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The closed-orbit and the photoabsorption spectra of the Rydberg hydrogen atom between two parallel metallic surfaces*

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Using the closed orbit theory, we study the classical motion and calculate the photoabsorption spectra of Rydberg hydrogen atom between two parallel metallic surfaces. The results show that the metallic surfaces have a significant effect on the photoabsorption process. When the distances between the hydrogen atom and the two metallic surfaces are close to a critical value d_c, the number of the closed orbits is the greatest. When the distance larger or smaller than d_c, the number of the closed orbits decreases and the absorption spectra are shown to exhibit a damping oscillation. This work is an interesting new application of closed-orbit theory and is of potential experimental interest.

Keywords: closed orbit, photoabsorption spectra, metallic surface
PACC: 3450D, 0365S

1. Introduction

The photoabsorption properties of Rydberg atom near a metallic surface have attracted much attention in recent years. As a typical theoretical and experimental model system, it can simulate many dynamical effects of atoms in strong fields, such as Zeeman–Stark effect, diamagnetic effect, instantaneous van der Waals interaction, etc.[1−3] In this system, the metallic surface appears as an external perturbation to the electronic motion, with the atom–surface distance used as the parameter. Its classical motion is very complex. When the distance between the atom and the surface is very large, the problem of an atom near a metallic surface can be considered in the approximation of the so-called instantaneous van der Waals interaction;[3] regular and irregular motions merge with each other and chaos appears. As the atom–surface interaction is weak, the perturbation theory may be applied. In 1997, Simonovic[4] studied the classical chaos in the hydrogen atom near a metallic surface by analysing the Poincaré surface of section of this system. Ganesan and Taylor[5] have studied the quantum spectra of Rydberg hydrogen atom near a metallic surface. Salas and Simonovic[6] have presented a quantum mechanical, classical and semi-classical studies of the energy spectrum of a Rydberg hydrogen atom in the instantaneous van der Waals potential. In our previous work,[7] we have studied the photoabsorption and recurrence spectra of Rydberg hydrogen atom near a metallic surface. But for the photoabsorption spectra of Rydberg hydrogen atom between two parallel metallic surfaces, no relevant analysis and calculation have been found.

Following Gutzwiller's original periodic-orbit theory for the density of states,[8] the closed-orbit theory (COT) was developed to study the oscillations in the complicated absorption spectrum for atoms, molecule and negative ions in strong external fields.[9−12] It has been demonstrated in the closed-orbit theory that each classical electron closed orbit that begins and ends near the nucleus contributes an oscillation to the average oscillator strength. Recently, this theory has been successfully applied to many other systems. In Ref.[12], Du *et al*[13] have used this theory to study the spontaneous emission of atoms in the environments. Yang *et al*[14] have investigated the photodetachment of H^- near an elastic interface by using the closed orbit theory. In this paper, we firstly develop the closed-orbit theory to calculate the absorption spectra of Rydberg hydrogen atom in the presence of two metallic surfaces. Considering the interaction of the Rydberg

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atom with the metallic surfaces, the photoabsorption process of this system is quite similar to the case of the Rydberg atom in strong external fields. The results show that the metallic surfaces have a significant effect on the photoabsorption process. The oscillations in the absorption spectra are related to the classical electron closed orbits: orbits going out from the atom and returning to the atom. This is another example to show the use of the closed orbit theory and this study provides a visual insight into the dynamical behaviour of Rydberg atom in the presence of metallic surfaces.

2. The classical dynamics of Rydberg hydrogen atom near two parallel metallic surfaces

The schematic of the system is shown in Fig.1. A hydrogen atom nucleus sits at the origin and a z-polarized laser is applied to the photoabsorption process. Two parallel metallic surfaces perpendicular to the z-axis are located at $z = -d_1$ and $z = d_2$ separately. So the excited electron is bounced back by the metallic surfaces. Considering the interaction between the hydrogen atom and the metallic surfaces, the physical picture of the closed orbit theory can be described as follows: assume that a hydrogen atom is irradiated by a continuous laser, it would be excited from the ground or a low excited state to a high Rydberg state. When the atom absorbs a photon, the electronic wave representing the electron is created near the atom. This wave propagates outwards from the atom in all directions. In the region that is close to the nucleus, the Coulomb interaction between the electron and the nucleus dominates and the influence of the metallic surface could be neglected. With the increase of the distance, the electrical image potential induced by the metallic surface becomes more important. For distances greater than $50a_0$ (a_0 is the Bohr radius), the electron waves travel along the classical trajectories under the action of both the Coulomb potential and the atom–surface interaction potential. The waves propagation can be approximated by semiclassical mechanics and they are correlated with outgoing classical trajectories, which are determined by the classical Hamiltonian canonical equations. When a group of trajectories hit the metallic surface, some of the associated waves will turn back due to the action of the combined external potential. The waves turning back to the nucleus would interfere with the outgoing waves, thus giving rise to the oscillation in the absorption spectra.

