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Recurrence spectra of non-hydrogen Rydberg atoms in parallel electric and magnetic fields*

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We present a new method for computing the recurrence spectra of $n \approx 40$, $m=0$ lithium Rydberg atoms in strong parallel external electric and magnetic fields. This method is based on an extended closed-orbit theory allowing the computation of the scattering of the electron by the ionic core. We pay particular attention to the scaling properties, which are extremely important for understanding the correspondence between classical and quantum mechanics. The spectra with a constant scaled electric field $\tilde{F}=0.01$ and a scaled energy $\varepsilon = -0.03$ are recorded and compared with those of hydrogen obtained by the standard closed-orbit theory. The result shows that the additional strong resonance structures can be interpreted in terms of the core-scattered classical closed orbits.

Keywords: closed-orbit theory, recurrence spectrum, region-splitting consistent and iterative method, core scattering

PACC: 0365S, 3210W, 3290, 3150

1. Introduction

Rydberg atoms in static external fields have attracted much interest in recent years[1] because they are the most basic quantum systems in which chaos can be observed experimentally.[2] In the regions of near ionization threshold, the transitions between individual discrete energy levels are no longer resolved and the interest, therefore, shifts to characterize the global properties of the system,[3] particularly the large-scale structures of the smoothed oscillator strength distribution.

As far as we know, until now there has been no effective quantum mechanical method for calculating the photoabsorption spectra of high-lying Rydberg states of non-hydrogen atoms in strong electric and magnetic fields. Since the quantum mechanics treatment requires the truncation of an immense basis expansion, such as the R-matrix method,[4] thus it is especially difficult to obtain converged results. The semiclassical calculation can provide an interpretation and insight that cannot easily be obtained from the quantum calculation, particularly in the region of chaos. And now, the semiclassical closed-orbit theory has been extensively used to explain the photoabsorption spectra of atoms in external fields due to its clear physical picture and wide applicability. The theory for hydrogen atoms in a magnetic field was developed by Du and Delos,[5] and from then on, this theory has reached the maturity that semiclassical calculations of spectra are highly feasible, such as investigating the photoabsorption of H^- in various external fields, as well as the highly excited states of the hydrogen atom.[6–9] This theory can also be applied to describe the recurrence spectra, the Fourier transform of photoabsorption spectra,[10,11] of non-hydrogen atoms. In this case, it is necessary to take into account the scattering contributions arising from the ionic core, and a complex spectral calculation is needed. We can anticipate that, due to the stronger action of the core potential, the effect of the ionic core in the $\varepsilon < 0$ case is more obvious than that for $\varepsilon > 0$.

In this paper we report on the extension of a method developed by Dando *et al*[12] that has already been used successfully in studying the spectra of non-hydrogen atoms in pure electric or pure magnetic fields. First, we introduce a new scaling property and a concept of the effective Planck constant. Then we employ an extended closed-orbit method to search all closed orbits and calculate the recurrence spectra of the lithium Rydberg atom with the quantum numbers n=37–46 and m=0 in the parallel electric and magnetic fields. Finally we compare them with those

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of hydrogen obtained by the standard closed-orbit theory. The results show clearly that the additional resonances can be explained in terms of core-scattered classical closed orbits.

2. Basic theory

The Hamiltonian with zero angular momentum for a lithium Rydberg atom in parallel electric and magnetic fields directed along the z-axis is, in cylindrical coordinates (ρ, z, θ) and atomic units, given by

$$H = \frac{1}{2}(p_\rho^2 + p_z^2) + \frac{1}{8}\gamma^2\rho^2 + Fz + V(r). \qquad (1)$$

Here we have adopted the short-range core potential[13]

$$\begin{aligned} V(r) =& V_{\text{Coulomb}}(r) + V_{\text{core}}(r) \\ =& -\frac{1}{r} - \frac{Z-1}{r}\mathrm{e}^{-r/a}\gamma \\ =& B(\text{T})/2.35 \times 10^5, \end{aligned} \qquad (2)$$

where $\gamma = B(\text{T})/2.35\times10^5$, Z=3 and a is a free length parameter determining the range of the core, which we chose to be 1/2.13 to obtain measured quantum defects.

