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SCALED-ENERGY SPECTROSCOPY OF HELIUM AND BARIUM RYDBERG ATOMS IN EXTERNAL FIELDS

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INTRODUCTION

Rydberg atoms in many aspects strongly resemble the hydrogen atom and are readily accessible for detailed investigation of e.g. the influence of external magnetic or electric fields on the level structure. The hydrogen atom in a sufficiently strong magnetic field B is a prototype system that classically exhibits chaotic behaviour. This is a consequence of the diamagnetic effect, which is proportional to $(B \times r)^2$ and which at low field strength is responsible for the mixing of angular momentum states (r is the radius of the electron orbit). When diamagnetism dominates only two conserved quantities (energy W and z -component of angular momentum l) exist for this system with three spatial degrees of freedom, a prerequisite for chaos in classical physics. The diamagnetic effect not only becomes important for large values of B , but also for large values of r . As the radius r scales as n^2 (n is principal quantum number) it follows that the diamagnetic effect grows with n^4 , so that in highly-excited states it may be studied at relatively moderate magnetic field strengths. The classically chaotic regime in the hydrogen atom is reached when the Lorentz force exerted by the field on the electron about equals the Coulomb force binding the electron. In the hydrogen ground state ($n=1$) this condition can only be fulfilled for the huge field strength of 2.35×10^5 Tesla, whereas in a Rydberg state with $n=100$ a field of 0.60 Tesla suffices. This, together with the fact that the hydrogen atom is experimentally not easily accessible, constitutes the major argument to investigate this effect also in Rydberg states of more complex atoms. An additional point of interest then relates to the influence of an extended atomic core, represented by a quantum defect in regular Rydberg sequences, on the observed spectra in the presence of a magnetic field.

Similar arguments hold for the investigation of Rydberg states in the presence of an external electric field E . Now it is the linear Stark effect, that at moderate field strengths induces mixing of angular momentum l -states, reflected in the appearance of angular momentum manifolds in the spectra, and at high field strengths also mixing of n -states. Finally field ionization will occur in sufficiently strong fields. In Rydberg states the sensitivity for electric fields is again strongly enhanced. In contrast to the

magnetic field case the Hamiltonian of the hydrogen atom in an external electric field can be fully separated and chaos in the classical problem does not occur. It is a problem of significant interest in atomic physics to investigate Rydberg states of non-hydrogen atoms in the presence of strong electric fields as well, in particular with respect to core-induced effects.

In this contribution experimental results of a study of Rydberg states in the He atom in the presence of a magnetic field as well as of Rydberg states in the Ba atom, including autoionizing states, in the presence of an electric field will be presented and discussed. Spectra are recorded under conditions where the classically important scaled-energy parameter is kept constant. This allows for a Fourier transform of the experimental spectra to so-called scaled-action spectra, which reflect the closed periodic orbits of the system. A direct comparison with semi-classical periodic-orbit theory then is feasible, resulting in a beautiful interpretation of many of the observations.

This contribution is organized as follows. First the Hamiltonians of the hydrogen atom in the presence of external fields and the appropriate scale transformations will be discussed. Next the semi-classical periodic-orbit theory will be briefly summarized. Then the He experiment in a magnetic field will be presented, followed, finally, by a discussion of the Ba experiment in electric fields.

HAMILTONIAN OF H-ATOM IN MAGNETIC AND ELECTRIC FIELDS

The Hamiltonian of a spinless H-atom in a magnetic field B along the z-axis is:

$$H = H_0 + H_p + H_d = \frac{p^2}{2} - \frac{1}{r} + \frac{\gamma}{2} L_z + \frac{1}{8} \gamma^2 \rho^2 \quad (1)$$

