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Semiclassical calculation of the recurrence spectra of He Rydberg atom in perpendicular electric and magnetic fields*

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Closed orbit theory is a semiclassical technique for explaining the spectra of Rydberg atoms in external fields. By developing the closed orbit theory from two degrees of freedom to three non-separable degrees of freedom, we calculated the recurrence spectra of He Rydberg atom in perpendicular electric and magnetic fields. The closed orbits in the corresponding classical system have also been obtained. Fourier transformed spectra of He atoms have allowed direct comparison between the resonance peaks and the scaled action values of closed orbits, whereas the nonhydrogenic resonance can be explained in terms of the new orbits created by the core scattering. The semiclassical result is in good agreement with the quantum spectra, which suggests that our method is correct.

Keywords: closed orbit theory, recurrence spectra, core scattering
PACC: 0547, 0365, 3290

1. Introduction

The effects of external electric and magnetic fields on atomic spectra is an important problem in atomic physics. Du and Delos's semiclassical closed orbit theory [1] introduced new ways of analyzing the spectra of atoms in strong external fields. However, up to now most researchers have concentrated on atoms in only pure electric or magnetic field or in parallel electric and magnetic fields.[1−11] Because these systems possess a cylindrical symmetry about the direction of the magnetic field, the angular motion perpendicular to the magnetic field can be separated, which reduces the problem to a two-dimensional one. As to the case of perpendicular electric and magnetic fields, the cylindrical symmetry is broken, and the Hamiltonian is nonseparable in three degrees of freedom, therefore, the theoretical treatment is more complicated. According to the closed orbit theory, each classical orbit that starts at the atom carries along with it a portion of the outgoing electron waves. If such an orbit returns to the nucleus, its companion wave interferes with the steady stream of outgoing waves, and this interference produces an oscillation in the absorption spectrum. The return of the electron and the resulting interference oscillation in the spectrum is called a recurrence. The more the orbits are focused on the atom, the stronger the recurrence. Motivated by non-hydrogenic resonance discovered in experimental scaled recurrence spectra of rubidium atoms,[12] we investigated the influence of the ionic core on the three-dimensional closed orbits and adopt a new model potential as given in Ref.[2], which includes not only the Coulomb interaction potential and the core-attractive potential, but also the exchange potential between the excited electron and other electrons. We found that the introduction of the core potential results in an immense increase of the number of closed orbits as compared with that of hydrogen atom. The novel orbits appear to be composed of two or more hydrogen orbits and are created through scattering by the core potential. In this paper, we develop the closed orbit theory from two-dimensional to three-dimensional one and calculate the recurrence spectra of He Rydberg atom in perpendicular electric and magnetic fields. The calculation of the recurrence spectra by using the closed orbit theory requires the numerical search for closed orbits starting and ending exactly at the nucleus. This leads to numerical difficulties because of the Coulomb singularity of the potential at the ori-

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gin. In order to remove this kind of singularity, we adopted the Kustaanheimo-Stiefel transformation,[13] which transforms the system from a three-dimensional one to a four-dimensional one. Our results are in good agreement with the quantum spectra, suggesting that our results are correct.

2. Theory and quantitative formula

2.1. Hamiltonian and scaled transformation

Since we are dealing with the He Rydberg atom, where only one electron is highly excited, while the ionic core remains in the ground state. We can simplify the system by considering the Hamiltonian of a single electron moving in core potential and external fields. Assuming the magnetic field pointing in the z-axis and the electric field pointing in the x-direction, then the Hamiltonian of the excited electron of He Rydberg atom is described in atomic units as [14]:

$$H = \frac{1}{2}P^2 + \frac{\gamma}{2}l_z + \frac{1}{8}\gamma^2\rho^2 + Fx + V(r), \qquad (1)$$

where $\gamma = B/2.35 \times 10^5$ (B is the magnetic field strength), F is the electric field strength, $V(r)$ is the model potential.[2]

