ON A POSSIBLE DYNAMICAL SYMMETRY IN DIAMAGNETISM

D. DELANDE and J.C. GAY

Laboratoire de Spectroscopie Hertzienne [1] *de l'Ecole Normale Supérieure, 75230 Paris Cedex 05, France*

Received 3 November 1980
Revised manuscript received 11 February 1981

Simple theoretical calculations on atomic diamagnetism are presented and compared with heuristic arguments. The existence of exponentially small anticrossings of energy levels in the B field diagram as pointed out recently is confirmed, and tentatively discussed in terms of general theorems of classical mechanics. Conjectures on conservation of nodal surfaces between the low and high field limits are shown to violate our results in the low field Coulomb limit.

The strongly magnetized hydrogen problem is the subject of many conjectures attempting to obtain a better understanding of the energy diagram and the physics of this non-separable problem. But a synthetic view as provided e.g. by a correlation diagram in the B field between the Coulomb and Landau regions is far from being obtained.

Recently, the low field Coulomb region has been shown to be of major interest, as precursors of the quasi Landau regime are existing in high resolution spectra [1–3]. We here present and discuss the results of quantum calculations performed in the so-called inter-l and inter-n mixing regimes, the main purpose of which was the interpretation of our experimental data [4], obtained in high resolution conditions, on highly hydrogenic states of cesium. The evidence of quasi crossing behaviour for *a part of the diamagnetic spectrum* is emphasized, thus confirming the results of recent work [5]. The striking, exponentially small size of these anticrossings for purely Coulomb potentials, which has been conjectured as a proof of the existence of an approximate dynamical symmetry [5], is tentatively compared with some situations arising in classical mechanics for a non-separable hamiltonian.

The non-separable hamiltonian of the problem is:

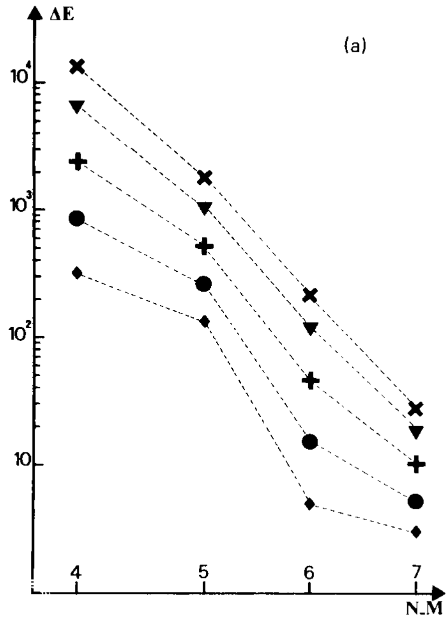
$$H = H_0 + H_D ,$$

where $H_D = \frac{1}{8}\gamma^2 r^2 \sin^2\theta$ is the diamagnetic term in reduced units (with $\gamma = \hbar\omega_c/2R$, ω_c the cyclotron frequency, R the Rydberg constant) responsible for quasi Landau condensation of the Rydberg spectrum near and above threshold. H_0 includes the Coulomb and paramagnetic terms.

The basic feature of the calculations is to diagonalize the diamagnetic hamiltonian on a hydrogenic basis, L_z and parity being constants of the motion. In the actual lack of an approximate theoretical separation of the problem, this is the more straightforward quantum approach at low fields. This has been performed both in spherical and parabolic coordinates allowing an additional check of the accuracy of the calculations (the usual criteria being the obvious one of stability after successive iterations). Calculations have been done for the lowest hydrogenic states, for various M and parity values, as shown in fig. 1, and for $M = \pm 3$ odd-parity states for higher n values, in order to compare to experimental results.

