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LETTER TO THE EDITOR

Stark effect on diamagnetic Rydberg states

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Abstract. The Stark effect on diamagnetic Rydberg states of hydrogen is investigated. In the perturbative regime, re-ordering of levels within a hydrogenic n manifold leads to level repulsion when the ratio of field strengths is varied. When the fields become strong enough very narrow avoided crossings between levels from *different* n manifolds appear. In addition we study the Stark effect on recently observed quasi-Landau structure in the irregular part of the spectrum.

The study of highly excited states of atoms in external fields is a topical subject of interest (for a review see, e.g., Nayfeh and Clark 1985). Because of its separability in hydrogen, the Stark effect is well understood and theory has reached a high level of development (Luc-Koenig and Bachelier 1980, Harmin 1982), as has the experimental work on hydrogen (Rottke and Welge 1986, Ng *et al* 1987) and other atoms (Zimmerman *et al* 1979, Rinneberg *et al* 1985). Atomic diamagnetism is much more difficult to handle because of the non-separability of the problem. A breakthrough in the theoretical treatment has been made only very recently (Wintgen and Friedrich 1986a, b, c, d, 1987a, b, Delande and Gay 1986, O'Mahony and Taylor 1986, Wunner *et al* 1986, Wintgen 1987a, Du and Delos 1987), although some remarkable quantum calculations exist from Clark and Taylor (1980, 1982). The situation is similar on the experimental side, where high-resolution spectroscopic data are now available for hydrogen (Holle *et al* 1986, 1987, Wintgen *et al* 1986, Main *et al* 1986) and also for other atoms (Cacciani *et al* 1986a, Rinneberg *et al* 1987).

In contrast to these separate treatments the study of atoms in combined electric and magnetic fields is still in its infancy. Here, we concentrate on the case of parallel electric and magnetic fields. Theoretically, the problem has been treated so far only by semiclassical methods or quantum perturbation theory for weak fields (Braun and Solov'ev 1984, Cacciani *et al* 1986b, Waterland *et al* 1987) and experimental data exist only for weak fields (Cacciani *et al* 1986b, Main 1987, Rinneberg *et al* 1987). In this letter we will report on the first non-perturbative quantum calculations for a hydrogen atom in strong parallel electric and magnetic fields.

The Hamiltonian for a hydrogen atom in a combined electric and magnetic field parallel to the z axis reads (atomic units are used throughout this letter)

$$H = \tfrac{1}{2}p^2 - 1/r + \tfrac{1}{2}\gamma l_z + \tfrac{1}{8}\gamma^2(x^2+y^2) + \phi z \qquad (1)$$

where γ measures the magnetic field in units of $B_0 = 2.35\times10^5$ T and ϕ measures the electric field in units of $F_0 = 5.14\times10^9\ \mathrm{V\,cm^{-1}}$. The only good quantum number is the azimuthal quantum number m, so that the paramagnetic term in equation (1) becomes a trivial energy shift and is dropped from now on. However, no further separation of variables is possible and the remaining problem is two-dimensional.

We have attacked the problem numerically by using semiparabolic coordinates and expanding the Hamiltonian in the complete set of oscillator functions. The algorithm is a straightforward extension of the method described in Wintgen and Friedrich (1987b). An additional difficulty arises here because, strictly speaking, bound states do not exist for $\phi \neq 0$. However, an L_2-basis expansion is justified well below the classical escape threshold $E_{esc} = -2\sqrt{\phi}$ (for $m = 0$), where the electron is trapped by a large potential barrier and the tunnelling widths of the resulting resonances become negligibly small for our purpose. Details of the numerical procedure will be published elsewhere.

Since the Hamiltonian (1) depends on two parameters, a graphical representation of the evolution of the spectrum requires a restriction to a particular choice of ratio of field strengths. This ratio may be arbitrary in its analytic form, but here we propose two meaningful kinds of ratio.