The classical motion of Hamiltonian (Eq.(1)) exhibits an important scaling property. If we transform the variables according to $\tilde{r} = r\gamma^{2/3}$, $\tilde{P} = P\gamma^{-1/3}$, $\varepsilon = E\gamma^{-2/3}$, $f = F\gamma^{-4/3}$, $\tilde{t} = t\gamma$, then the classical motion is governed by the scaled Hamiltonian:

$$\tilde{H} = \frac{1}{2}\tilde{P}^2 + \frac{1}{2}\tilde{l}_z + \frac{1}{8}\tilde{\rho}^2 + f\tilde{x} + V(\tilde{r}) = \varepsilon. \qquad (2)$$

From Eq.(2), we find that the scaled Hamiltonian does not depend on the energy E and field strength separately but only on the scaled energy ε and the scaled electric field f, thus remove a parameter B.

2.2. Closed orbits search

In order to solve numerically the equations of motion and remove the Coulomb singularity generated by Hamiltonian (2), we implement the Kustaanheimo-Stiefel transformation [13] which transform the system from a three-dimensional to a four-dimensional one. Removal of the Coulomb singularity makes the numerical calculation very stable and an orbit launched from the origin can evolve quite a long time without loss of numerical accuracy. Due to the increased dimensionality of the phase space involved, the number of the closed orbits is large and a search for the closed orbits is by no means trivial. The difficulty lies in the fact that for a three degrees of freedom system, one has to scan two independent parameters (θ, φ) to locate the closed orbits, while in the pure magnetic field only one parameter θ needs to be scanned. We use the fifth-order Cash-Karp Runge-Kutta method to integrate the equations of motion. In previous works,[6,14] the core potential is to be switched-off at the beginning and the end of the orbits. In contrast, in our calculation, we always take into account the core potential. Therefore, this treatment is more complete and more general. By integrating the Hamiltonian equations of motion, we found the closed orbits with scaled actions smaller than 10 at $\varepsilon = -0.6$, f=0.06. For each closed orbit, we evaluate the classical action, the classical amplitude as well as the Maslov index. Some of the orbits are given in Fig.1.

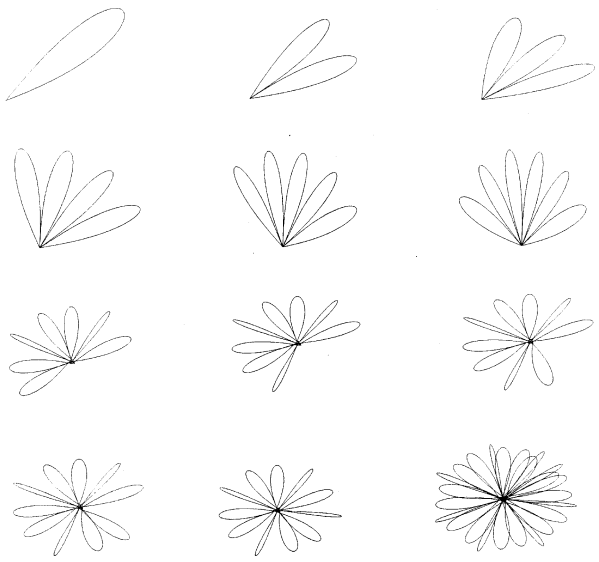


Fig.1. Some closed orbits of Rydberg He atom in perpendicular electric and magnetic fields with different stating angles at $\varepsilon = -0.6$, $f = 0.06$.These orbits are drawn in the x-y plane. (Horizontal axis: x; Vertical axis: y).