In inter-l mixing conditions, $2R/n^3 \gg H_D$ or $\gamma^2 n^7 \ll 1$ and the diagonalization is performed inside the n hydrogenic manifold. This provides a set of eigenvalues $E^{nK}(M, \Pi)$ and wavefunctions $\Psi^{nK}(M, \Pi)$. K is just a label. The $K = 1$ state retains the dominant part of the nF wavefunction and E^{n1} has the fastest increase with B field once the Zeeman effect is overcome. In this regime, E^{nK} varies as B^2 (if quantum defects are neglected). The wavefunctions Ψ^{nK} do

[1] Associé au CNRS, LA 18.



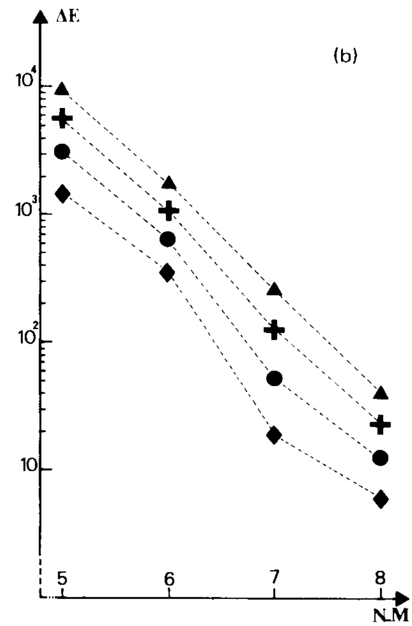


Fig. 1. Sizes of the anticrossings (in GHz) between $(n, K = 1)$ and $(n + 1, K_{max})$ for the lowest energy levels, in logarithmic coordinates, as a function of $(n - M)$. The sizes are almost varying as e^{-2n} with n. The two systems of curves correspond to states having (a) even parity along the Z axis, (b) odd parity along the Z axis, respectively. Each broken curve corresponds to a fixed M value which is, from the upper to the lower one, (a) $M = 0$ to $M = 4$, (b) $M = 1$ to $M = 4$, respectively. For $M = 4$, even parity along Z, the first anticrossing between $(n, K = 1)$ and $(n + 1, K_{max} = 2)$ occurs for $n = 8$ at a magnetic field strength of 588 T while for $(M = 4$, even, $n = 12)$ it is 144 T.

not depend on B and are eigenstates of H_0, for the Coulomb problem in zero field. They are also eigenstates of H when B tends to 0 (but is non-zero, then definitively breaking the zero-field supersymmetry of the Coulomb problem). Surprisingly, the characteristics of such states which are of major importance for the present problem have never been studied.

In the inter-n mixing regime, $2R/n^3 \sim H_D$ $(\gamma^2 n^7 \approx 1)$ and n is no longer a good quantum number. Diagonalization of H_D must be performed on a wider set of hydrogenic states including several manifolds. The sizes of the bases which are used in this regime are up to 500 states, following the stability of successive iterations. Nevertheless, the accuracy of calculations can never be perfect as one neglects the role of continua. This is also the problem in the calculations of ref. [5]. The use of a sturmian approach [3] is certainly the only way of overcoming these difficulties. In this regime, the eigenvalues are $E^{\widetilde{nK}}(M, \Pi)$ and the eigenstates $\Psi^{\widetilde{nK}}(M, \Pi)$ which adiabatically branch to the $\Psi^{nK}(M, \Pi)$ states when $2R/n^3 \gg H_D$. A typical plot of the energy diagram is shown in fig. 5 of ref. [4]. It has been obtained including several hydrogenic manifolds around $n = 50$.