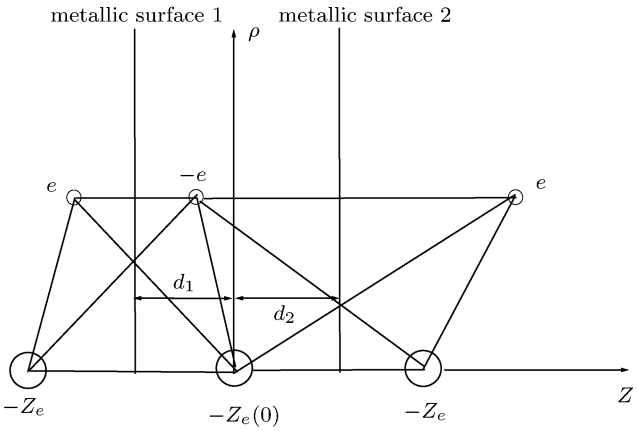


Fig.1. The pictorial representation of an atom and its images near two parallel metallic surfaces. A hydrogen atom nucleus sits at the origin and two parallel metallic surfaces perpendicular to the z-axis are located at the $z = -d_1$ and $z = d_2$.

According to the electrical image method,[15] the Hamiltonian of the electron in hydrogen atom near two parallel metallic surfaces is in following form (atomic units are used):

$$H = \frac{p^2}{2} - \frac{1}{r} + \frac{1}{\sqrt{\rho^2 + (2d_1 + z)^2}} - \frac{1}{4(d_1 + z)} + \frac{1}{\sqrt{\rho^2 + (2d_2 - z)^2}} - \frac{1}{4(d_2 - z)}, \quad (1)$$

in which $r = \sqrt{\rho^2 + z^2}$ and $p^2 = p_\rho^2 + l_z^2/\rho^2 + p_z^2$, the last four terms on the right-hand side of the above expression are referred to as image potential and induced by the metallic surfaces. In the above expression, the interaction between the hydrogen atom and metallic surfaces is approximated by the Coulomb interaction between the hydrogen atom and their direct images at the surfaces. The higher-order images are neglected. Owing to the cylindrical symmetry, the z-component of angular momentum is conserved and the φ-motion is separated from that in the (ρ, z) plane. Therefore, the system is reduced to a problem with two degrees of freedom. Here we consider the case of $l_z = 0$.

The classical motion of the excited electron is closely related to the excited state and the atom–surface distance parameters d_1 and d_2. If d_1 and d_2 are sufficiently large, the image potential could be regarded as a perturbation and the system is completely integrable. Define d_c as the critical value when the Coulomb term and the electronic image potential could be compared with each other: $d_c(E) =$

$1.4922/|E|$,[4] where $E = -1/(2n^2)$ that is the energy of the hydrogen atom in atomic units. If the hydrogen atom is excited to the state with principle quantum number $n = 20$, then $E = -1.25\times10^{-3}$, and $d_{\rm c} = 1193.76$.

In order to find the closed orbits of this system, we must integrate the Hamiltonian's motion equations. The main difficulty in integrating the equations of motion for the Hamiltonian (Eq.(1) is the Coulomb singularity at the origin. As a classical electron approaches the origin, its velocity becomes infinite, thereby the arbitrarily fine time steps are necessitated to be used in a numerical integration of the classical equations of motion. This difficulty can be surmounted easily by making a regularized transformation that removes the Coulomb singularity. For the case considered here, this can be achieved by transforming its coordinates to semi-parabolic coordinates (u, v), where $u^2 = r+z$, and $v^2 = r-z$, with conjugate momenta, $p_u = {\rm d}u/{\rm d}\tau$, and $p_v = {\rm d}v/{\rm d}\tau$, defined with respect to a rescaled time τ through the following way:[16]

$$\frac{{\rm d}\tau}{{\rm d}t} = \frac{1}{2r(t)} = \frac{1}{(u^2+v^2)}\,, \tag{2}$$