2.3. Oscillator strength density

According to the basic ideas presented by Du and Delos,[1] the exact quantum expression for the oscillator strength density in dipole transitions of an initial state ψ_i to final states at energy E can be expressed as

$$f(E) = -\frac{2}{\pi}(E - E_i)\mathrm{Im}\int \mathrm{d}^3\boldsymbol{r}(D\psi_i)^*(\boldsymbol{r})\psi(\boldsymbol{r}), \qquad (3)$$

where D is the dipole operator, ψ_i is the initial wave function and $\psi(\boldsymbol{r})$ is a solution to the inhomogeneous Schrödinger equation:

$$\psi(\boldsymbol{r}) = \psi^{\mathrm{out}}(\boldsymbol{r}) + \psi^{\mathrm{ret}}(\boldsymbol{r}). \qquad (4)$$

Here $\psi^{\rm out}$ is the outgoing wave and $\psi^{\rm ret}$ is the returning wave.

In order to obtain the outgoing and returning electron wavefunctions, we divide the whole space into two regions: the inner region and the outer region. In the inner region the influence of the external fields can be neglected. The outgoing wave can be seen as if no external fields were present [6]

$$\psi^{\rm out}(r,\theta,\varphi) = \sum_{l=|m|}^{\infty} \int Y^*_{l,m}(\theta',\varphi') g_l^{E=0}(r,r') Y_{l,m}(\theta,\varphi) D\psi_i(\boldsymbol{r}') {\rm d}\boldsymbol{r}', \tag{5}$$

with $g_l^{E=0}(r,r')$ being the radial Green's function,

$$g_l^{E=0}(r,r') = \frac{2R_l^{0,{\rm reg}}(r_<)R_l^{0,{\rm out}}(r_>)}{r'^2 w[R_l^{0,{\rm reg}}(r'), R_l^{0,{\rm out}}(r')]}, \tag{6}$$

where $r_< = \min\{r,r'\}, r_> = \max\{r,r'\}$ and w is the Wronskian determinant. The functions $R_l^{0,{\rm reg}}$ and $R_l^{0,{\rm out}}$ are the regular and outgoing solutions to the radial part of the Schrödinger equation at E=0. By using the asymptotic forms of them,[15] we obtain the outgoing wave

$$\psi^{\rm out}(r,\theta,\varphi) = -\pi {\rm i} \sum_{l=|m|}^{\infty} R_l^{0,{\rm out}}(r) Y_{l,m}(\theta,\varphi) B_{lm}, \tag{7}$$

where

$$B_{lm} = \int {\rm d}\boldsymbol{r}' D\psi_i(r') R_l^{0,{\rm reg}}(r') Y^*_{l,m}(\theta',\varphi').$$

In the outer region where the external fields become important, the outgoing wave $\psi^{\rm out}$ can propagate semiclassically:

$$\psi^{\rm semi}(\boldsymbol{r}) = \sum_k \sqrt{|J_k(0,\theta^k,\varphi^k)/J_k(t_k,\theta^k,\varphi^k)|} \times {\rm e}^{{\rm i}[s_k - (\pi/2)\mu_k]} \psi^{\rm out}(\boldsymbol{r}_i^k). \tag{8}$$

The sum includes all the classical trajectories with energy E starting radially on the initial sphere; $J(t,\theta,\varphi)$ is the Jacobian determinant, t_k is the time when the trajectory reaches the point (r,θ,φ), S_k is the action and μ_k is the Maslov index.

Due to the influence of the external fields, some of the trajectories will turn back. Let $S_0^{(k)}$ and $\mu_0^{(k)}$ be the action and Maslov index of the closed orbit respectively. Using the expressions for the Jacobian determinant as given in Ref.[14], we obtain the semiclassical approximation for $\psi^{\rm ret}$:

$$\begin{aligned}\psi^{\rm ret}_{\rm semi}(r) =& \sum_k -64\pi^2 \frac{{\rm e}^{{\rm i}(S_0^{(k)} - (\pi/2)\mu_0^{(k)})}}{\sqrt{|M_0^{(k)}|}} \\ &\times \sum_{l',m'} (-1)^{l'} {\rm e}^{{\rm i}\delta_{l'}} B_{l'm'} Y_{l'm'}(\theta_i^{(k)}, \varphi_i^{(k)}) \\ &\times \sum_{l,m} (-1)^l Y^*_{lm}(\theta_f^{(k)}, \varphi_f^{(k)}) \\ &\times Y_{lm}(\theta,\varphi) {\rm e}^{{\rm i}\delta_l} R_l^{0,{\rm reg}}(r).\end{aligned} \tag{9}$$