States as Ψ^{n1} and $\Psi^{n'1}$ are strongly interacting with each other in the inter-n mixing regime, while Ψ^{n1} and $\Psi^{n'K \neq 1}$ have usually weak interactions. In this later case, the sizes of the anticrossings are usually exponentially small, varying approximately as e^{-2n} with the n value (fig. 1). This means almost crossing behaviour with the approximate rule that the more parallel the behaviour of the energies with B, the stronger the anticrossings. Practically, the anticrossing sizes for $K = 1$ states are always small compared to $2R/n^3$ and smaller than 1 MHz for $n = 50$. The class of states with $K = 1$ seems to behave almost independently of the other ones for high n values. They also retain most of the optically excited $(l = 3, M = \pm 3)$ hydrogenic state and will have dominant character *in experimental investigations with one-photon absorption* from $l = 2$ atomic levels [2,4]. This class of $K = 1$ states seems to have major importance for the formation of the quasi Landau spectrum as inferred from experimental data [1,2] and numerical simulations [3]. Then, this justifies why strong interactions between Ψ^{n1} and $\Psi^{n'1}$ states must exist. The variations of their energies with B must pass from B^2 in the Coulomb limit to B in the Landau one. The other series with $K \neq 1$, for which optical excitation is less efficient, having almost crossing behaviour with the $K = 1$ series, may contribute to the formation of other Landau series of slightly different characteristics. Mixing of them will result in the so-called fine structure with chaotic character.

Close range corrections to the Coulomb potential will not affect the gross features of the energy diagram [6,7] in practical experimental conditions. As shown in ref. [4] where quantum defects of $\delta = 0.033$ for the F states are included, the quasi crossing behaviour will not disappear. Though they are no longer exponentially small, the anticrossings are still small compared to $2R/n^3$ and almost impossible to measure for high n values [5,8]. Nevertheless, if the existence of exponentially small anticrossings is really connected with the existence of an approximate dynamical symmetry, close range corrections to the Coulomb potential could be important in the sense that they may break the possible hidden symmetry. In addition, departures from hydrogenic behaviour are surely of importance as concerns *the oscillator strengths of the recorded experimental spectrum*, when optical excitation is used. This is the main reason for which no dominant structures exist in the experimental results [9,10], in inter-n mixing conditions but for those obtained on highly hydrogenic states [2,3]. Obviously, they also completely modify the appearance of the spectrum in the inter-l mixing region [1].

Nodal surfaces and conservation laws. Nodal surfaces of $\Psi^{nK}(M, \Pi)$ in the inter-l mixing regime are shown in fig. 2 for $n = 17$, $M = 3$, odd-parity states. From the zero field and ultra high field limits [11], one can imagine how the deformations occur, due to merging of cones and spheres, or planes and cylinders. The *inadequacy of conjectures* of Kleiner [12] and of ref. [13] based on conservation of nodal surfaces between the low and high field limits is exemplified in fig. 2. A similar feature has already been reported in ref. [11], but in ultra high field conditions ($R \ll \hbar\omega_c$) when the adiabatic separation of the longitudinal and transverse motions is almost valid. These conditions are far from being valid in the Rydberg atomic physics conditions we presently investigate. But the two approaches are a contribution to the same problem, with identical conclusions, though they are obtained in completely different ways. Our contribution is the first one to the important problem of symmetries of the wave functions in the low field limit, for quasi-hydrogenic atoms.

$K = 1$ states seem to be associated to excited motion along the B direction as may be inferred counting the nodes along B. But for values greater than 3×10^{-5}, $\Psi^{K=1}$ is concentrated in the shaded area meaning that it really corresponds to *low excitation out of the $Z = 0$ plane*. This is in agreement with the basic hypothesis of semi-classical calculations of the Landau spectrum [7]. It is no longer surprising that experimental positions, quantum inter-n mixing calculations, and two-dimensional semi-classical ones assuming $Z \approx 0$ for the $(n\widetilde{K} = 1)$ lines are in agreement. These lines are really low field precursors of the quasi Landau spectrum [1–3].

$K = 7$ states, from the nodes, seem to correspond to low excitation in the B direction for $\rho \approx 0$ and excited motion for $\rho \sim n^2 a_0$ (a_0 the Bohr radius). Once again, the wave function for $|\Psi| > 3 \times 10^{-5}$ is concentrated in the shaded area and then corresponds to excited motion along B.