Here H_0 is the zero-field Hamiltonian, H_p the paramagnetic effect (Zeeman effect, $H_p=0$ for $L_z=0$) and H_d the diamagnetic effect ($\rho^2 = x^2 + y^2$); $\gamma = B/B_c$ with $B_c = 2.35 \times 10^5$ Tesla (critical field). This Hamiltonian (for $H_p=0$) can be transformed using scaling parameters

$$\tilde{r} = \frac{1}{2} \gamma^{-2/3} r; \tilde{p} = \gamma^{-1/3} p \quad (2)$$

resulting in the *scaled Hamiltonian* $\tilde{H}$:

$$\tilde{H} = \gamma^{-2/3} H = \frac{\tilde{p}^2}{2} - \frac{1}{\tilde{r}} + \tilde{\rho}^2 \quad (3)$$

This transformation shows that the classical motion of the electron is not determined by two independent parameters (energy W and magnetic field strength B) but only by a single parameter (*scaled energy*):

$$\tilde{\epsilon} = \gamma^{-2/3} W \quad (4)$$

This scaled energy $\tilde{\epsilon}$ is a measure for the onset of chaos ($\tilde{\epsilon}=-1$ corresponds to the situation where Coulomb force and diamagnetic force on the electron are equal). It is of interest to note that also the commutator $[p_z, z] = i\hbar$ scales, resulting in:

$$[\tilde{p}_z, \tilde{z}] = i\gamma^{1/3}\hbar = i\hbar_{eff} \quad (5)$$

The semi-classical quantization condition for the action S along a given electron orbit n also transforms:

$$S_n = \frac{1}{2\pi} \int p dq = n\hbar \quad (6)$$

$$\tilde{S}_n = \frac{1}{2\pi} \int \tilde{p} d\tilde{q} = n\gamma^{1/3}\hbar = n\hbar_{eff} \quad (7)$$

The *scaled action* $\tilde{S}_n = \gamma^{1/3}S_n$ only depends on $\tilde{\epsilon}$. This implies that the Fourier transform of a spectrum recorded at constant scaled energy, which is linear in the variable $\gamma^{-1/3}$, shows resonances at values of the scaled action $\tilde{S}_n = n\gamma^{1/3}\hbar$; $n\gamma^{1/3}$ is the conjugated variable.

The Hamiltonian of the H-atom in an electric field E along the z-axis is:

$$H = H_0 + H_e = \frac{1}{2}p^2 - \frac{1}{r} + fz \quad (8)$$

Here H_0 is the zero-field term, H_e the field term; $f = E/E_c$ with $E_c = 5.14 \times 10^9$ V/cm. Also in this case a scale transformation is possible:

$$\tilde{r} = f^{1/2}r; \tilde{p} = f^{-1/4}p \quad (9)$$

resulting in the *scaled Hamiltonian* $\tilde{H}$:

$$\tilde{H} = f^{-1/2}H = \frac{1}{2}\tilde{p}^2 - \frac{1}{\tilde{r}} + \tilde{z} \quad (10)$$

This again shows that the classical motion of the electron is governed by the single *scaled-energy parameter* $\tilde{\epsilon}$:

$$\tilde{\epsilon} = f^{-1/2}W \quad (11)$$

The value $\tilde{\epsilon} = -2$ corresponds to the saddle-point energy, related to the maximum in the combined Coulomb and electric field potential determining the classical field ionization limit.

The recognition that scaling transformations do exist for the H-atom in external fields [1] now forms the basis of *scaled-energy spectroscopy*. Complex excitation spectra of Rydberg atoms in strong external fields, when recorded at constant scaled energy, show remarkable regular Fourier transforms. A limited set of resonances shows up in the scaled-action spectra, each resonance corresponding to a classical periodic orbit. This type of spectroscopy in the presence of a magnetic field was first applied to the H-atom itself, using pulsed laser excitations, by Holle et al [2]. Pioneering scaled-energy experiments in an electric field were performed by Eichmann et al [3] in Na Rydberg states and in Kleppners group on Li Rydberg states [4].

SEMI-CLASSICAL PERIODIC-ORBIT THEORY

In semi-classical periodic-orbit theory (see [5] and references therein) it is assumed that a frequency spectrum contains numerous sinusoidal oscillations, each related to a classical periodic orbit. These orbits may be regular or chaotic, depending on the parameters of the system. When an orbit is regular, it may be traversed many times. These multiple traverses give rise to higher harmonics in the frequency spectrum and interferences between all harmonics result in sharp resonances, corresponding to the quantum states. For chaotic orbits, however, the probability of repeated traversals is exponentially small. The signature of a chaotic orbit in the frequency spectrum is a deformed sine wave.

