Semiclassical calculation of ionisation rate for Rydberg helium atoms in an electric field*

Wang De-Hua(王德华)†

College of Physics, Ludong University, Yantai 264025, *China*

(Received 4 March 2010; revised manuscript received 28 March 2010)

The ionisation of Rydberg helium atoms in an electric field above the classical ionisation threshold has been examined using the semiclassical method, with particular emphasis on discussing the influence of the core scattering on the escape dynamics of electrons. The results show that the Rydberg helium atoms ionise by emitting a train of electron pulses. Unlike the case of the ionisation of Rydberg hydrogen atom in parallel electric and magnetic fields, where the pulses of the electron are caused by the external magnetic field, the pulse trains for Rydberg helium atoms are created through core scattering. Each peak in the ionisation rate corresponds to the contribution of one core-scattered combination trajectory. This fact further illustrates that the ionic core scattering leads to the chaotic property of the Rydberg helium atom in external fields. Our studies provide a simple explanation for the escape dynamics in the ionisation of nonhydrogenic atoms in external fields.

Keywords: ionisation rate, closed orbit theory, core scattering, pulse train

PACS: 34.50Gb, 32.80Fb, 03.65Sg **DOI:** 10.1088/1674-1056/20/1/013403

1. Introduction

The ionisation dynamics of the highly excited Rydberg atoms in external fields have been studied extensively over the past decade. Most experimental studies focused on the ionisation dynamics of alkali-metal atoms in an electric field.[1−4] If the Rydberg electron the energy above the saddle point of the classical ionisation threshold, it escapes in a picoseconds time scale. In 1996, Lankhuijizen and Noordam studied the autoionisation of a rubidium atom excited by a short laser pulse in a constant applied electric field.[3,4] The ionisation of the Rydberg rubidium atom occurred via a train of electron pulses, rather than an exponential decay of the ionisation of the hydrogen atom in an electric field. Motivated by this experiment, Mitchell *et al.* predicted that a hydrogen atom can also ionise through emission of an electron pulse train when placed in combined electric and magnetic fields.[5] Later, they gave a detailed analysis of chaos-induced pulse trains in the ionisation of hydrogen by using semiclassical method.[6−8] Their results suggested that the classical chaos in the ionisation of hydrogen atom is caused by the magnetic fields. Subsequently, they studied the transport and escape dynamics concerning the chaos-induced pulse trains in the ionisation of hydrogen atom by a homoclinic tangle technique.[9,10] Although Noordam found that the pulses appeared in the rubidium were related to the core scattering, they did not give a clear description about the escape dynamics and the ionisation property of non-hydrogen atoms in external fields.

In this paper, using the semiclassical theory, we calculate the ionisation of Rydberg helium atom in an electric field and analyse the influence of the core scattering on the escape of the electron. The connection of the peaks in the ionisation rate to the ionisation trajectory is given clearly. Our studies provide a simple explanation for the escape dynamics in the ionisation of nonhydrogenic atoms in external fields.

2. Theory and quantitative formula

The Hamiltonian of Rydberg helium atom in an applied electric field directed along the z-axis is given in cylindrical coordinates (ρ, z) and atomic units by

$$H = \frac{1}{2}(p_\rho^2 + p_z^2) - \frac{1}{\sqrt{\rho^2 + z^2}} + Fz + V_{\rm core}(\rho, z), \quad (1)$$

where F is the electric field and the z component of angular momentum is set to zero. $V_{\rm core}(\rho, z)$ is the

*Project supported by the National Natural Science Foundation of China (Grant Nos. 10604045 and 11074104), the University Science and Technology Planning Program of Shandong Province of China (Grant No. J09LA02), and the Discipline Construction Fund of Ludong University of China.