Obvious features of the inter-n mixing regime can be deduced from fig. 2. Interaction between nK and $n'K$ states will be strong as matrix elements of ρ^2 between states of the same general symmetry will be non-zero. In contrast, interactions between $K = 1$ and $K = 7$ states will obviously be weak. Optical excitation of $K = 1$ states in σ polarization will also be more efficient. Additionally, it is clear that no a priori choice of a basis having convenient symmetries is possible; especially choice of spheroidal functions [14] is not better than others *in this regime.*

It appears that the strange distribution of nodal surfaces gives a distorted representation of the real problem, especially contradicting major arguments on the modulus and extension of the wave function. In addition, there is some fundamental ambiguity in the way of counting the nodes, and finally on their interpretation for a *non-separable problem.* The results of fig. 2 contradict both Kleiner's conjecture [12] and other simple adiabatic pictures [13], confirming that there is no obvious argument for assuming nodal surface conservations, between the low and high field limits. The *anticrossing behaviour* of $K = 1$ and K_{max} states just proves the opposite.

Heuristic point of view. This is not a new feature but indeed suitable here to question upon classical mo-

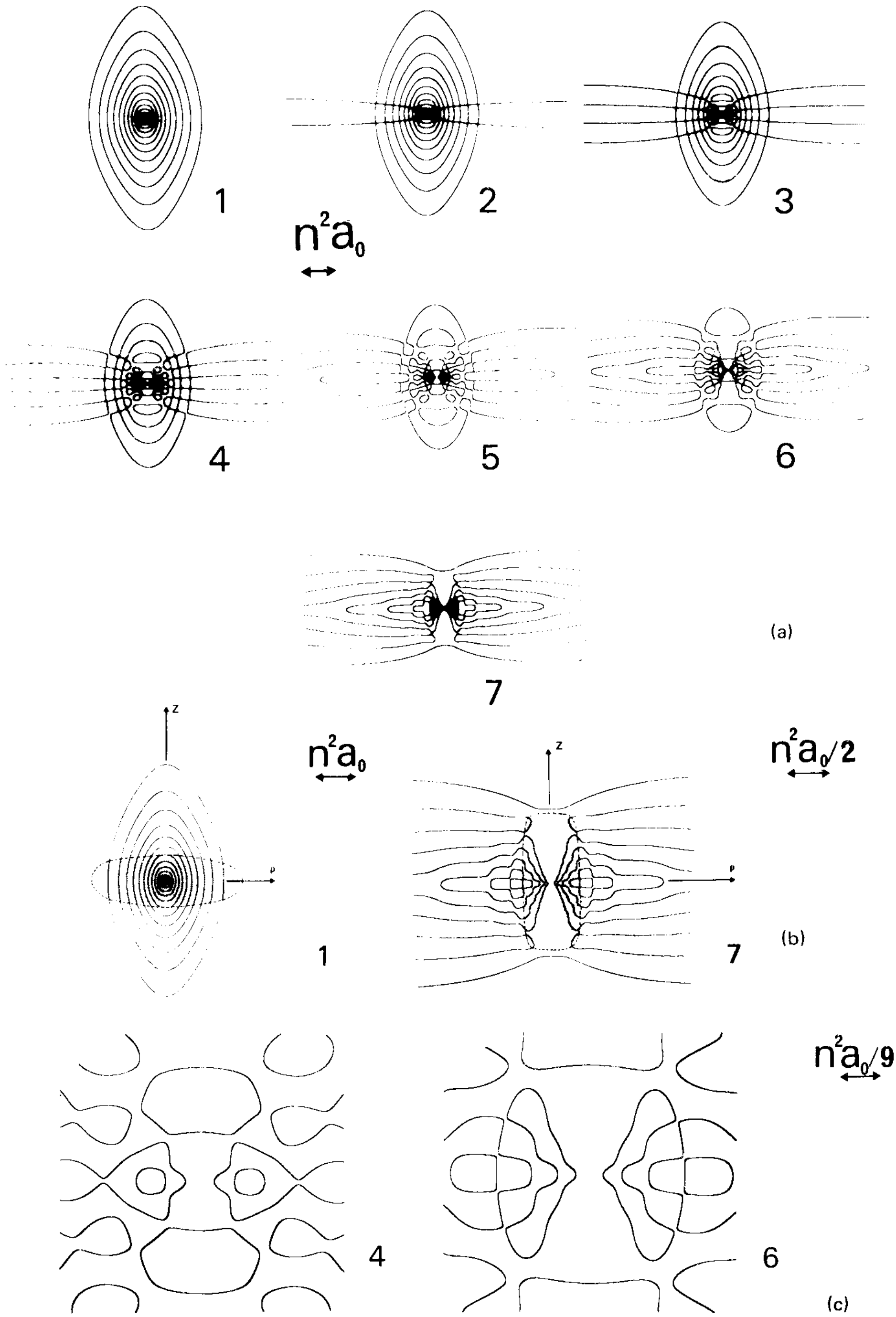


Fig. 2. Inter-l mixing calculations of the nodal surfaces for n = 17 and different values of K (M = 3, odd-parity states). Rotational symmetry along the Z axis has been omitted. (a) General aspects for K = 1 to K = 7 states. (b) The shaded area in the curves corresponds to values of the wave functions greater than 3×10^{-5}. K = 1 states then correspond to motion near the Z = 0 plane while for K = 7 the behaviour is that of excited motion along B. This partly contradicts the distribution of nodal surfaces. (c) Enlargements for K = 4 and K = 6 of the central regions exhibiting a general non-crossing rule of the nodal surfaces. The size of the "anti-crossing" is of the order of $3\,a_0$.

tion, way of quantization, ergodicity and dynamical symmetries for a non-separable problem [15,16]. But these important questions implied in various domains of physics are highly difficult and still open. Only limited results have been obtained for classical systems. They are essentially due to Kolmogorov and Arnold [17,18].