Du and Delos [6] and Gao and Delos [7] give a physical picture explaining the spectral features of hydrogenlike atoms in the presence of external fields using closed-orbit theory. In their model incident electromagnetic radiation excites the electron

Figure 1. Schematic representation of closed-orbit theory [6,7]. (1) Outgoing Coulomb waves generated in the excitation process. (2) In the classical region the wave fronts follow classical trajectories. (3) Returning waves interfere with outgoing Coulomb waves producing oscillations in the frequency spectrum.

close to the nucleus in an outgoing, zero-energy Coulomb wave as near the core effects of the external field are negligible small (see Fig. 1). At large distances ($\sim 50a_0$, with a_0 the Bohr radius) the wave is assumed to propagate semi-classically. The outgoing wave fronts then follow classical trajectories and propagate far out in the field. As the influence of the magnetic or electric field grows with distance some of the trajectories will eventually curve back to the nucleus. Near the nucleus the incoming waves have to be treated quantum mechanically again. Incoming Coulomb waves interfere with the outgoing waves, resulting in oscillations in the absorption spectrum. These oscillations are described by the well known Gutzwiller trace formula [5,6], involving a summation over all closed periodic orbits and their traversals. A Fourier transform of the absorption spectrum results in an action spectrum, where each peak corresponds to a classical orbit. The intensity of each action peak then is determined by the stability of the orbit and by the external parameters (excitation process and light polarization). The effect of the core in a non-hydrogenic system may be incorporated by introducing a phase shift in the scattered wave function as in standard quantum defect theory.

Closed periodic orbits may be found assuming that the electron leaves the core region with a momentum directed perpendicular to the sphere with radius $r \sim 50a_0$ and with a value determined from eq. (1) for given value of the energy W :

$$p_r = \sqrt{W - \frac{1}{r} + \frac{1}{8}\gamma^2\rho^2} \quad (12)$$

Then the classical Hamilton equations are integrated. A closed orbit is found when the final momentum upon reaching the core region is again perpendicular to this sphere. The orbit may be traversed again in the same or reversed direction after scattering on the nucleus. The stability of the orbit follows from its sensitivity to small variations in

Figure 2. Experimental constant-scaled-energy spectrum for $\tilde{\epsilon} = -0.700$. Laser frequency and magnetic field were varied simultaneously to keep the scaled energy constant.

the initial conditions, which becomes extreme in the high-field regime. For each closed orbit the (scaled) classical action can be calculated by straightforward integration along the closed trajectory and its value compared with experimental data.

Frequency spectra in the presence of external fields can also be calculated quantum mechanically. For highly-excited states this involves the diagonalization of huge energy matrices, for which special numerical techniques have been developed [8,9]. Such calculations are also possible under conditions of constant-scaled energy, so that a direct comparison with experimental data and Fourier transforms can be made.

HELIUM RYDBERG ATOMS IN A MAGNETIC FIELD

Scaled-energy experiments in a magnetic field were performed in helium Rydberg states at two values of the scaled energy: $\tilde{\epsilon} = -0.700$ and $\tilde{\epsilon} = -0.400$. Helium was excited to $1snp$ Rydberg states (up to $n = 200$) in a collimated beam of discharge-populated 2^3S_1 metastable atoms with 260 nm light from a frequency-doubled CW ring dye laser pumped by an Ar-ion laser. Atomic beam and laser beam perpendicularly intersected (in a well-shielded interaction region) to eliminate Doppler effects to a large extent, resulting in a residual linewidth of 25 MHz. The magnetic field was produced with conventional current-driven coils (maximum value about 0.2 Tesla) and aligned very carefully parallel to the atomic beam to avoid motional Stark effects. The He Rydberg atoms were field-ionized after leaving the interaction chamber, mass selected with a quadrupole filter and counted with an electron multiplier. The visible CW laser was scanned continuously over 30 GHz (60 GHz at 260 nm), monitoring the change in frequency ν on-line with a Fabry-Perot etalon. This calibration spectrum in turn was used to control the current through the magnetic field coils in such a way that the scaled energy $\tilde{\epsilon}$ is kept constant. In extended spectra several of these 30 GHz scans were overlapped, using the zero-field Rydberg resonances as references. More details of the experimental setup can be found in [10] and [11]. As an example in Fig. 2 a recorded spectrum at $\tilde{\epsilon} = -0.700$ is shown, where the excitation energy was varied from -9.7 cm^{-1} to -8.7 cm^{-1} and the magnetic field simultaneously from 1193 Gauss to 1002 Gauss. From this dense spectrum not much information can be extracted directly.