The recurrence spectra calculations require knowledge of the connections between quantum structure and classical dynamics. Using the scaled-energy spectroscopic technique,[2] we can keep ε and $\tilde{F}$ constant while varying simultaneously the laser frequency and the strength of the external fields. Because the dynamic behaviours depend only on the scaling variables and the effective Planck constant, it is convenient to compare directly with experiment. Performing the transformation

$$\begin{aligned} &\tilde{r} = r\gamma^{2/3}, \quad \tilde{p} = p\gamma^{-1/3}, \quad \varepsilon = E\gamma^{-2/3}, \\ &\tilde{F} = F\gamma^{-4/3}, \quad \tilde{t} = t\gamma, \quad \hbar_{\text{eff}} = \gamma^{1/3}, \end{aligned} \qquad (3)$$

in Eq.(1), we obtain a scaled Hamiltonian $\tilde{H}$

$$\tilde{H} = \frac{1}{2}\tilde{p}^2 + \frac{1}{8}\tilde{\rho}^2 + \tilde{F}\tilde{z} - \frac{1}{\tilde{r}} - \frac{2}{\tilde{r}}\mathrm{e}^{-\bar{r}/\bar{a}} = \varepsilon, \qquad (4)$$

where $[\tilde{r}, \tilde{p}] = \mathrm{i}\hbar_{\text{eff}}$, $\hbar_{\text{eff}}$ is the effective Planck constant, and $\hbar_{\text{eff}}^{-1}$ is approximately equal to the main quantum number n in the situation of intense external fields.

The average oscillator-strength density consists of a smooth background term and an oscillation part $f(E)$ which involves a sum of over all allowed closed-orbit contributions[12]

$$f(E) = [-2(E - E_i)/\pi]\mathrm{Im}\sum_j N_j(-1)^m\sqrt{8\pi}y^*(\theta_f^j). \qquad (5)$$

Here $E - E_i$ is the electron energy with respect to the zero-field ionization limit of the atom E_i and only the final states with m=0 and odd l are excited. $Y(\theta)$ is the angular function for photoabsorption from the s state of the atom with π polarization of the laser, and its complex conjugate[14] takes the form

$$Y^*(\theta) = -B_{10}\mathrm{e}^{\mathrm{i}\pi_l}\sqrt{\frac{3}{4\pi}}\cos\theta\,\Psi_i. \qquad (6)$$

Following the suggestion of Clark and Taylor,[15] we employ the region-splitting method, which divides the space into three characteristic spatial regions according to the dynamics of the excited electron: the core region; the surrounding Coulomb region where the dynamics of the excited electron is predominantly determined by the Coulomb force of the ionic core; and the outer region where the influence of the external field is at least as important as the Coulomb force of the ionic core. To gain the coefficients N_j, we match the semiclassical wavefunction returning to the core with the asymptotic form of the incoming part of the scattered wavefunction, $\Psi_{\text{inc}}(r_0, \theta_f^j)$, obtained in the core and Coulomb regions. In the matching procedure, the coordinate values in both wavefunctions are taken on the surface of the sphere of a radius r_0 within the Coulomb region where both the semiclassical and quantum calculations are valid. For the laboratory strength of fields, any distance between 30–100 Bohr radii is acceptable. The crucial point is that the matching should take place at a radius large enough to use the semiclassical approximation but small enough to neglect the effect of the external field, so that the quantum defect solution still holds. The matched equation is

$$\begin{aligned} &\Phi_{\text{initial}}(r_0, \theta_i^j)A_j\exp\{\mathrm{i}(S_j - \alpha_j\pi/2) \\ =& N_j\,\Psi_{\text{inc}}(r_0, \theta_f^j). \end{aligned} \qquad (7)$$

The left-hand side of this equation is a semiclassical wavefunction, where $\Phi_{\text{initial}}(r_0, \theta_i^j)$ is the initial outgoing wavefunction on the spherical surface of the sphere of radius r_0. The amplitude of the wavefunction, A_j, contains the information about the stability of the trajectory and is given by Eq.(6) of Ref.[12]

$$A_j(r, \nu) = \left|\frac{J_2(0, \theta_i^j)}{J_2(t, \theta_i^j)}\right|^{1/2}\left|\frac{r_0^2\sin\theta_i^j}{r\sin\nu}\right|^{1/2}, \qquad (8)$$

where the two-dimensional Jacobian, $J_2(t, \theta_i) = \dfrac{\partial(r, \theta)}{\partial(t, \theta_i)}$, can be calculated from the neighbouring trajectories.