†Corresponding author. E-mail: jnwdh@sohu.com

© 2011 Chinese Physical Society and IOP Publishing Ltd

model potential for the Rydberg helium atom, which has the explicit form[11]

$$V_{\rm core}(\rho,z) = -\frac{z_e-1}{\sqrt{\rho^2+z^2}} \times\left(1+\frac{\sqrt{\rho^2+z^2}}{a}\right)\mathrm{e}^{-\frac{\sqrt{\rho^2+z^2}}{a}}, \quad (2)$$

where z_e is the atomic number and a is the parameter of the model potential, which is chosen to give the measured quantum defects. For the Rydberg helium atom, $z_e = 2$ and the optimized value of the parameter is given by $a = 1/3$.[11]

If we transform variables according to $\tilde{\rho} = \rho F^{1/2}$, $\tilde{z} = zF^{1/2}$, $\tilde{a} = aF^{1/2}$, $\tilde{p}_\rho = p_\rho F^{-1/4}$, $\tilde{p}_z = p_z F^{-1/4}$, $\tilde{t} = tF^{3/4}$ and $\varepsilon = EF^{-1/2}$,[12] then the transformed Hamiltonian after omitting the tilde '$\sim$' is

$$H = \frac{1}{2}(p_\rho^2+p_z^2) - \frac{1}{\sqrt{\rho^2+z^2}} - \frac{z_e-1}{\sqrt{\rho^2+z^2}} \times\left(1+\frac{\sqrt{\rho^2+z^2}}{a}\right)\mathrm{e}^{-\frac{\sqrt{\rho^2+z^2}}{a}} + z = \varepsilon. \quad (3)$$

From the above equation, we find that the Hamiltonian does not depend upon the energy E and field strength separately, but only on the scaled energy ε, and thus reduce a parameter F.

Since the applied fields point in the positive z direction, the ionising trajectories eventually escape in the negative z direction. In our calculation, the detector is placed at $z = -4$. When we seek the ionisation trajectories that reach the detector, we must integrate the Hamiltonian motion equations. In order to remove the Coulomb singularity at the origin, we transform the cylindrical coordinates into parabolic coordinates.[13] We define parabolic coordinates (u, v) and their conjugate momenta (p_u, p_v) by: $u = \pm\sqrt{r+z}$, $v = \pm\sqrt{r-z}$, $p_u = vp_\rho + up_z$, $p_v = up_\rho - vp_z$, and a new time-like variable τ, defined by $\mathrm{d}\tau/\mathrm{d}t = 1/(u^2+v^2)$. We introduce an effective Hamiltonian $h = 2r(H-\varepsilon)$, which equals

$$h = \frac{1}{2}(p_u^2+p_v^2) - V_{uv}(u,v) - 2, \quad (4)$$

$$V_{uv}(u,v) = -\varepsilon(u^2+v^2) - 2(z_e-1) \times\left(1+\frac{u^2+v^2}{2a}\right)\mathrm{e}^{-\frac{u^2+v^2}{2a}} + \frac{1}{2}(u^4-v^4) - 2. \quad (5)$$

Setting h=0, the Hamiltonian (Eq. (4)) generates the same trajectories as setting $H = \varepsilon$. After the removal of the Coulomb singularity, each trajectory can launch from the origin. These trajectories are integrated until they reach the detector, located at $z = -4$.

Using a short laser pulse, the helium atom can be excited from the ground state to energy above the classical saddle point energy $\varepsilon_{\rm c} = -2$, $\varepsilon_{\rm c}$ being the lowest energy at which a classical electron can pass over the potential barrier. For energies larger than $\varepsilon_{\rm c}$, an excited electron can go over the top of the barrier and escape. Photoexcitation of the helium atom produces a spherical outgoing wave packet, which propagates away from the atom in all directions. As in Ref. [14], we use the semiclassical approximation theory to describe the outgoing wave. Photoabsorption promotes the valence electron from a low-energy bound state into an outgoing wave, which can be modeled semiclassically as an ensemble of trajectories propagating away from the atomic core in all directions and in a narrow range of energies. Some trajectories head directly downhill, and are accelerated by the external field toward a detector, creating an initial prompt pulse of electrons. Other trajectories initially head uphill, are turned around by the field, and return to the core where they scatter in all directions. Some of these scattered trajectories head downhill, creating a second pulse of electrons. Subsequent rescattering events create additional pulses. Therefore, the outgoing wave can be described by an ensemble of classical trajectories moving radially away from the core; these trajectories have a fixed energy and are continuously emitted from the source for all times. At a sufficiently large distance from the nucleus, this wave is approximately expressed as:[15]