which results in a regularized Hamiltonian

$$\begin{aligned} h = {} & \frac{1}{2}(p_\mu^2 + p_v^2) - E(\mu^2+v^2) \\ & + \frac{\mu^2+v^2}{\sqrt{\mu^2v^2 + \Big(2d_1 + \dfrac{\mu^2-v^2}{2}\Big)^2}} - \frac{\mu^2+v^2}{4\Big(d_1 + \dfrac{\mu^2-v^2}{2}\Big)} \\ & + \frac{\mu^2+v^2}{\sqrt{\mu^2v^2 + \Big(2d_2 - \dfrac{\mu^2-v^2}{2}\Big)^2}} - \frac{\mu^2+v^2}{4\Big(d_2 - \dfrac{\mu^2-v^2}{2}\Big)} \\ = {} & 2\,. \end{aligned} \tag{3}$$

After this transformation, the Coulomb singularities have been removed. Therefore, we can integrate numerically the Hamiltonian motion equations for (u, v, p_u, p_v) of Eq.(3), namely

$$\dot{u} = \frac{\partial h}{\partial p_u}\,, \quad \dot{v} = \frac{\partial h}{\partial p_v}, \quad \dot{p}_u = -\frac{\partial h}{\partial u}, \quad \dot{p}_v = -\frac{\partial h}{\partial v}.$$

We use a standard fifth-order Runge–Kutta method to integrate the motion equations. The removal of Coulomb singularity makes the numerical calculation very stable and each orbit launched from the origin can evolve quite a long time without loss of numerical accuracy. By integrating the Hamiltonian motion equations, we find out all the closed orbits of Rydberg hydrogen atom near metallic surfaces with different atom–surface distance. For each closed orbit, we calculate the classical action $S_{\rm k}$, the period of the closed orbit $T_{\rm k}$, the amplitude $A_{\rm k}$ and the Maslov index $\mu_{\rm k}$. Some of the closed orbits are given in Fig.2.

3. The photoabsorption spectra of Rydberg hydrogen atom near metallic surfaces

According to the closed-orbit theory, the photoabsorption cross section is related to the oscillator-strength density ${\rm D}f(E)$ by[9]

$$\sigma(E) = \frac{2\pi^2}{c}{\rm D}f(E)\,. \tag{4}$$

The oscillator strength for dipole transitions from an initial state ψ_i to final states at energy E is

$${\rm D}f(E) = -\frac{2}{\pi}(E-E_i){\rm Im}\langle {\rm D}\psi_i|\hat{G}^+|{\rm D}\psi_i\rangle, \tag{5}$$

where $\hat{G}^+$ is the outgoing Green function, and D is the dipole operator, which equals the projection of the electron coordinate onto the direction of the polarization of the laser field. If we consider the z-polarized light, then ${\rm D} = z = r\cos\theta$.

Equation (5) has the following physical meaning: the initial state ψ_i is modified by the dipole operator related with the incident laser field to become the source wavefunction; the Green function propagates these waves outward to become the outgoing waves; and finally the waves overlap with the source wave to give the absorption spectrum.

The Green function in Eq.(5) propagates the 'source function' from the source point to the field point along two or more paths. The first path is associated with waves propagating from the source point to the field point without ever leaving the atomic region. This is the direct path. Additionally, there are waves propagating outward from the source point and entering the external region where they are bounced back by the metallic surface and return to the vicinity of the atomic core where they arrive at the field point. Therefore, the Green function in Eq.(5) can be divided into a direct part and a returning part as follows:

$$\begin{aligned} \hat{G}^+|{\rm D}\psi_i> &= \hat{G}^+_{\rm dir}|{\rm D}\psi_i\rangle + \hat{G}^+_{\rm ret}|{\rm D}\psi_i\rangle \\ &\equiv \psi_{\rm dir} + \psi_{\rm ret}\,. \end{aligned} \tag{6}$$

Accordingly, the oscillator-strength density can be split into two parts:

$${\rm D}f(E) = {\rm D}f_0(E) + {\rm D}f_1(E)\,. \tag{7}$$

The first term on the right-hand side of the above expression is the smooth background term, which is precisely the oscillator strength density that would be obtained without any external field as follows:[9]

$$\mathrm{D}f_0(E) = -4E_i \sum\nolimits_{l'} |b^i_{l'm} I(n,l,l')|^2 \,, \tag{8}$$

where $b^i_{l'm}$ is the coefficient of the partial-wave expansion and $I(n,l,l')$ is the radial dipole integral involving the initial state and the regular zero-energy coulomb wavefunction.[9]