The classical action $S_j = \int \boldsymbol{p}_j \cdot \mathrm{d}\boldsymbol{q}_j$ is accumulated along the jth trajectory, and the Maslov index, α_j, is an integer that equals the number of caustics and foci encountered by the closed trajectory. Ψ_{inc} is given by Delos and co-workers.[5,14]

In the case of hydrogen, the initial outgoing Coulomb wave at the boundary r_0 is used as a starting point for constructing the semiclassical wavefunction

$$\Phi_{\mathrm{initial}}(r_0, \theta_i^j) = \Phi_{\mathrm{out}}(r_0, \theta_i^j). \tag{9}$$

The calculated N_j is then proportional to $A_j \mathrm{e}^{\mathrm{i}(S_j - \alpha_j \pi/2)}$. The jth closed orbit results in an oscillation with a frequency S_j and an amplitude A_j in the absorption spectrum. It is noticed that the Coulomb-scattered waves that arise from the hydrogenic nucleus are strongly backwardly focused, which result in the repetition of the trajectory.[12]

For a non-hydrogen atom, the scattered quantum wavefunction consists not only of an incoming part returning to the core and a Coulomb part scattered back in the direction of the incoming trajectory, but also of an additional outgoing core-scattered wave

$$\Psi_{\mathrm{return}}(r, \theta) = \Psi_{\mathrm{inc}}(r, \theta) + \Psi_{\mathrm{Coulomb}}(r, \theta) + \Psi_{\mathrm{core}}(r, \theta). \tag{10}$$

The effect of the core-scattered wave is to redistribute its amplitude among all other closed orbits. As the electron returns to the nucleus, it is scattered by the atomic core potential in an arbitrary direction, which may be close to the starting angle of another hydrogenic closed orbit. Such a core scattering may be repeated several times until the electron returns to the origin.

As the semiclassical wavefunction is constructed, the core-scattered outgoing wavefunction must be added to the starting point so as to incorporate the effect of the core-scattered waves in the sum

$$\Phi_{\mathrm{initial}}(r, \theta_i^j) = \Phi_{\mathrm{out}}(r_0, \theta_i^j) + \sum_k N_k \Psi_{\mathrm{core}}^k(r_0, \theta_0^j). \tag{11}$$

Finally, we obtain the expression for N_j

$$N_j = \left\{ D_j(\theta_i^j) + \sum_{k,p} N_k \beta_j^{k,p}(\theta_i^j) \right\} \times \sum_n A_j^n \exp\{\mathrm{i}(S_j^n - \alpha_j^n \pi/2 - \phi_j)\}, \tag{12}$$

where $\phi_j = \pi/2$ for $\theta_i^j = 0$ or $\theta_i^j = \pi$, and $\phi_j = 3\pi/4$ for other values of θ_i^j. The core-scattered contributions can be written as[12]

$$\begin{aligned}
\beta_j^{k,p}(\theta_i^j) =& \mathrm{i}2^{7/4}\pi^{3/2} \sum_{l \geq |m|}^{\infty} Y_{lm}^*(\theta_f^{k,p}, 0) \times Y_{lm}(\theta_i^j, 0)(\mathrm{e}^{2\pi \mathrm{i}\mu_l} - 1) \\
& (\theta_i^j \neq 0, \pi), \\
\beta_j^{k,p}(\theta_i^j) =& 2^{3/2}\pi \sum_l Y_{10}^*(\theta_f^{k,p}, 0) \times Y_{10}(\theta_i^j, 0)(\mathrm{e}^{2\pi \mathrm{i}\mu_l} - 1) \\
& (\theta_i^j = 0, \pi),
\end{aligned} \tag{13}$$

and the quantum defect has the following form[13]

$$\mu_l = \frac{\sqrt{2}}{\pi} \lim_{R \to \infty} \left[\int_{r_{0,c}}^{R} \sqrt{-V(r) - \frac{(l+1/2)^2}{2r^2}} \mathrm{d}r - \int_{r_0}^{R} \sqrt{\frac{1}{r} - \frac{(l+1/2)^2}{2r^2}} \mathrm{d}r \right]. \tag{14}$$