However, after Fourier transformation to a scaled-action spectrum a much more insightful picture results, revealing the closed periodic orbits of the system. In Fig. 3 (upper part) such a Fourier transformation is shown for $\tilde{\epsilon} = -0.400$ (mixed regular-

Figure 3. Squared Fourier transform of the constant-scaled energy spectrum at $\tilde{\epsilon} = -0.400$. Upper part: transform of a spectrum in the range $112.3 < \gamma^{-1/3} < 161.1$. Lower part: transform of a quantum mechanically calculated energy spectrum.

chaotic regime). Although this scaled-action spectrum is less regular than the spectrum recorded at $\tilde{\epsilon} = -0.700$ [11], many resonances still can be connected to two types of closed orbits and their traversals. The resonance at $\tilde{S}=1.12$ corresponds to the fundamental, so-called vibrator orbit V_1 , directed parallel to the magnetic field, whereas there is a weak indication for the presence of the fundamental rotator orbit R_1 , in the plane perpendicular to the magnetic field at $\tilde{S}=0.92$ (see [2] for this classification). The first bifurcation from the fundamental vibrator, traversed twice, appears at $\tilde{S}=2.21$ and is denoted V_2^1 . Its action is about equal to the action of the two times traversed fundamental vibrator V_2 . Higher traversals appear at $\tilde{S}=3.27$ (V_3^1) and 4.31 (V_4^1). The second bifurcation of the vibrator orbit, traversed three times, lies at $\tilde{S}=3.35$ (V_3^2). The first strong rotator orbit at $\tilde{S}=2.80$ corresponds to three traversals of the first bifurcation of the fundamental orbit (R_3^1), at $\tilde{S}=3.84$ to R_4^1 . The number of unstable orbits in the case of $\tilde{\epsilon} = -0.400$ drastically increases compared to $\tilde{\epsilon} = -0.700$, and also the number of resonances that does not fit the classification in terms of rotator - vibrator orbits, called exotics. Such exotic orbits may contribute to the resonances observed at e.g. $\tilde{S} = 6.64, 8.85$ and 9.15 . In the action spectrum for $\tilde{\epsilon} = -0.400$ there is clear evidence for electron scattering from vibrator into rotator orbits and vice versa. This is evidenced e.g. by the occurrence of resonances at $\tilde{S} = 5.01$ and 9.03 , which are the sum of actions for V_2^1 and R_3^1 orbits (2.21 and 2.80), respectively V_4^2 and R_5^2 orbits (4.42 and 4.61). The occurrence of sum actions (not possible in the hydrogen atom) was for the first time observed in the $\tilde{\epsilon} = -0.700$ spectrum [8]. The proliferation of unstable orbits as well as the more pronounced occurrence of sum orbits results in less good agreement between the observed and calculated (with classical closed-orbit theory) action spectrum in the case of $\tilde{\epsilon} = -0.400$ than in case of $\tilde{\epsilon} = -0.700$ [11].