Here, $(\theta_i^{(k)},\ \varphi_i^{(k)})$ are the starting angles of the kth closed orbit and $(\theta_f^k, \varphi_f^k)$ are the returning angles of the kth closed orbit, δ_l is the phase shift caused by the core potential and $M_0^{(k)}$ is related to the stability matrix of the closed orbit.[14]

By substituting the above functions into Eq.(3), we can obtain the oscillator strength density:

$$\begin{aligned}f(E) =& -\frac{2}{\pi}(E-E_i){\rm Im}\int {\rm d}^3\boldsymbol{r} (D\psi_i)^*(\boldsymbol{r}) \psi^{\rm out}(\boldsymbol{r}) \\ &-\frac{2}{\pi}(E-E_i){\rm Im}\int {\rm d}^3\boldsymbol{r} (D\psi_i)^*(\boldsymbol{r}) \psi^{\rm ret}(\boldsymbol{r}) \\ =& f^0(E) + f^{\rm osc}(E).\end{aligned} \tag{10}$$

The first term is a slowly varying background of the spectra, which can appear without the external fields. The second part, which is composed of contributions from the returning closed orbits, leads to oscillations in the absorption spectra. Inserting $\psi^{\rm ret}$ into the above equation, we have

$$\begin{aligned}f^{\rm osc}(E) =& \sum_k 64\pi \frac{{\rm e}^{{\rm i}(S_0^{(k)} - (\pi/2)\mu_0^{(k)})}}{\sqrt{|M_0^{(k)}|}} \\ &\times \sum_{l',m'} (-1)^{l'} {\rm e}^{{\rm i}\delta_{l'}} B_{l'm'} Y_{l'm'}(\theta_i^{(k)}, \varphi_i^{(k)}) \\ &\times \sum_{l,m} (-1)^l Y^*_{lm}(\theta_f^{(k)}, \varphi_f^{(k)}) B_{lm} {\rm e}^{{\rm i}\delta_l}.\end{aligned} \tag{11}$$

For the He Rydberg atom, the electron is excited to the 2s state, thus the initial state wave-function is: $\psi_i(r,\theta,\varphi) = R_{20}(r)Y_{00}(\theta,\varphi)$. If we use the x-polarized light, then the nonvanishing coefficients of B_{lm} are B_{11} and B_{1-1} with the relation $B_{11} = -B_{1-1}$. In this case, the oscillating part of the absorption spectra is

$$\begin{aligned}f^{\rm osc}(E) =& B_{11} 64\pi \\ &\times \sum_k \frac{\sin\theta_i^{(k)} \sin\theta_f^{(k)} \cos\varphi_i^{(k)} \cos\varphi_f^{(k)}}{\sqrt{|M_0^{(k)}|}} \\ &\times \sin\left[S_0^{(k)} - \frac{\pi}{2}\mu_0^{(k)} + 2\delta_1\right].\end{aligned} \tag{12}$$

2.4. Recurrence spectra

Recurrence spectra are the Fourier transformation of the photoabsorption spectra. After the scaled transformation $\tilde{S}_0^{(k)} = \gamma^{1/3} S_0^{(k)}$ and $\tilde{M}_0^{(k)} = \gamma^{2/3} M_0^{(k)}$, we obtain

$$f^{\rm osc}(z) = \frac{1}{z} \sum_k A_k \sin\left[\tilde{S}_0^{(k)} z - \frac{\pi}{2}\mu_0^{(k)} + 2\delta_1\right], \quad (13)$$

where $z = \gamma^{-1/3}$ and

$$A_k = B_{11} 64\pi \frac{\sin\theta_i^{(k)} \sin\theta_f^{(k)} \cos\varphi_i^{(k)} \cos\varphi_f^{(k)}}{\sqrt{|\tilde{M}_0^{(k)}|}}.$$