In the calculation, we have taken μ_0=0.40 and μ_1=0.06 while D_j and A_j^n [12] read as

$$\begin{aligned}
D_j &= 2^{9/4}\pi^{3/2} y(\theta_i^j), \quad (\theta_i^j \neq 0, \pi) \\
D_j &= -\mathrm{i}4\pi y(\theta_i^j), \quad (\theta_i^j = 0, \pi) \\
A_j^n &= r_0^{1/4} \sin\theta_f^{j,n} A_j^n, \quad (\theta_i^j \neq 0, \pi) \\
A_j^n &= r_0^{1/2} A_j^n, \quad (\theta_i^j = 0, \pi).
\end{aligned} \tag{15}$$

The first term on the right-hand side of Eq.(12) accounts for the oscillation caused by the hydrogenic orbit, and the second term indicates the contribution of all orbits that the electron is scattered from the kth orbit into the jth orbit by the ionic core. The superscripts n and p denote the numbers of repetitive time of the orbits.

3. Conclusion and discussion

From the well-defined Hamiltonian of Eq.(1), we have found 31 stable closed orbits in the angular range between 0° and 180° at $E = -4.3\mathrm{cm}^{-1}$, F=18.7V/cm and B=3.5T. The values of the corresponding scaled variables are $\varepsilon = -0.03$, $\tilde{F}$=0.01 and $\gamma^{-1/3} = 37.4 - 45.5$. Some orbits are shown in Fig.1, where the vertical and horizontal axes represent $z(t)$ and $\rho(t)$, respectively.

We have calculated the recurrence spectra of the lithium atom and compared them with those of hydrogen obtained by the standard closed-orbit theory. Figures 2(a) and (b) show the recurrence spectra of

lithium and hydrogen, respectively. Here the vertical and horizontal axes represent the modulus of the Fourier transform and the scaled classical action $\tilde{S}$, respectively. In these figures the position of each peak in the recurrence spectra is associated with a closed orbit, and the amplitude of the peak is proportional to A_j. The core-scattered contributions are the peaks located at the sums $\tilde{S}_j + \tilde{S}_k$ and the Coulomb-scattered contributions appear at the location of $n\tilde{S}_j$.

There are substantial differences between the re-

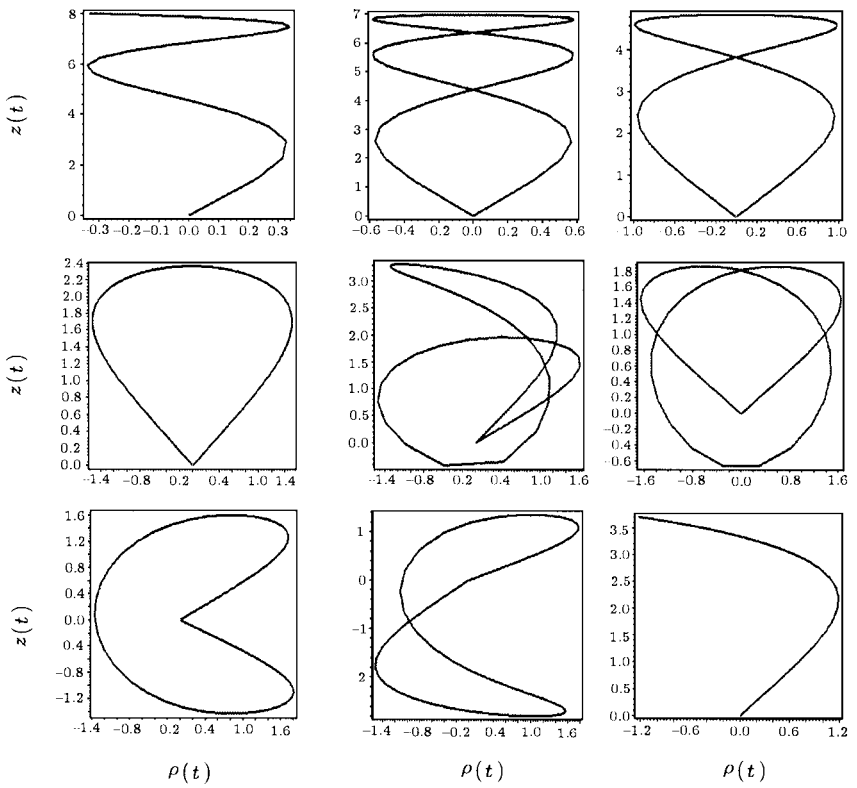


Fig.1. Closed orbits in cylindrical coordinates.