We also calculated a quantum mechanical scaled-energy spectrum for $\tilde{\epsilon} = -0.400$ applying the same computer code as in the $\tilde{\epsilon} = -0.700$ case [8]. The Fourier transform

Figure 4. Calculated distribution of nearest neighbour separations for $\tilde{\epsilon} = -0.710$ for the hydrogen (left) and helium (right) atom in a magnetic field. In helium the quantum defect of the $1snp$ Rydberg series has been taken into account. Along the horizontal axis the scaled separation $\gamma^{-1/3}$ is plotted.

of this quantum mechanically calculated spectrum is plotted in the lower part of Fig. 3. These calculations result in a much better agreement with observations, not only for the S -values where resonances occur but also with respect to their strengths, when compared with the closed-orbit calculations. Such quantum mechanical calculations may also be used to determine level statistics, i.e. the distribution of nearest neighbour separations (s). We calculated these distributions in the regular regime ($\tilde{\epsilon} = -0.700$) for both the hydrogen and the helium atom. In hydrogen a clear Poissonian distribution $P(s) = P(0)\exp(-s)$ is obtained, which is shown on the left in Fig. 4. However, in helium a Wigner type of distribution $P(s) = P(0)s \cdot \exp(-\pi s^2/4)$ is found, as shown on the right in Fig. 4. To get good agreement between the experimental and calculated energy spectrum for the helium atom in a magnetic field for this case of $\tilde{\epsilon} = -0.700$ it was necessary to include the quantum defect of 0.0684 for the $1snp$ Rydberg series [8]. Then the level statistics shown in Fig. 4 results. A change from Poisson to Wigner type of distributions as a function of the scaled-energy parameter is considered to be a signature of the transition from a classically regular system to a chaotic system. However, the effect calculated for the level statistics in the helium atom in the still regular regime does not relate to this type of transition. In helium it is the core-scattering effect (represented by the quantum defect) that is responsible for the change in level statistics as compared to hydrogen; it is therefore referred to as core-induced chaos [4]. The agreement between experimental and calculated energy spectrum at $\tilde{\epsilon} = -0.700$ is excellent. This stimulates experiments to measure the level statistics under various circumstances. Such experiments are in progress.

BARIUM RYDBERG ATOMS IN AN ELECTRIC FIELD

Scaled-energy experiments in an electric field have been performed in three barium Rydberg series $6snf$, $5d_{3/2}nf$ and $5d_{5/2}nf$ at two values of the scaled energy below the field-ionization limit ($\tilde{\epsilon} = -2.94$ and -2.35) and one value above this limit ($\tilde{\epsilon} = -1.76$). The $6snf$ states are bound and converge to the $6s$ -ionization limit of the atom, whereas the $5dnf$ states belong to weakly autoionizing series converging to the $5d_{3/2}$ - and $5d_{5/2}$ -excited states of the ion. So a study of these various series in principle allows for a comparison of differences induced by core effects. The states of interest were populated in a one-photon excitation process with narrowband, tunable CW laser radiation from

the $6s5d^1D_2$ and $5d^2G_4$ metastable states. For the excitation of $6snfJ = 3$ states from $6s5d$ UV light from an intra-cavity frequency-doubled ring dye laser operating around 600 nm was used, whereas $5dnf$ states (dominantly $J=5$) were excited with a CW Stilbene ring dye laser operating in the blue spectral range around 450 nm. The laser beam perpendicularly intersected a well-collimated beam of Ba atoms between two capacitor plates in an excitation chamber, well shielded against stray electric fields. In the beam metastable states were populated by running a DC-discharge between the oven and a tungsten filament heating the oven. As in the helium case the frequency of the laser was scanned in the presence of an electric field and continuously monitored with a Fabry-Perot etalon. This etalon signal was used to adjust the electric field in such a way that spectra were recorded at constant-scaled-energy value. We quote an absolute uncertainty in the value of $\tilde{\epsilon}$ of about 0.06, determined by the uncertainty in the measurement of the distance between the capacitor plates. Electrons directly produced by autoionization were, in case of the $5dnf$ -series, detected using an electron multiplier positioned above one capacitor plate which contained a grounded fine wire mesh. In case of the $6snf$ states electrons were produced by field-ionizing the highly-excited atoms downstream from the excitation chamber in a second chamber. More details may be found in refs. [12,13].