$$\psi_{\rm out}(r,\theta,\varphi) = -\,\mathrm{i}\,\pi^{1/2}2^{3/4}\mathrm{e}^{\mathrm{i}(\sqrt{8r}-3\pi/4)}y(\theta,\varphi), \quad (6)$$

where $y(\theta,\varphi)$ gives the outgoing wave's angular distribution. Assuming the z component of the angular momentum to be zero, we reduce $y(\theta,\varphi)$ to $y(\theta)$.

Since the radially outgoing wave far away from the nucleus can be expressed by an ensemble of classical trajectories, the ionisation rate can be calculated by integrating an initial ensemble of radial outgoing trajectories with a single fixed scaled energy from the nucleus to the detector. The ionisation rate is the total number of trajectories striking the detector per unit time, which can be recorded as a function of time.[6]

3. Results and discussion

In order to show the important effect of core scattering on the ionisation of helium atom, we first draw the ionisation rate curve and the escape-time plot of hydrogen atom in the electric field at the scaled energy of the excited electron $\varepsilon = -1.3$, see Fig. 1.

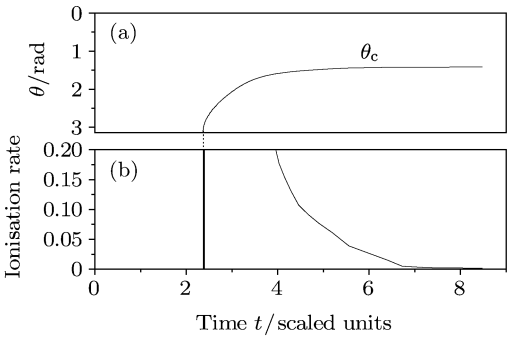


Fig. 1. (a) The escape-time plot for hydrogen atom in electric field, at $\varepsilon = -1.3$. The time it takes a trajectory to strike the detector is plotted as a function of the initial outgoing angle θ, θ_c being the critical angle. (b) The ionisation rate for hydrogen atom in electric field. The dashed line connects the direct ionising trajectory to the corresponding pulse in (a).

The time t takes a trajectory to strike the detector depends on its initial outgoing angle θ, defined as the angle between the initial momentum and the positive z-axis. The curve of the angle θ versus escape time t is called the escape-time plot. The ionisation rate curve is equal to the slope $\mathrm{d}\theta/\mathrm{d}t$ of the escape-time plot weighted by the initial angular distribution $|y(\theta)|^2 \sin\theta$.[6] As shown in Ref. [16], the trajectory that heads directly downhill ($\theta = 180°$) reaches the detector first. As θ decreases, the initial velocity points further and further away from the downhill direction, and the trajectory takes longer and longer time to strike the detector. Eventually, a critical angle θ_c is reached, at which the trajectory takes an infinite time to escape. All trajectories above the critical angle θ_c are bound forever. In the ionisation rate curve, there is only a single pulse of electrons, with an exponentially decaying tail.

Next, we calculate the ionisation rate of Rydberg helium atom in an electric field. In our calculation, we consider the excitation from the helium 2s state to the final states corresponding to the principal quantum numbers around N=80, m=0, the electric field $F = 8.5$ V/cm, the scaled energy of the excited electron is $\varepsilon = -1.3$. Integrating the Hamiltonian motion equations, we find out all the ionisation trajectories of the helium atom in an electric field with the scaled time less than 15. The escape-time plot and the ionisation rate curve are given in Fig. 2. The dashed lines connect the escape segments (i.e. icicles) in Fig. 2(a) to their corresponding pulses in Fig. 2(b). Each peak corresponding to an escape segment implies that a different ensemble of electron trajectories reach the detector at different time intervals.