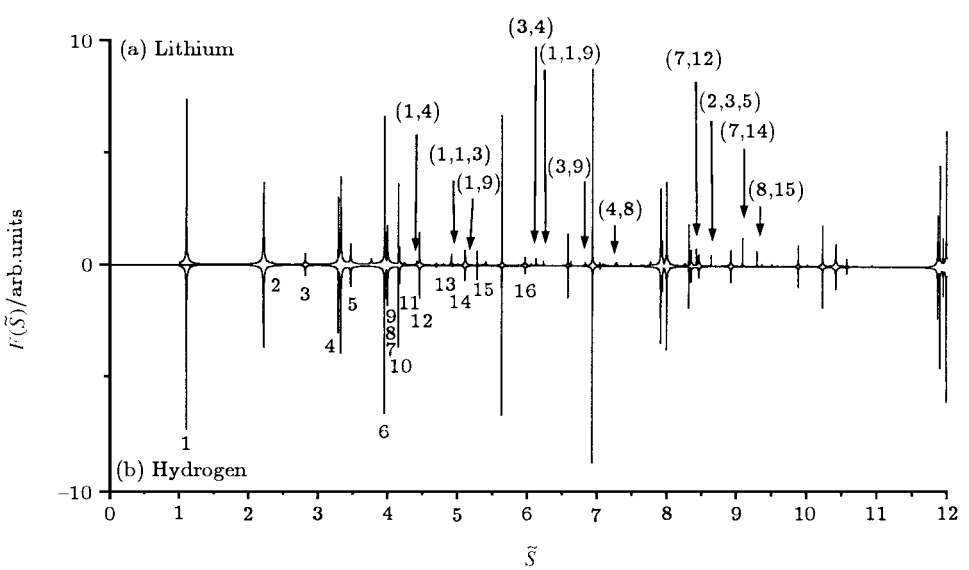


Fig.2. Fourier transform of the photoabsorption spectra for lithium in the parallel electric and magnetic fields with a constant scaled energy $\varepsilon = -0.03$ and a scaled electric field $\tilde{F}$=0.01. The value of $\gamma^{-1/3}$ varies in the range of 37.4–45.5. (a) The recurrence spectra of lithium including the ionic core-scattered terms. (b) The recurrence spectra of hydrogen based on the standard closed-orbit theory.

currence spectra of the lithium and hydrogen atoms due to the effect of the ionic core. Figure 2 shows that at the positions indicated by the arrows, peaks are not associated with any single classical closed orbits, which do not exist in hydrogen. The significant distinctions between the two spectra appear when $\tilde{S} > 4$. The larger the action is, the more additional peaks there are in the lithium spectra. This is consistent with the proliferation of closed orbits in chaotic systems.

The single integers in Fig.2(b) indicate the primary orbits. The double integers in Fig.2(a) characterize the combinations of orbits corresponding to the electron encountering one-time core scattering. The triple integers indicate the peaks arising from double core scattering, which is tantamount to a combination of three orbits. For example, the peak marked by (1,4) and located at $\tilde{S}_1 + \tilde{S}_4 \approx 4.415$ means the combination of orbits 1 and 4 caused by one-time core scattering. The peak marked by (2,3,5) and located at $\tilde{S}_2 + \tilde{S}_3 + \tilde{S}_5 \approx 8.63$ denotes the contributions of orbits 2, 3 and 5, obtained from two-time core scattering. Theoretical analysis indicates that the influences of the higher-order core scattering can be ignored. There are also some peaks which cannot be explained by using the closed-orbit theory. These correspond to the ghost peaks.[16]

By extending the closed-orbit theory and using the region-splitting method, we have presented the recurrence spectra of the lithium Rydberg atom in parallel electric and magnetic fields. It is demonstrated that the ionic core plays a very important role. The spectra presented here for $\varepsilon < 0$ are considerably different from those for $\varepsilon > 0$.[17] We have not accounted for the effects of diffraction and bifurcations here, and these need further study elsewhere.

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