The first one is where we tune ϕ and γ^2 such that ϕ/γ^2 remains constant. In this case the first-order energy shifts of the unperturbed atom become linear in the field strengths ϕ and γ^2. In figure 1 we have diagonalised the Hamiltonian within the hydrogenic manifold $n = 24$, $m = 0$ for all ratios of ϕ/γ^2. The left-hand side of the figure shows the Stark map only, while the evolution of a pure diamagnetic manifold is shown on the right-hand side. The middle part shows the re-ordering of levels due to the different ratios of field strengths. The levels belong to three categories of states characterised by specific values of an adiabatic invariant, which is a combination of components of the Runge-Lunz vector. The boundaries of these subsets are seen clearly on the figure. All the features shown in figure 1 can be explained by a semiclassical treatment of the Runge-Lenz vector, which has been done in the literature (Braun and Solov'ev 1984, Cacciani *et al* 1986b, Waterland *et al* 1987).

The second kind of ratio is where we fix $\phi\gamma^{-4/3}$. This is just the ratio with which the classical forces scale. Figure 2(*a*)-(*c*) shows the evolution of the spectrum in the n-mixing regime for three values of this ratio: (*a*) 0.007, (*b*) 0.070 and (*c*) 0.105. These

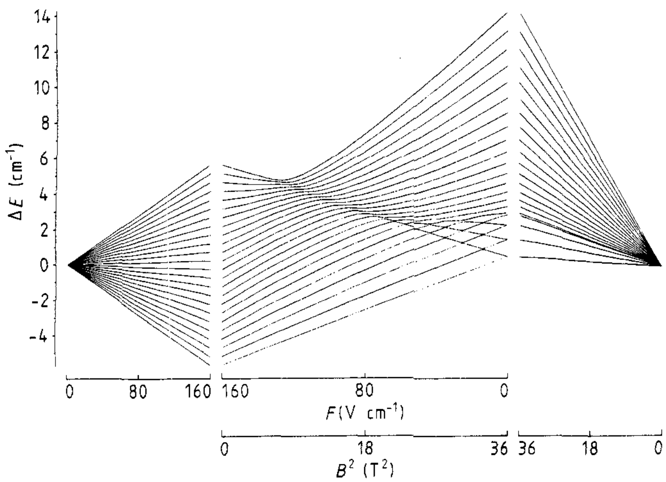

Figure 1. Perturbative energy shift for the states belonging to the $n = 24$, $m = 0$ manifold. Stark (diamagnetic) map in the left (right) panel, arbitrary ratio of field strengths in the middle part.