Extensive studies have been performed on the class of systems for which the non-separable hamiltonian can be written down as [17]:

$$H = H_S + V ,$$

where H_S is a separable hamiltonian and V the symmetry breaking perturbing part. Here separable means the existence of conservation laws in phase space for some set of operators and does not necessarily imply a simple geometrical interpretation, in R^3.

General results show that, for initial conditions close to an invariant torus of H_S, the phase space trajectories of H will be of two types. The first one is associated with quasi-periodic trajectories lying in the vicinity of those associated with H_S meaning that the invariant non-resonant torus is slowly deformed and evolving with the perturbation V. The other class is associated with instability regions or faults between the non-resonant invariant tori. For initial conditions belonging to these faults, the trajectories will go away and the variations of the adiabatic invariants (associated with H_S) will be of the order of $\exp(-1/V^{n_d})$ per unit time (where the value of n_d depends on V and $0 < n_d < 1$). The difference in energy between the quasi-periodic orbits which still respect symmetries of H_S and those which do no longer obey the classification is then of the order of $\exp(-1/V^{n_d})$.

For a two-dimensional problem, and for initial conditions lying in faults between two non-resonant tori, the phase curves will always be confined near the faults, whatever the complexity of the resulting motion. These additional constraints are due to topological arguments. The associated physical quantities will always be close to their initial values, if *there is no energetic degeneracy of the hamiltonian.* When energetic degeneracy occurs, which is precisely true near crossing of energy levels, there are no general results but the role of the faults and instabilities will no longer be limited by topological arguments. It is likely that one turns back to the general situation previously described.

There is no rigorous proof that the strongly magnetized hydrogen atom problem will really behave as previously described. It can be shown to be equivalent to a *two-dimensional*, symmetric, anharmonic oscillator but a study of the stability of the classical motion is rather complicated.