The second part is the contribution of the returning wave

$$\mathrm{D}f_1(E) = -\frac{2}{\pi}(E-E_i)\mathrm{Im}\langle \mathrm{D}\psi_i|\psi_{\mathrm{ret}}\rangle \,, \tag{9}$$

where ψ_{ret} is the returning wave, which overlaps with the outgoing source wave to give the interference pattern in the absorption spectrum. We use the semiclassical approximation to construct the returning wave. In the vicinity of the nucleus, the influence of the metallic surface is very small, the orbits behave like the regular Keplerian orbits and each closed orbit will be surrounded by an ensemble of nearby rays. The ensemble of rays is the family of trajectories that initially are close enough to the closed orbit and each trajectory in the family returns crossing the final sphere almost simultaneously. Associated with this pencil of trajectories is a wave function, which is given by[9]

$$\psi^{mk_m}_{\mathrm{ret}} = \mathrm{e}^{\mathrm{i}m\phi} A_2^{mk_m}(r,\theta) \times \exp[\mathrm{i}S_2^{mk_m} - \mu^{mk_m}\pi/2] \times \frac{r_{\mathrm{b}}}{r}\left|\frac{\sin\theta_i^{mk_m}}{\sin\theta}\right|^{1/2} \times \psi^m_{\mathrm{out}}(r,\theta) \,. \tag{10}$$

In the vicinity of the nucleus, the full returning wave ψ_{ret} is the sum of contributions arising from each closed orbit:

$$\psi_{\mathrm{ret}}(r,\theta,\phi) = \sum_{m,k_m} \psi^{mk_m}_{\mathrm{ret}}(r,\theta,\phi) \,. \tag{11}$$

Substituting the above formula into Eq.(9), we have the oscillating part of the oscillator-strength density:

$$\mathrm{D}f_1(E) = \sum_{k_m} C_{mk_m}(E)\sin(T_{mk_m}E + \Delta_{mk_m}) \,. \tag{12}$$

Each oscillatory term arises from a closed orbit labelled with k_m. T_{mk_m} is the period of the kth closed orbit. The amplitude and phase constants, C_{mk_m} and Δ_{mk_m} in the sum of sinusoidal oscillation terms, are given by the following formula:

$$C_{mk_m} = (-E_i)2^{19/4}\pi^{3/2} r_{\mathrm{b}}^{-1/4} (\sin\theta_i^{mk_m}\sin\theta_f^{mk_m})^{1/2} \times A_{mk_m} y(\theta_i^{mk_m}) y^*(\theta_f^{mk_m}) \,, \tag{13}$$

$$\Delta_{mk_m} = S^{mk_m} - \frac{\pi}{2}\mu^{mk_m} + 2\sqrt{8r_{\mathrm{b}}} - \frac{3\pi}{4} \,, \tag{14}$$

in which

$$y(\theta) = \sum_{l'} (-1)^{l'} b^i_{l'm} I(n,l,l') Y_{l'm}(\theta,0) \,. \tag{15}$$

We will consider the transition of hydrogen atom from the ground state to the high Rydberg state. The whole oscillator strength density can be written as a combination of a smooth background term and a sum of sinusoidal oscillations as follows:

$$\mathrm{D}f(E) = \mathrm{D}f_0(E) + \sum_{k_m} C_{mk_m}\sin(T_{mk_m}E + \Delta_{mk_m}) \,. \tag{16}$$

By using the closed orbits we found for the Rydberg hydrogen atom near two metallic surfaces, we calculate the absorption spectra. The results are given in Figs.3 and 4.

4. Results and discussion

Considering the hydrogen atom to be excited from the ground state to the high Rydberg state with $n = 20$, then $E = -1.25\times10^{-3}$, and $d_{\mathrm{c}} = 1193.76$. By integrating the Hamiltonian motion equations, we find out all the closed orbits of Rydberg hydrogen atom between two metallic surfaces with different distances of the atom from the surfaces. The results show that when the atom–surface distances d_1 and d_2 are near d_{c}, i.e. $d_1 = d_2 = 1200$ a.u., the number of the closed orbits is the largest. If we kept the first metallic surface at $d_1 = 1200$ a.u., this distance is 63.6 nm that is quite typical in surface physics and cavity quantum dynamics,[17] and then we change the distance of the second metallic surface from the hydrogen atom. The results show that when $d_2 > d_{\mathrm{c}}$, with the increase of the distance between the atom and the metallic surface 2, the potential of the metallic surface 2 acting on the atom decreases, which is equivalent to a weaker electric field, and the number of the closed orbits reduces gradually. For example, when the distance $d_2 = 1200$ a.u., we find 42 closed orbits; when $d_2 = 2400$ a.u, there are all together 36 closed orbits. As d_2 is increased to 10000 a.u, only 23 closed orbits can be found, which equals the number of the closed orbits of the Rydberg atom near one metallic surface.[7]