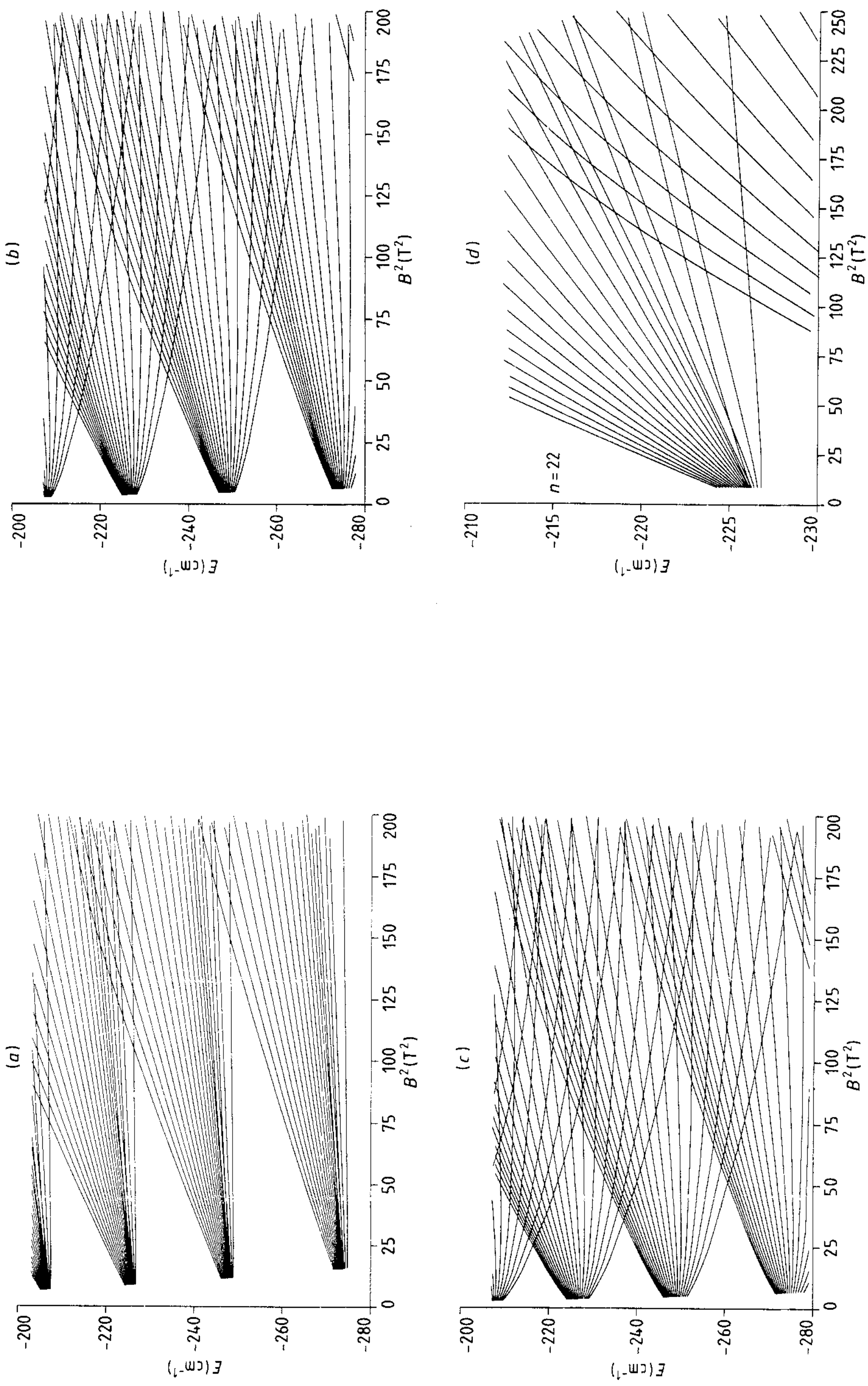

Figure 2. Evolution of the spectrum in the $m = 1$ subspace for fixed values of $\phi\gamma^{-4/3}$. (*a*) 0.007, (*b*) 0.070, (*c*) 0.105. (*d*) shows a part of (*a*) on an expanded scale. Note that the electric field, not labelled on the axes, is also varying.

ratios belong to cases where the diamagnetic force (a) or the electric force (c) is dominant, or where both forces have comparable magnitude (b). Surprisingly, different n manifolds overlap without significant mutual interaction. The avoided crossings of states belonging to adjacent n manifolds shown in figure 2 are far beyond graphical resolution. For pure electric fields states may cross because of the separability of the Hamiltonian and approximate separability is also present for pure magnetic fields (Wintgen and Friedrich 1986a). We suppose the very small avoided crossings for combined fields to be a consequence of the structure of classical phase space which remains regular for the energies and field strengths shown in figure 2, although three forces with completely different symmetries (Coulomb, electric and magnetic field) are acting on the electron (Wintgen 1987b).

A part of the spectrum of figure 2(a) is shown on an expanded scale in figure 2(d). Note the re-ordering of levels within the $n = 22$ manifold that leads to level repulsions, whereas states belonging to *different* n manifolds nearly cross. The re-ordering takes place because $\phi\gamma^{-4/3}$ remains constant and not ϕ/γ^2.

The systematic near-degeneracies shown in figure 2 are only present as long as the classical phase space is filled with tori. These break up for smaller energies or larger field strengths and the classical dynamics become more and more chaotic (Wintgen 1987c). In this regime and for pure magnetic fields new 'quasi-Landau' structure has been discovered recently (Holle *et al* 1986, Wintgen and Friedrich 1986d, Al-Laithy *et al* 1986, Main *et al* 1986). These structures were traced back to be a quantum manifestation of classical periodic orbits (Wintgen 1987a, Du and Delos 1987; for a review of periodic-orbit theory see, e.g., Berry 1983). The periodic orbits of the classical system appear as peaks in the Fourier transform of the quantum level density. The positions of the peaks are given by the action S_i of the closed orbits. Since for a vanishing electric field the orbits are partly degenerate with respect to the $z = 0$ plane, we expect a linear Stark splitting of Fourier peaks belonging to orbits with permanent dipole moment.