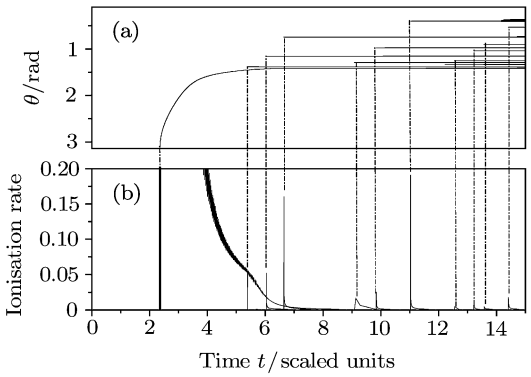


Fig. 2. (a) The escape-time plot for helium atom in an electric field, with the scaled energy $\varepsilon = -1.3$. (b) The ionisation rate for helium atom in an electric field. The dashed lines connect the direct ionising trajectory to the corresponding pulse in (a).

Compared to the ionisation of hydrogen atom in an electric field, different escape segments appear. Rather than a single critical angle, there are many critical angles. Accordingly, there are many pulses in the ionisation rate curves. Each icicle in the escape time plot contributes one pulse to the ionisation rate curve. Thus, there is a one-to-one correspondence between icicles and pulses. The pulse begins at the tip of an icicle, and then it exhibits exponential decay as one moves out along an edge of the icicle.

The difference between the ionisation rate of the helium atom and hydrogen atom can be qualitatively explained by analysing the structure of the electron trajectories. The helium atom is firstly placed in an electric field, after the electron absorbs a photon from the laser pulse, it is promoted from a low-energy bound state into an outgoing wave. This wave can be modeled as an ensemble of trajectories propagating away from the nucleus in all directions. Depending on the launch angle, some of the trajectories return to the nucleus, forming closed orbits.[14] Other trajectories, however, escape the atom and arrive at the detector, contributing to the total ionisation flux. These ionising trajectories may form an interference pattern, which can be measured on a detector in a train of pulses. Alternatively, in a pulsed time-domain experiment, such trajectories may arrive at the detec-

tor in a train of pulses. The classical electron dynamics is determined by the modified Coulomb force plus the external fields applied to the system. For Rydberg hydrogen atom, the trajectories return to the vicinity of the nucleus and the waves associated with them, which are incoming Coulomb waves, produce Coulomb scattered waves. The Coulomb scattered waves are strongly backward focused and retrace the orbit, generating repeated traversals of the trajectory. This pure Coulomb scattering does not bring about any chaotic behaviour and there is no chaos for Rydberg hydrogen atom in the electric field, hence there is only one electron pulse decayed at exponential rate. However, for nonhydrogenic atoms, except the Coulomb scattering, the core-scattering plays an important role.[17] Core scattering in effect constitutes a new source: the core-scattered outgoing waves can be turned around by the external fields, and return to the atom and produce a whole new set of trajectories. The core-scattered wave redistributes the amplitude of one closed orbit into other orbits. Combination of orbits is created by scattering of one closed orbit into other escape trajectory. Because the core induces important dynamical effects, it is now apparent that the contribution of the core-scattered trajectory in any semiclassical approach is necessary to be included. Some of the core-scattered trajectories are given in Fig. 3.