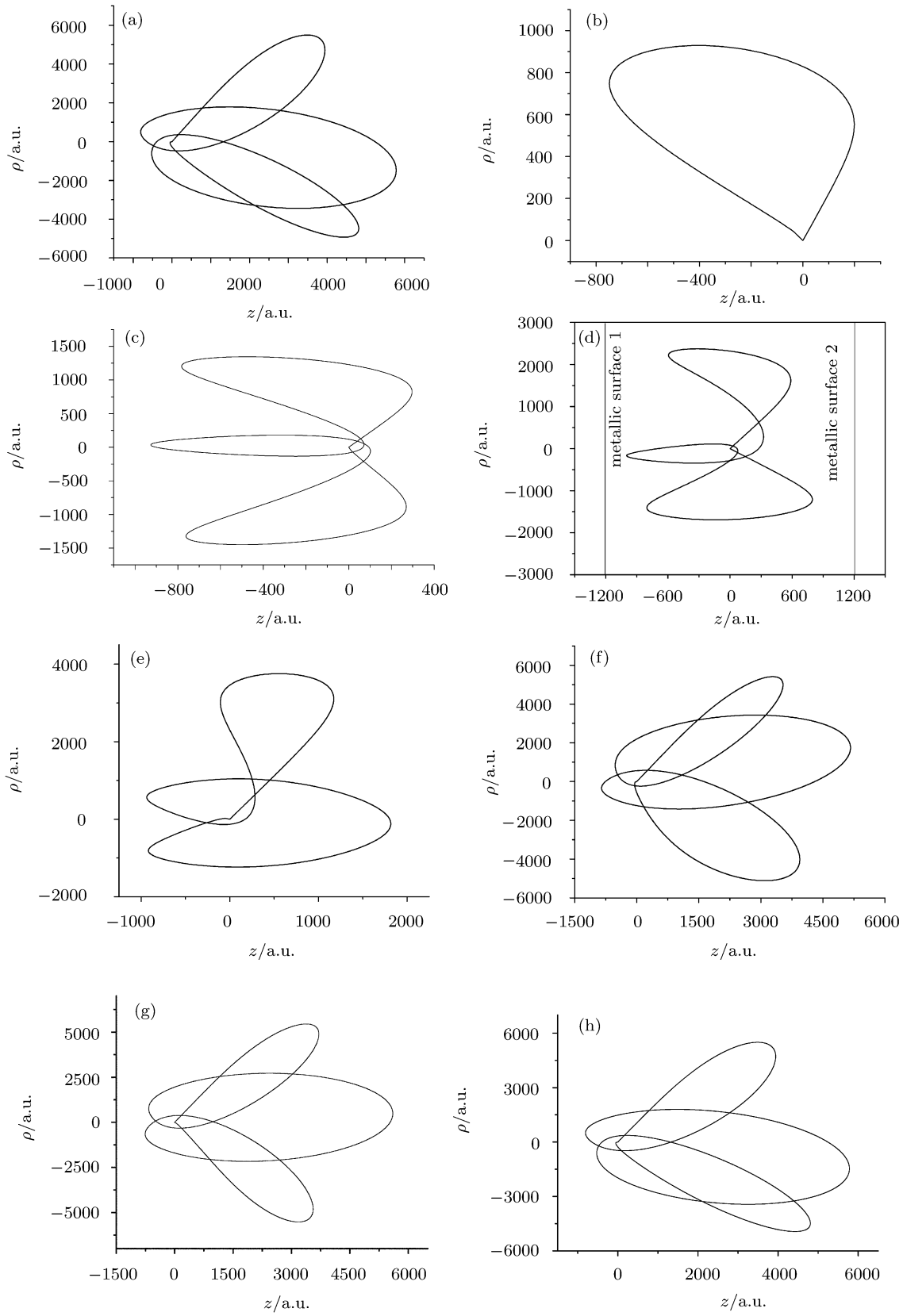


Fig.2. Some electronic closed orbits emitted from the hydrogen atom under the same initial condition, except for the atom–surface distances d_1 and d_2 being different: (a) $d_1 = 1200$ a.u., $d_2 \to \infty$; (b) $d_1 = 1200$ a.u., $d_2 = 400$ a.u., (c) $d_1 = 1200$ a.u., $d_2 = 600$ a.u., (d) $d_1 = 1200$ a.u., $d_2 = 1200$ a.u., (e) $d_1 = 1200$ a.u., $d_2 = 2400$ a.u., (f) $d_1 = 1200$ a.u., $d_2 = 8000$ a.u., (g) $d_1 = 1200$ a.u., $d_2 = 12000$ a.u., (h) $d_1 = 1200$ a.u., $d_2 = 60000$ a.u..

The reason is as follows: when the distance of the atom from the metallic surface is large, the interaction between the atom and the metallic surface is very weak, the electron can travel much farther from the nucleus and the electrical image potential cannot turn the electron back to the nucleus, therefore the number of the closed orbits decreases. On the other hand, when $d_2 < d_c$, with the decrease of d_2, the number of the closed orbits is also reduced. Because when the atom–surface distance d_2 is small, the electrical image potential acting on the electron is very strong, and the electron can travel a much short time in the outer region before it turns back to the nucleus. As the distance d_2 is very small, some of the electronic trajectories can