Figure 3(a) shows the Fourier transform of the fluctuating part of the quantum level density (level density minus mean level density) for a pure diamagnetic spectrum ($m = 1$) where we have fixed the ratio of energy and magnetic field strength to $E\gamma^{-2/3} = -0.3$. (This means that the Coulomb and diamagnetic forces have always the same ratio; for further details see Wintgen and Friedrich 1987b.) Figure 3(b)-(e) show the Fourier transforms of spectra with additional electric fields. The peaks at $S_i = 0.98$, 1.13 and 1.95 belong to orbits which are symmetric with respect to the $z = 0$ plane. Note that these peaks are roughly stable against additional (parallel) electric fields. The Coulomb orbit parallel to the field (labelled as C) and the orbit belonging to the $0.64\hbar\omega$ quasi-Landau modulation near $E = 0$ (I_2) (Holle *et al* 1986, Wintgen and Friedrich 1987b) are responsible for the peak at $S = 1.27$ in figure 3(a). These two orbits have permanent dipole moments and this leads to a splitting of the corresponding Fourier peak. The semiclassical predictions, that are the actions of the orbits, are marked as arrows. The agreement between the splitting of the Fourier peaks and semiclassical predictions is evident.

In conclusion, we have studied the influence of parallel electric and magnetic fields on a hydrogen atom quantum mechanically. In the perturbative regime all features can be understood within a semiclassical treatment of the Runge-Lenz vector. In the low inter-n mixing regime levels of adjacent n manifolds nearly cross as a consequence of the regular structure of classical phase space. In the strong field mixing regime we have shown the Stark splitting of quasi-Landau modulations.

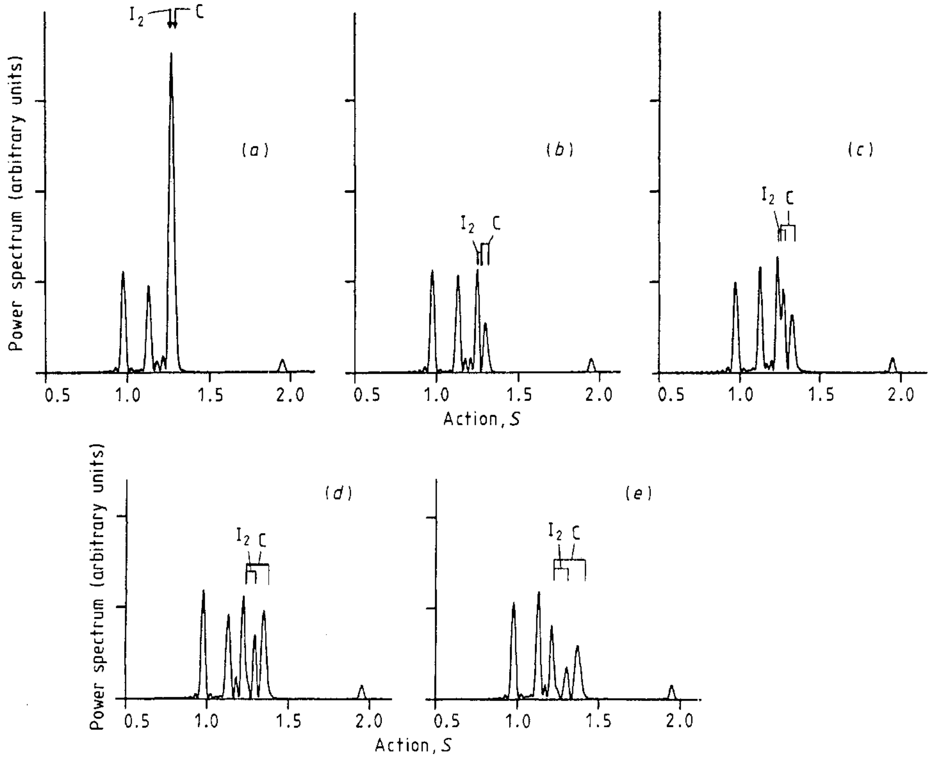

Figure 3. Power spectrum of the fluctuating part of the quantum level density for fixed scaled energy $E\gamma^{-2/3} = -0.3$ and fixed field strength ratios $\phi\gamma^{-4/3}$: (*a*) 0, (*b*) 0.004, (*c*) 0.008, (*d*) 0.012, (*e*) 0.016. The arrows mark the actions of orbits with dipole moments, see text. Six hundred levels are included for each Fourier transform.

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