Classical dynamics of s-wave helium

G. Handke, M. Draeger and H. Friedrich

Physik Department, Technische Universität München, W-8046 Garching, Germany

Received 29 January 1993

s-wave helium is a model of a two-electron atom in which both electrons are restricted to spherical states. The classical dynamics of this model exhibits considerable resemblance to the collinear approximation of real helium, which has been studied in detail by Ezra et al. [J. Phys. B 24 (1991) L413] and Wintgen et al. [Chaos 2 (1992) 19]. Both systems are fully chaotic, the periodic orbits are all unstable, proliferate exponentially and can be labelled by the same simple binary code. In s-wave helium, non-periodic trajectories can unambiguously be assigned an ionisation time or a scattering time delay, and they can be classified using this binary code. Ionisation times impose a fractal structure on phase space and the initial conditions of the non-ionising trajectories form a Cantor set. The density of bound trajectories decreases exponentially with increasing number of encounters of the two electrons, but follows a power law as a function of the ionisation time. The mean lifetime of the bound atom against autoionisation is infinite.

1. Introduction

Despite the growing interest and multifarious efforts in recent years the strongly correlated motion of the electrons in a two-electron system still remains a challenge for physicists. The progress made experimentally [1,2] and theoretically [3–13] in studies of doubly excited helium reveals complex physical properties in this seemingly simple system. The quantum mechanical energy spectrum becomes complicated and interesting below the two-electron ionisation threshold, where many Rydberg series and continua interact [6,11,12]. Classically the chaotic motion of the electrons in large parts of phase space is attracting increasing attention [9,13]. Helium is one of the simplest, but nevertheless not fully understood, realistic physical systems with chaotic classical dynamics.

In this paper we study s-wave helium, a model in which both electrons are restricted to spherical states with individual angular momentum zero. It is perhaps the simplest model for two electrons interacting with each other and with a nucleus via long-ranged Coulomb forces. It can be interpreted as the
first term of a partial waves expansion of spherical real helium, where only the
total angular momentum is zero [14]. Classically, s-wave helium corresponds to
two spherical shells of charge \(-1\) expanding and contracting around a fixed
nucleus of charge \(Z\). The restriction to s-waves eliminates all angular correla-
tions between the electrons, but the dynamics in the resulting model closely
resembles that of the collinear approximation of real helium, where angular
correlations are maximal. The potentials are similar, and the periodic orbits
have the same structure. Real helium might be expected to correspond to
something in between these two extremes.

As in collinear helium [8,9,13], the classical dynamics of s-wave helium is
chaotic; the trajectories are typically unbound and describe ionisation and
scattering. Hence the present results are also of general interest for the study
of chaotic ionisation and scattering [15–27].

The equations of motion of s-wave helium are discussed in section 2. In
section 3 we describe the general behaviour of periodic and non-periodic
trajectories. Section 4 presents results based on calculations of many trajec-
tories and reveals a fractal structure of classical phase space.

2. Equations of motion

Our starting point is the nonrelativistic two-electron atom \((Z = 2)\) or ion
\((Z > 2)\) neglecting mass polarisation. (Numerical calculations are for \(Z = 2\)
throughout the paper.) Projecting all one-particle states onto angular momen-
tum zero reduces the number of degrees of freedom to two. These are the
radial distances \(r_1\) and \(r_2\) of the two electrons from the nucleus. The Hamilto-
nian function is

\[
H(r_1, r_2) = \frac{p_1^2}{2/r_1} + \frac{p_2^2}{2/r_2} - \frac{Z}{r_1} - \frac{Z}{r_2} - \frac{1}{r_\infty} = \frac{p_1^2}{2/r_\infty} + \frac{p_2^2}{2/r_\infty} - \frac{Z}{r_\infty} - \frac{Z-1}{r_\infty},
\]

where atomic units are used here and in the following. \(r_\infty\) stands for the smaller
and \(r_\infty\) for the greater of \(r_1\) and \(r_2\). The collinear approximation of real helium
(with electrons on opposite sides of the nucleus) is defined by a Hamiltonian
differing from (1) only in that the repulsive interaction term \(1/r_\infty\) is replaced by
\(1/(r_1 + r_2)\).

Within the regions \(r_1 > r_2\) (region 1) and \(r_1 < r_2\) (region 2) the classical
equations of motion generated by (1) are separable. They describe one-
dimensional Kepler motion in each of the two uncoupled coordinates:

\[
\frac{\dot{r}_1^2}{2} - \frac{Z - 1}{r_1} = E_1, \quad \frac{\dot{r}_2^2}{2} - \frac{Z}{r_2} = E_2 \quad (\text{region 1}),
\]
\[ \frac{\dot{r}_1^2}{2} - \frac{Z}{r_1} = E_1, \quad \frac{\dot{r}_2^2}{2} - \frac{Z-1}{r_2} = E_2 \]  
(region 2).  