be absorbed onto the metallic surface 2. In order to see the evolution of the closed orbit with the distance of the atom from the metallic surfaces, in Fig.2 we plot eight closed orbits of hydrogen atom near two parallel metallic surfaces with the atom–surface distance $d_1 = 1200$ a.u. under the same initial condition except for the values of d_2 being different. From the figures we found that when d_2 is less than 10000 a.u., the electron can travel between the two surfaces. But when d_2 is large than 10000a.u., the effect of the second metallic surface is very little and this case is equivalent to the photoabsorption process of hydrogen atom near one metallic surface. In Table 1, we give the action and the period of the closed orbits plotted in Fig.2.

Table 1. The action and the period of the classical closed orbits plotted in Fig.2.

	1	2	3	4	5	6	7	8
Period/(a.u.)	3.34×10^6	6.77×10^4	2.56×10^5	4.09×10^5	6.74×10^5	2.92×10^6	3.08×10^6	3.34×10^6
Action	1230.02	154.79	548.04	636.83	688.66	1124.22	1210.32	1228.94

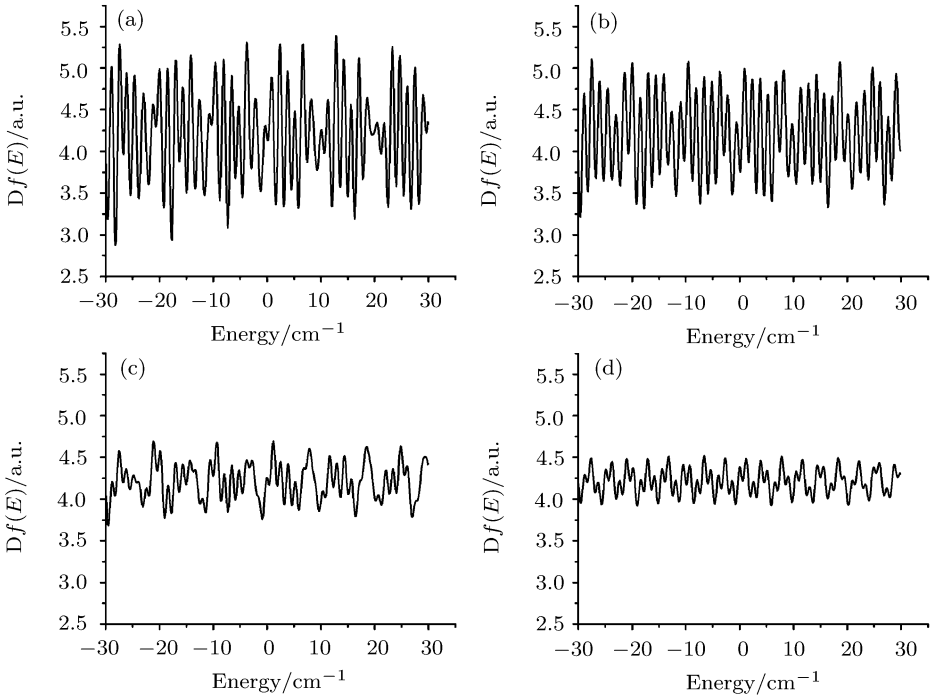


Fig.3. The photoabsorption spectra of Hydrogen atom in the presence of only one metallic surface 1. The four values of the atom–surface distances are (a) $d_1 = 1200$ a.u.; (b) $d_1 = 2000$ a.u.; (c) $d_1 = 3000$ a.u.; (d) $d_1 = 6000$ a.u..