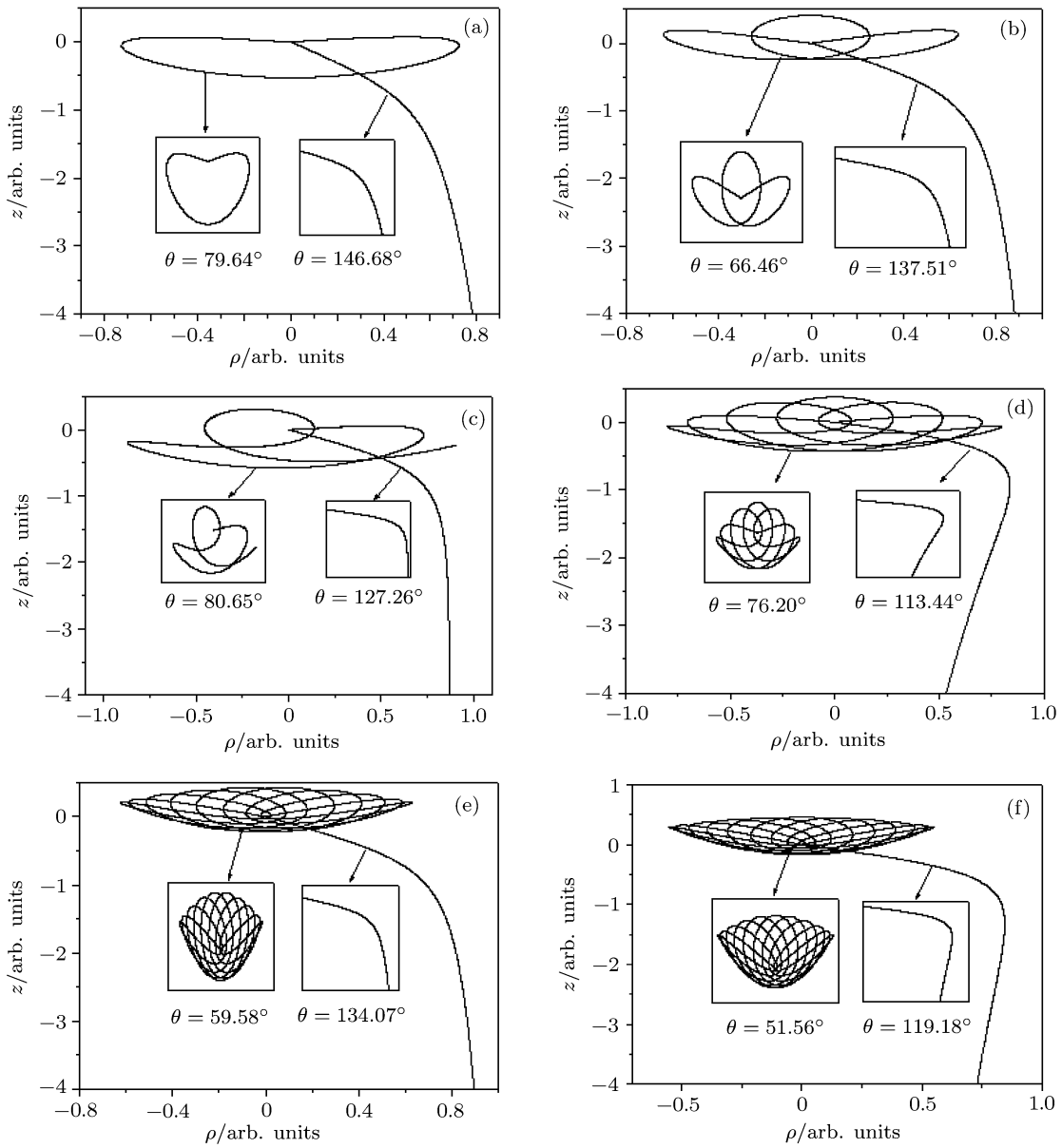


Fig. 3. Some core-scattered combinated ionising trajectories of helium atom in an electric field at the scaled energy $\varepsilon = -1.3$. The emission time of the ionisation trajectories are as follows: (a) $t = 5.38$; (b) $t = 6.02$; (c) $t = 9.08$; (d) $t = 11.02$; (e) $t = 13.23$; (f) $t = 14.42$.