The existence of exponentially small anticrossing in the quantum problem seems to be a clue of the existence of trajectories obeying in the classical limit some of the general features previously described. One must stress upon the fact that the smallness of the anticrossing is justified if a decomposition such as (2) holds but the quasi-exponential behaviour is a rather singular feature, which especially cannot be understood in terms of perturbation theory for classical or quantum systems. In contrast, close range corrections to the Coulomb potential are responsible for an enhancement of the sizes of anticrossings (through straightforward mixing of the channels) which is perfectly understood in terms of conventional perturbation theory. It is very enticing and enlightening to compare this exponential feature to what happens for trajectories associated with the faults between invariant tori, and to correlate the exponential behaviour with the existence of instability regions in phase space, for the conjunctions of Coulomb and oscillator-like potentials.

A rough justification could be choosing for H_S the hamiltonian for $Z = 0$ (depending on ρ and including both the Coulomb and diamagnetic potentials), which respects the gross features of the spectrum. Then:

$$V \sim \left(\frac{1}{(\rho^2 + Z^2)^{1/2}} - \frac{1}{\rho} \right) ,$$

which is an oscillator-like potential for small Z. In reduced units, V is of order Z^2/ρ^3 and $V \sim n_Z/n_\rho^6$, where n_ρ is of the order of the Coulomb principal quantum number if $2R/n^3 > H_D$. From the anticrossing behaviour, it follows that $e^{-2n} \approx \exp(-K/V^{1/6})$ which is of the order of the estimation from classical mechanics.

Then a possible origin of the exponential size of the anticrossings might be sought in an enhanced contribution of waves from the instability regions to the semi-classical Green functions, for some special values of energy and magnetic field. Whether such a hypothesis is correct and a manifestation of chaotic behaviour in the system, needs a detailed study of the classical motion of the electron. This is of importance anyway for probing and finding the nature of a possible underlying dynamical symmetry. This has been a complex task for other problems with in the margin interesting questions about chaotic behaviour, way of quantization and quantum analogs. Very old problems laid out in 1917 [15].

References

[1] J.C. Castro, M.L. Zimmerman, R.G. Hulet and D. Kleppner, Phys. Rev. Lett. 45 (1980) 1780.
[2] J.C. Gay, D. Delande and F. Biraben, J. Phys. B13 (1980) L729.
[3] C.W. Clark and K.T. Taylor, J. Phys. B13 (1980) L737.
[4] D. Delande and J.C. Gay, following letter.
[5] M.L. Zimmerman, M.M. Kash and D. Kleppner, Phys. Rev Lett. 45 (1980) 1092.
[6] A.R. Edmonds, J. de Phys. (Paris) C4 (1970) 31, 71.
[7] A.R.P. Rau, Phys. Rev. A16 (1977) 613.
[8] M.L. Zimmerman, J.C. Castro and D. Kleppner, Phys. Rev. Lett. 40 (1978) 1083.
[9] R.J. Fonck, F.L. Roessler, D.H. Tracy and F.S. Tomkins, Phys. Rev. A21 (1980) 861.
[10] N.P. Economou, R.R. Freeman and P.F. Liao, Phys. Rev. A18 (1978) 2506.
[11] J. Simola and J. Virtamo, J. Phys. B11 (1978) 3309.
[12] W.H. Kleiner, Lincoln Lab. Progress Report (1958).
[13] M. Shinada, O. Akimoto, H. Hasegawa and T. Tanaka, J. Phys. Soc. Japan 28 (1970) 4.
[14] A.F. Starace and G.L. Webster, Phys. Rev. A19 (1979) 1629.
[15] A. Einstein, Verh. Dtsch. Phys. Ges. 19 (1917) 82, from ref. [13].
[16] I.C. Percival, Adv. Chem. Phys. 36 (1977) 61.
[17] V. Arnold, Méthode mathématique de la mécanique classique (Mir, Moscow, 1976).
[18] V. Arnold and A. Avez, Problèmes ergodiques de la mécanique classique (Gauthier-Villars, Paris, 1967).