From the calculation we can see that the first orbit can travel a much long distance in the outer region before it returns to the nucleus and the distance it can travel is much farther. Its period is $T_1 = 3.34 \times 10^6$ a.u.. Comparing with the first orbit, the second orbit can travel a much short time and its period is $T_2 = 6.77 \times 10^4$ a.u. and the maximum distance it can travel in the outer region is short. When d_2 increases further, the action and the period are increased correspondingly. As d_2 is increased to

12000 a.u., the action and the period of the seventh closed orbit are close to those of the first closed orbit and the effect of the second metallic surface can be ignored. Therefore, the case in which the second metallic surface is located at infinity amounts to the photoabsorption of atom in the presence of only one metallic surface 1.[7] In Fig.3 we calculate the photoabsorption spectra of hydrogen atom near one metallic surface (metallic surface 1) at different atom–surface distances. The values of the atom–surface distance are: (a) $d_1 = 1200$ a.u.; (b) $d_1 = 2000$ a.u.; (c) $d_1 = 3000$ a.u.; (d) $d_1 = 6000$ a.u.. As we can see from Figs.3(a)–(d), with the increase of the atom–surface d_1, the amplitude of the oscillating spectra decreases and the oscillations become weakened. It is easy to understand the physical reasons behind it. According to the closed-orbit theory, each electron closed-orbit contributes an oscillation to the absorption spectra. As the distance between the atom and metallic surface increases, the number of the closed orbits decreases, hence the absorption spectra are dampened out correspondingly.