Considering the influence of the core scattering, there are a lot of pulse trains appearing in the ionisation of helium atoms in an electric field. The first pulse in Fig. 2 is the initial prompt pulse induced by the electron trajectories, which heads directly downhill and reaches the detect plane. These trajectories are not affected by the core scattering. Other pulses could be attributed to the combined trajectories caused by the core scattering. For example, the second pulse that begins at the scaled time $t = 5.38$ is caused by the combinated trajectories in Fig. 3(a), which can be considered as the fact that the electron first travels along the closed orbit with the initial angle $\theta = 79.64°$ and then is scattered into the downhill ionisation trajectory with the initial angle $\theta = 146.68°$ through core scattering. Similarly, the origination of the third peak at scaled time of $t = 6.02$ is caused by the combinated trajectories in Fig. 3(b), which can be considered as the combination of the closed orbit with the initial angle $\theta = 66.46°$ and the downhill ionisation trajectory with the initial angle $\theta = 137.51°$ through core scattering. The pulse appears at scaled time of $t = 9.08$ is caused by the combinated trajectories of Fig. 3(c), which corresponds to the combinated trajectories of the closed orbit with the initial angle $\theta = 80.65°$ and the downhill ionisation trajectory with the initial angle $\theta = 127.26°$ through core scattering. Other pulses can be analysed in the same way. The closed orbit and the ionising trajectory are given in the inset of each plot.

4. Conclusions

We have studied the ionisation of a Rydberg helium atom in an electric field above the classical ionisation threshold. The influence of the core scattering on the escape dynamics of the electron has been discussed in great detail. The results show that due to the influence of the core scattering, a lot of core-scattered combinated ionisation trajectories appear. Each peak in the ionisation rate curve corresponds to one electron's ionisation trajectory. Compared to the case of the ionisation of hydrogen atom in an electric field, the escape-time plot and the ionisation pulse trains of the Rydberg helium atom in electric field become much more complicated. Our calculation result further verify that the existence of pulse trains is a direct consequence of core scattering, which occurs for all the multi-electron atoms, such as experimentally measured pulse trains by Noordham for the Rydberg Rb atom.[3] At present, experimental results on the ionisation of helium atom in electric field are not available for comparison. We hope that our results will be useful for guiding experimental studies of the ionisation and escape dynamics of multi-electron Rydberg atoms and ion in external fields or near metal surfaces.[18,19]

References

[1] Broers B, Christian J F and Hoogenraad J H 1993 *Phys. Rev. Lett.* **71** 344

[2] Zhou H, Li H Y, Gao S, Zhang Y H, Jia Z M and Lin S L 2008 *Chin. Phys.* B **17** 4458

[3] Lankhuijzen G M and Noordam L D 1996 *Phys. Rev. Lett.* **76** 1784

[4] Lankhuijzen G M and Noordam L D 1995 *Phys. Rev.* A **52** 2016

[5] Mitchell K A, Handley J P, Tighe B, Flower A and Delos J B 2004 *Phys. Rev. Lett.* **92** 073001

[6] Mitchell K A, Handley J P, Tighe B, Flower A and Delos J B 2004 *Phys. Rev.* A **70** 043407

[7] Mitchell K A and Delos J B 2006 *Physica* D **221** 170

[8] Mitchell K A and Steck D A 2007 *Phys. Rev.* A **76** 031403R

[9] Mitchell K A, Handley J P, Tighe B, Knudson S K and Delos J B 2003 *Chaos* **13** 880

[10] Mitchell K A, Handley J P, Tighe B, Knudson S K and Delos J B 2003 *Chaos* **13** 892

[11] Hupper B, Main J and Wunner G 1996 *Phys. Rev.* A **53** 744

[12] Delande D and Buchleitner A 1995 *Advances in Atomic Molecular and Optical Physics* **34** 85

[13] Courtney M, Spellmeyer N, Jiao H and Kleppner D 1995 *Phys. Rev.* A **51** 3604

[14] Du M L and Delos J B 1988 *Phys. Rev.* A **38** 1896

[15] Gao J and Delos J B 1992 *Phys. Rev.* A **46** 1455

[16] Wang D H, Huang K Y, Zhou H and Lin S L 2009 *J. Electron Spectrosc.* **166** 86

[17] Karremans K, Kips A, Vassen W and Hogervorst W 1999 *Phys. Rev.* A **60** R2649

[18] Wang D H and Wu A L 2008 *J. Electron Spectrosc.* **165** 36

[19] Huang K Y and Wang D H 2010 *Chin. Phys.* **19** 063402