The Fourier transformed recurrence spectra can be calculated in the interval $[z_1,\ z_2]$. With $\bar{z} = 1/2(z_1 + z_2)$, $\Delta z = z_2 - z_1$ and approximate $1/z$ by $1/\bar{z}$, we obtain the recurrence spectra:

$$F(\tilde{S}) = \frac{1}{\bar{z}} \sum_k A_k \frac{\sin[\Delta z(\tilde{S}_0^{(k)} - S)]}{(\tilde{S}_0^{(k)} - S)} \times {\rm i}\exp\left[-{\rm i}\left(\tilde{S}_0^{(k)}\bar{z} - \frac{\pi}{2}\mu_0^{(k)} + 2\delta_1\right)\right] \cdot {\rm e}^{{\rm i}\bar{z}\tilde{S}}. \quad (14)$$

For a finite length Δz, $F(\tilde{S})$ are complex numbers. In our calculation, we take the absolute value of $F(\tilde{S})$.

3. Results and discussions

Figure 1 gives the recurrence spectra of atoms in perpendicular electric and magnetic fields at $\varepsilon = -0.6$, $f = 0.06$, $z = \gamma^{-1/3}$ in the range of 60–120. Figure 2(a) is the recurrence spectra of He atoms, and (b) is the recurrence spectra of H atoms. Each peak is associated with a closed orbit. Some of the orbits are shown in Fig.1. Due to the core scattering, the orbits possess some repetition. With the change of the starting angles, the orbits consist of more and more "loops". For example, at the starting angles ($\theta = 90.4°$, $\varphi = 9.12°$), there are only two "loops" while at the starting angles ($\theta = 91.2°$, $\varphi = 5.02°$), there are five "loops". From Fig.2, we can see that for smaller $\tilde{S}$, the structures of spectra for He and H atoms are analogues. As $\tilde{S}$ increases, there are some new peaks (as indicated by arrows) appear in the spectra of He atoms. This can be interpreted as follows: in order to calculate the recurrence spectra of He atoms, we have taken into account the effect of the core potential. The main effect of the core potential lies in the creation of a huge number of new closed orbits. These new orbits appear to be composed of two or more hydrogenic orbits originating from the scattering of hydrogenic orbits by the core potential. This can be clearly seen from the closed orbits of Fig.1. Figure 3 displays the semiclassical recurrence spectra of Rydberg He atoms in perpendicular external fields at $\varepsilon = -0.45$, $f = 0.02$. In order to compare the results between the semiclassical and the quantum spectra, we calculate the recurrence spectra of He Rydberg atoms in perpendicular electric and magnetic fields by using quantum mechanical method as given in Ref.[16]. Figure 3 (a) is the quantum calculation result; (b) is the semiclassical calculation result. It is apparent from this figure that the quantum recurrence spectrum retains the pronounced peak structures as in the semiclassical spectra. Even for the largest action considered, the quantum and semiclassical recurrence spectra agree well quantitatively, suggesting that our method is valid.