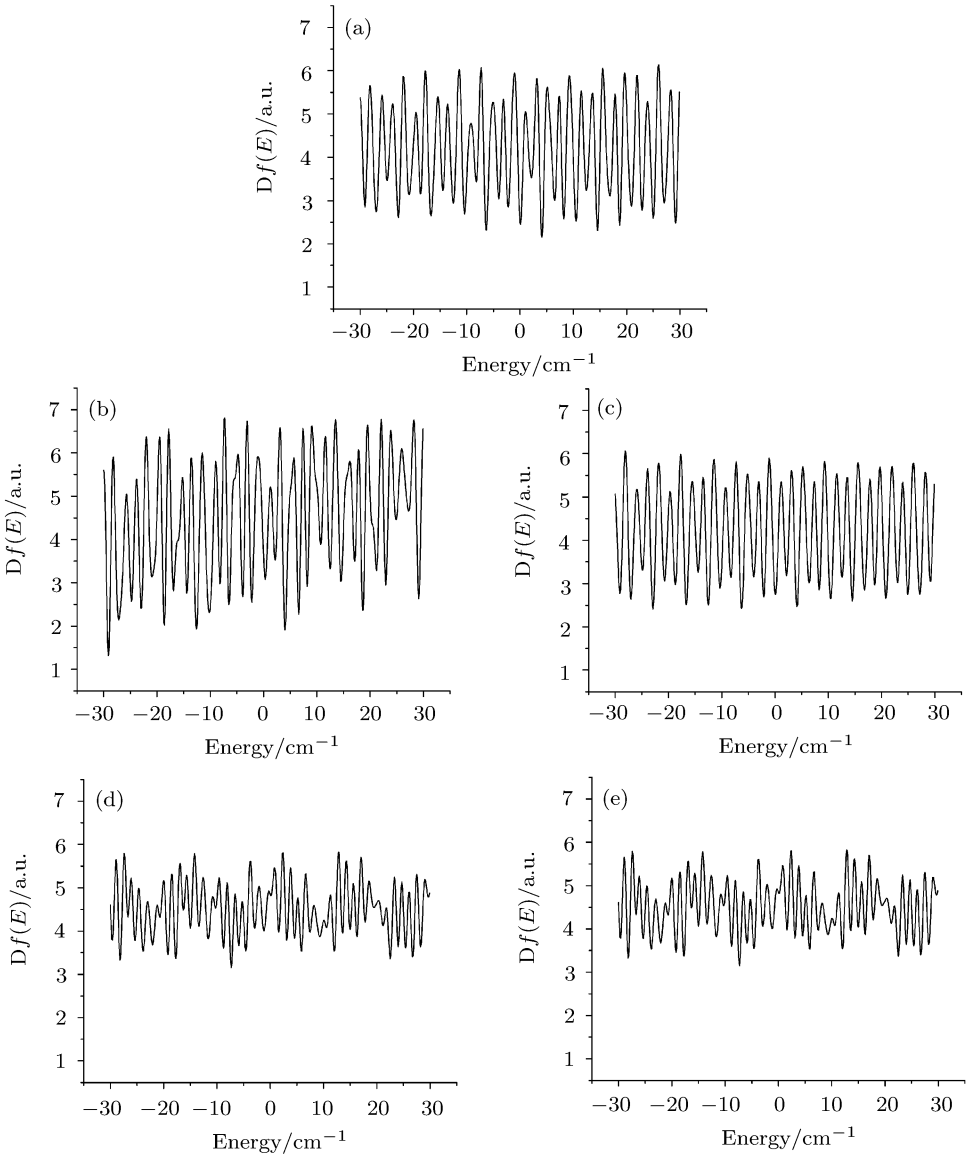


Fig.4. The photoabsorption spectra of Rydberg hydrogen atom near two parallel metallic surfaces. The first metallic surface is located $d_1 = 1200$ a.u., the distance between the atom and the second metallic surface are as follows: (a) $d_2 = 600$ a.u.; (b) $d_2 = 1200$ a.u.; (c) $d_2 = 3000$ a.u.; (d) $d_2 = 12000$ a.u. Figure 4(e) shows the photoabsorption spectra of Hydrogen atom in the presence of only one metallic surface 1 with the atom–surface distance $d_1 = 1200$ a.u..

In Fig.4 we calculate the photoabsorption spectra of Rydberg hydrogen atom in the presence of two metallic surfaces. The first metallic surface is located at $d_1 = 1200$ a.u. and the distance between the atom and the second metallic surface d_2 is varied. The results show, at $d_1 = d_2 = 1200$ a.u., the oscillation of the spectra is the greatest. When $d_2 > 1200$ a.u. or $d_2 < 1200$ a.u., the oscillations become damped. From this result, we conclude that the metallic surface 2 has a significant effect on the photoabsorption of atom. However, when d_2 is increased to ten times the value of d_1, i.e. $d_2 = 12000$ a.u., the photoabsorption spectra are nearly the same as those in the case of the hydrogen atom being near one metallic surface (see Fig.4(d) and Fig.4(e)). The reason is that when d_2 is very large we could ignore the interplay between hydrogen atom and surface 2.

5. Conclusion

In summary, the highly excited hydrogen atom in the presence of metallic surfaces is an ideal system to investigate the instantaneous van der Waals interaction and an important example to reveal quantum chaos. Using the closed orbit theory, we have studied its dynamical properties and calculated the photoabsorption spectra of Rydberg hydrogen atom between two parallel metallic surfaces. The oscillation in the absorption spectra can be attributed to the interferences induced by the outgoing and returning electron waves travelling along the closed orbits. Since the closed orbit theory is only of a semiclassical approximation, the bifurcation and recurrence of the closed orbit might bring some error to the photoabsorption spectra. In order to describe the electron motion exactly, we should use the time-dependent wave packet method.[18] This problem we will discuss in the future. In this paper, we calculate only the absorption spectrum. In our future work, we will calculate the recurrence spectra of Rydberg hydrogen atom near two parallel metallic surfaces and discuss the influence of the bifurcation of the closed orbit on the recurrence spectra. This study provides a visual demonstration of the dynamical behaviour of the Rydberg atom near metallic surfaces. Recently, the interactions of Rydberg atoms,[19] ions[20] and molecules[21] with metallic surfaces and thin metallic films, respectively, have been investigated, but no experiments on the photoabsorption process of Rydberg atom near a metallic surface have been carried out. Our calculation shows that we can use semiclassical closed orbit theory to treat these complicated systems, if an accurate potential between the atoms, ions or molecules and the surface is available. We hope that our results will be useful to understanding the photoabsorption process of atoms, ions or molecules in the vicinity of metallic surface.

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