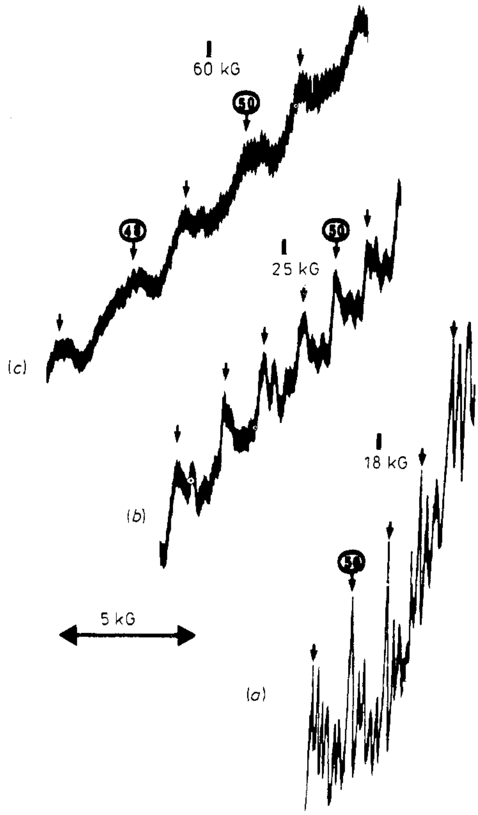

Figure 3. Aspects of the Landau spectrum below and above the ionisation threshold for $n_r \sim 50$ and energies (*a*) $E = -12{\cdot}56\ \mathrm{cm}^{-1}$, (*b*) $E = 0$, (*c*) $E = +99{\cdot}53\ \mathrm{cm}^{-1}$. Arrows indicate successive members of the series.

'theory' is then excellent over more than thirty levels and field variations of 6 T. The overall agreement shown in figure 4 is satisfying although some discrepancies persist in the Coulomb region. This possibly illustrates the limitations of (1) (variations of α with B) and deviations from the two-dimensional planar motion of the electron between 0 and 8 T. Energies in (1) will then be overestimated and energy levels will appear at higher field values. The effects of variations of α with B will be less important in the continuum as the Coulomb character is fainter, though the choice of $\alpha = \frac{1}{2}$ is obviously no longer appropriate.

The energy levels close to the threshold follow a $B^{-1/3}$ linear law as predicted through (1) and various classical models (O'Connell 1974, Rau 1977, Gay 1980) over a wide range of B. For negative and positive energies of the electron the $n_r(B)$ results curve away in opposite directions towards the Coulomb and Landau limits.

From our results, the appearance of the energy spectrum at fixed magnetic field can be deduced. The spacing of the resonances continuously decreases from the Coulomb to the Landau regime which is almost reached for levels at $E \simeq 100\ \mathrm{cm}^{-1}$, the spacing being $1{\cdot}1\ \hbar\omega_c$ for $B = 20$ kG. The spacing of the resonances near threshold is about $1{\cdot}5\ \hbar\omega_c$. For these highly hydrogenic $M = \pm 3$ states of caesium, the spacing of the resonances is just the same as for Sr, Ba and Rb, as expected. As concerns the *coarse*

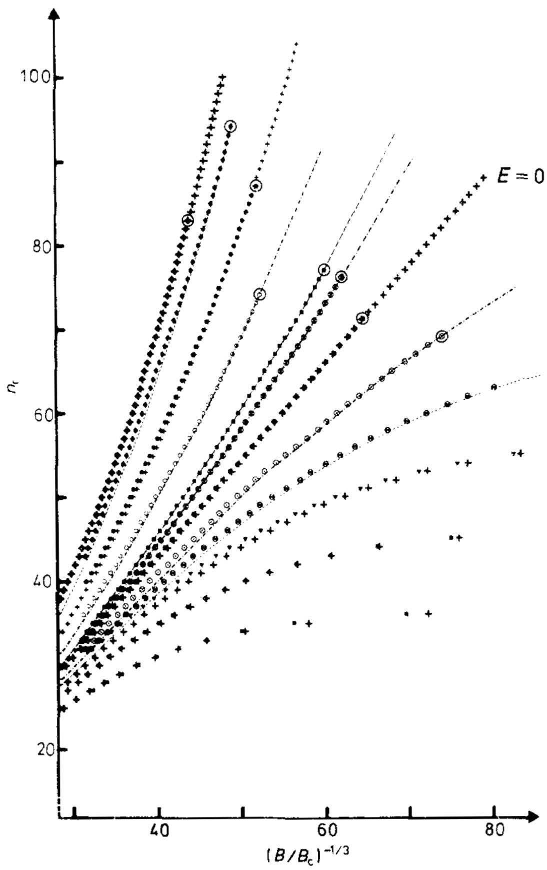

Figure 4. Experimental and theoretical plots of the radial quantum number against $(B/B_c)^{-1/3}$ at various energies for the dominant discrete series of the spectrum. Theoretical points (crosses or broken curves) are from formula (1) with $\alpha = \frac{1}{2}$. Experimental points are for energies 121·5 (top line), 99·5, 65·7, 37·3, 17·2, 11·5, 0, −10·9, −20, −29·3, −43·6, −66·6 cm^{-1} (lower line) respectively from the ionisation limit. Circles indicate assignment points when used. At the threshold the curve is a straight line. For positive and negative energies of the electron, the results curve in opposite directions towards the Landau and Coulomb regions.

features of the spectrum, there is no special behaviour associated with the hydrogenic character. Our results, therefore, confirm experimentally that the $\frac{1}{2}\hbar\omega_c$ spacing observed in the Li experiment (Lu *et al* 1978a, b) is due to strong motional Stark effects on the quasi-hydrogenic structure, as deduced theoretically (Crosswhite *et al* 1979, Rau 1979, Gay *et al* 1979).

However, the key feature of the data of figure 4 is that they have been obtained on *well defined sharp lines* of the spectrum *followed* from the Landau to the Coulomb limit where they branch to '$K = 1$' lines of the diamagnetic manifold and not on broad unresolved modulations. This is the first experimental proof of the existence of a set of discrete levels obeying the semiclassical quantisation conditions and exhibiting the strong-field mixing behaviour. Finally, our conclusions strongly suggest that the weaker lines of the spectrum can be classified under an analogous scheme, thus explaining the so-called 'fine structure' with chaotic character as the result of a superposition of several quasi-Landau series of slightly different characteristics and connected with different atomic states in the Coulomb limit.

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