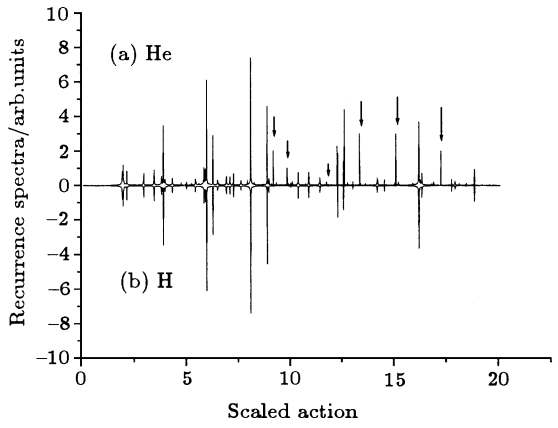


Fig.2. The recurrence spectra of atoms in perpendicular electric and magnetic field at $\varepsilon = -0.6$, $f = 0.06$. (a)semiclassical calculation for the He atom; (b)semiclassical calculation for the H atom.

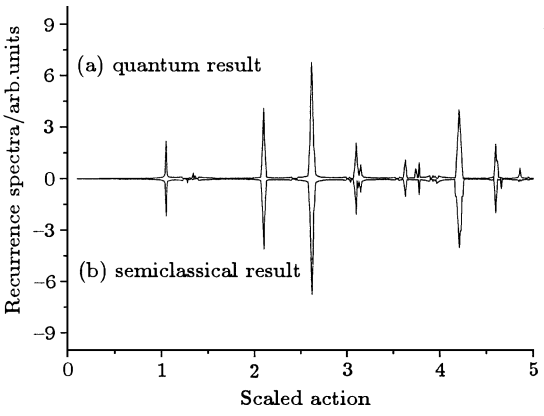


Fig.3. The recurrence spectra of Rydberg He atom in perpendicular electric and magnetic field at $\varepsilon = -0.45$, $f = 0.02$. (a) Quantum calculation result; (b) Semiclassical calculation result.

4. Conclusion

By extending the closed orbit theory to three nonseparable degrees of freedom, we have calculated the recurrence spectra of He Rydberg atoms in perpendicular electric and magnetic fields. The non-Coulombic core potential has been considered in the classical trajectory calculations, and some core-scattered nonhydrogenic closed orbits have been discovered. The new peaks in the recurrence spectra of He atoms have been considered as effects caused by the core scattering of returning waves at the ionic core, from which we gain a deep understanding of the highly excited Rydberg atoms in perpendicular electric and magnetic fields. Because these atoms represent a system nonseparable in three degrees of freedom which behaves classically chaotic. Our results are in good qualitative agreement with the quantum spectra. Presently no experiments on the recurrence spectra of He atoms in perpendicular external fields have been carried out. We hope that our calculations may be helpful in future measurements.

References

[1] Du M L and Delos J B 1988 *Phys. Rev.* A **38** 1896, 1913
[2] Wang D H and Ding S L 2003 *Phys. Rev.* A **68** 023405
[3] Wang D H, Ding S L and Lin S L 2003 *J. Phys.* B **36** 4225
[4] Peters A D, Jaffe C and Delos J B 1994 *Phys. Rev. Lett.* **73** 2825
[5] Dando P A *et al* 1995 *Phys. Rev. Lett.* **74** 1099
[6] Hupper Bruno, Main Jong and Wunner Gunter 1996 *Phys. Rev.* A **53** 744
[7] Dando P A *et al* 1996 *Phys. Rev.* A **54** 127
[8] Lin S L *et al* 2002 *Chin. Phys. Lett.* **19** 29
[9] Wang D H and Ding S L 2004 *Chin. Phys.* **13** 30
[10] Song X H and Lin S L 2003 *Acta. Phys. Sin.* **52** 1611 (in Chinese)
[11] Song X H *et al* 2002 *Chin. Phys.* **11** 656
[12] Raithel G *et al* 1994 *J. Phys.* B **27** 2849
[13] Kustaanheimo P and Stiefel E 1965 *J. Ang. Math.* **218** 204
[14] Weibert K, Main J and Wunner G 1998 *Ann. Phys.* **268** 172
[15] Gao J and Delos J B 1992 *Phys. Rev.* A **46** 1455
[16] Rao J G and Taylor K T 2002 *J. Phys.* B **35** L1