Scaled-energy spectra were recorded for high- n Rydberg states ($n=60-80$) by overlapping several laser scans. Linewidths of $6snf$ states below the field-ionization limit did have the Doppler-limited value of about 10 MHz, whereas broadening was apparent above this limit. The linewidth of the $5dnfJ = 5$ autoionizing levels in zero field was of the order of 30 MHz, but broadened in the field by the admixture of states with a lower l -value with enhanced autoionization rates. Above the field-ionization limit autoionization into the $5d$ -continuum resulted in broad peaks with a width of about 100 MHz. In Fig. 5 scaled-action spectra for all three Rydberg series for $\tilde{\epsilon} = -2.35$ are shown as obtained from a Fourier transform of the frequency spectra, whereas in Fig. 6 similar spectra for $\tilde{\epsilon} = -1.76$ are reproduced. The typical grouping of lines, in particular in the spectra for $\tilde{\epsilon} = -2.94$ and $\tilde{\epsilon} = -2.35$, is similar to that observed in Na [3] and Li [4] experiments and can be interpreted directly for large negative $\tilde{\epsilon}$. The energy difference between levels within a Stark manifold is responsible for the overall group structure, whereas the energy difference between adjacent Rydberg n -levels determines the splitting within each group. This latter difference also results in the resonance at low scaled action at about 0.44, which corresponds to the uphill / downhill orbits along the z -axis.

Comparing the Fourier spectra in Figs. 5 and 6 the resonances in the three series are found at nearly equal values of the scaled action, although for large scaled actions minor differences become apparent for the higher $\tilde{\epsilon}$ values. This is as expected in periodic-orbit theory, where only the motion of the excited electron far outside the core is important. This motion is governed by classical equations, which do not involve the state of the core. However, the heights of the resonances show significant differences, in particular when comparing the Fourier spectrum of the $6snf$ series with those of the $5dnf$ series and at increased $\tilde{\epsilon}$ values. The height of a resonance is determined by the intensity of the recurring excitations in the oscillator strength distribution, which directly relate to the transition probabilities at zero-field, and by core-scattering processes (sum orbits). The different probabilities for $6snf$, $5d_{3/2}$ - and $5d_{5/2}nf$ excitations only partly explain the observed peak-height differences. Near and above the classical field-ionization limit ($\tilde{\epsilon} = -2.0$) the frequency spectra for the $5dnf$ series show line broadening, resulting in high intensities for scaled actions close to zero as is clear in the $\tilde{\epsilon} = -1.76$ spectrum when comparing the $5dnf$ results with those for $6snf$ (see Fig. 6). The

Figure 5. Squared Fourier transform of experimental spectra for $\tilde{\epsilon} = -2.35$ (below the saddle point) for the 6snf and 5dnf Rydberg series.

Figure 6. Squared Fourier transform of experimental spectra for $\tilde{\epsilon} = -1.76$ (above the saddle point) for the 6snf and 5dnf Rydberg series.

Figure 7. Comparison between experimental Fourier spectrum and closed-orbit calculations for $\tilde{\epsilon} = -2.35$ for the $5d_{3/2}$ - (left) and $5d_{5/2}$ - (right) series.

number of experimental peaks, in particular at high scaled action (recurrence time) strongly decreases with increasing value of $\tilde{\epsilon}$ while also the typical grouping of peaks disappears. Above the saddle point only a limited number of closed orbits is observed. This can also be understood within the framework of closed-orbit theory. For $\tilde{\epsilon} > 0$ only one unstable orbit, the uphill orbit parallel to the electric field, exists. For $\tilde{\epsilon} < 0$ out of this orbit new orbits bifurcate, thus proliferating the number of peaks in the action spectrum. The downhill orbit, also parallel to the electric field axis, becomes possible at $\tilde{\epsilon} = -2$. Closed-orbit calculations were performed for all experimental conditions, taking into account the respective quantum defects for the Rydberg electron: 0.17 for $6snfJ=3$, 0.07 for $5d_{3/2}nfJ=5$ and 0.14 for $5d_{5/2}J=5$. Although in most case the positions of the resonances could be reproduced well and can be assigned to classical orbits, it turned out to be extremely difficult to reproduce peak heights. Surprisingly, none of the strong peaks are due to a repetition of a parallel orbit, but many peaks are due to recurrences of other types of orbits, and in particular in the $6snf$ spectra, sum orbits strongly proliferate. This latter phenomenon makes peak-height calculations extremely difficult. Many more orbits are calculated than experimentally observed, whereas their theoretical intensities turn out to be of the same order of magnitude (see Figs. 7 and 8). Four peaks in the $\tilde{\epsilon} = -2.35$ spectrum of the $5d_{5/2}$ -series could not be assigned. However, they show a remarkable connection: the strongest ones at $\tilde{S} = 6.33$ and $\tilde{S} = 9.34$ differ 3.01 in action, i.e. the action of the most intense experimental peak. The other two at $\tilde{S} = 5.87$ and $\tilde{S} = 9.78$ are shifted 0.44 in action from the stronger ones, which corresponds to the action of the uphill orbit. Remarkably the experimental spectra for $\tilde{\epsilon} = -2.35$ and $\tilde{\epsilon} = -1.76$ are dominated by a single strong peak, at $\tilde{S} = 3.01$ and at $\tilde{S} = 1.98$ respectively, and their recurrences. The corresponding orbits lie in the plane perpendicular to the electric field! Also the majority of other peaks are oriented in the same direction ($\theta = 90$ degrees), for which no simple explanation is available. It was a surprise in itself that for increasing value of $\tilde{\epsilon}$ the number of calculated orbits increases dramatically, in contradiction with observations. Especially for $\tilde{\epsilon} = -1.76$ it might be expected that only a few orbits would remain that do not lead to escape of the Rydberg electron over the saddle point.

Figure 8. Comparison between experimental Fourier spectrum and closed-orbit calculations for $\varepsilon = -1.76$ for the $5d_{3/2}$ - (left) and $5d_{5/2}$ - (right) series.

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REFERENCES

- [1]. D. Wintgen and H. Friedrich, Phys. Rev. **A35** (1987) 1464.
- [2]. A. Holle, J. Main, G. Wiebusch, H. Rottke and K.H. Welge, Phys. Rev. Lett. **61** (1988) 161.
- [3]. U. Eichmann, K. Richter, D. Wintgen and W. Sandner, Phys. Rev. Lett. **61** (1988) 2438.
- [4]. M. Courtney et al., Phys. Rev. Lett. **73** (1994) 1340; Phys. Rev. Lett. **74** (1994) 2438; Phys. Rev. **A51** (1995) 1538.
- [5]. M.C. Gutzwiller, Chaos in classical and quantum mechanics, (1991 Springer Verlag NY).
- [6]. M.L. Du and J. Delos, Phys. Rev. **A38** (1988) 1896, 1913; Phys. Rev. Lett. **58** (1987) 1731; IEEE J. Quantum Elec. **24** (1988) 1445.
- [7]. J. Gao, J. Delos, Phys. Rev. **A46** (1994) 869, 1455; Phys. Rev. **A42** (1992) 1449.
- [8]. D. Delande, K.T. Taylor, M.H. Halley, T. van der Veldt, W. Vassen and W. Hogervorst, J. Phys. **B27** (1994) 2771.
- [9]. P.A. Dando, T.S. Monteiro, D. Delande and K.T. Taylor, Phys. Rev. Lett. **74** (1995) 1099.
- [10]. T. van der Veldt, Diamagnetism in helium Rydberg atoms (1993, thesis Vrije Universiteit Amsterdam).
- [11]. T. van der Veldt, W. Vassen and W. Hogervorst, Europhys. Lett. **21** (1993) 903.
- [12]. G.J. Kuik, High resolution studies of electric field effects on autoionising Rydberg states of barium (1997, thesis Vrije Universiteit Amsterdam).
- [13]. G.J. Kuik, A. Kips, W. Vassen and W. Hogervorst, J. Phys. **B29** (1996) 2159.