(2)

Within each region the energies \( E_1 \) and \( E_2 \) for the two degrees of freedom are conserved, but the only global constant of motion is the total energy \( E = E_1 + E_2 \). Coupling between the two degrees of freedom occurs on the line \( r_1 = r_2 \), where the equipotential lines in the \( r_1, r_2 \)-plane show a kink (fig. 1). When this line is crossed there is a sudden change of acceleration and an energy exchange between the two electrons. The amount of energy exchanged depends only on the position \( s = r_1 = r_2 \) where the electrons meet. If the transition goes from region 1 to region 2, then

\[ E_1^{(a)} = E_1^{(b)} - 1/s, \quad E_2^{(a)} = E_2^{(b)} + 1/s, \]  

(3)

where \( E_i^{(a)} \) and \( E_i^{(b)} \) are the energies after and before the transition respectively. If the transition is from region 2 to region 1, then

\[ E_1^{(a)} = E_1^{(b)} + 1/s, \quad E_2^{(a)} = E_2^{(b)} - 1/s, \]  

(4)

i.e. after an encounter at \( r_1 = r_2 = s \), the now larger coordinate has gained the energy \( 1/s \) at expense of the now smaller coordinate. For any given (negative) initial energies \( E_1^{(b)} \) and \( E_2^{(b)} \), the energy gain can lead to a positive energy of one electron if the encounter occurs so close to the nucleus that \( 1/s \) is sufficiently large. Due to this possibility the volume of phase space is infinite.

Fig. 1. Equipotential lines of the potential of (1) (for \( Z = 2 \)) from \(-5.0\) to \(-1.0\) atomic units in steps of 0.5.
Since the potential in (1) is homogeneous, the equations of motion can be scaled to energy independent form. It is only necessary to distinguish between positive, negative or vanishing total energy. Bound motion (i.e. negative energy) of both electrons is only possible for negative total energy corresponding to the domain below the double ionisation threshold. This domain is the subject of present investigation and we choose $E = -1$.

Although the equations of motion can be solved analytically in the uncoupled regions there is no explicit formula for the trajectories. The solutions are expressions for time as a function of coordinates such as

$$t(r_1) = \sqrt{\frac{1}{-2E_1}} \left\{ \sqrt{\frac{Z-1}{-E_1}} r_1 - r_1^2 + \frac{Z-1}{E_1} \left[ \arcsin\left(\sqrt{\frac{-E_1 r_1}{Z-1}}\right) - \frac{\pi}{2} \right] \right\}, \quad (5)$$

which is an example for region 1. They cannot be resolved for $r_1$ or $r_2$, so trajectories have to be calculated point by point by numerical root finding.

3. Trajectories

Detailed studies of the periodic orbits of the collinear configuration in real helium have recently been performed by Wintgen et al. [9,13]. These orbits are all unstable and can be enumerated by a simple symbolic binary code. This code registers collisions with the $r_1$-axis and the $r_2$-axis during one period. Two consecutive collisions with the same axis are coded as "+", with different axes as "-". Since the number of possible codes grows exponentially with its length and a longer code corresponds to an orbit with higher action one can conclude that the number of periodic orbits proliferates exponentially with the action. Together with the instability of the periodic orbits this implies that the system is fully chaotic.

The structure of the periodic orbits in s-wave helium is essentially the same as for collinear helium, see fig. 2 for two examples and for comparison fig. 4 of [13]. All periodic orbits found are unstable, and they can be uniquely labelled using the same symbolic code registering successive collisions with the same or different axes. The Liapunov exponents are of the same order as those for the corresponding orbits in collinear helium. Because of the kink in the potential, the orbit along the line $r_1 = r_2$ has an infinite Liapunov exponent. Instability and exponential proliferation imply complete chaos in s-wave helium as in collinear helium.

Calculation of a large number of non-periodic trajectories shows that they typically are unbound. After several crossings of the line $r_1 = r_2$ one electron acquires positive energy and ionisation takes place. It can be shown that when
a crossing leads from negative energies of both electrons to a positive energy of one (the outer) electron, this is the final crossing of the line $r_1 = r_2$, i.e. once the electron has acquired a positive energy, it moves forever away from the nucleus and never again meets the other (inner) electron (see the Appendix). Thus for any given initial conditions corresponding to bound motion, $E_1 < 0$, $E_2 < 0$, we can unambiguously define the ionisation time as the duration of the bound motion of both electrons. We can adapt the binary code used to label periodic orbits to classify the non-periodic trajectories: successive collisions with the same or different axes are registered as "+" or "-" respectively, up to the final crossing of the line $r_1 = r_2$, which is followed by ionisation. Two examples are shown in fig. 3.

Initial states in which one electron has a positive energy (and is moving towards the nucleus) correspond to a scattering process. The first encounter of the two electrons on the line $r_1 = r_2$ may lead to bound motion of both electrons, $E_1 < 0$, $E_2 < 0$. In this case further crossings of the line $r_1 = r_2$ take

Fig. 2. Two periodic orbits together with the equipotential line at $-1$ atomic unit. The codes of the orbits are also given.

Fig. 3. Two examples of ionising trajectories with the equipotential line at $-1$ atomic unit and the codes of the classes to which they belong.
place and the bound motion of both electrons persists until the last crossing. The time delay during the scattering process can be unambiguously defined as the time elapsed between the first and the last crossing. This corresponds to adding two (usually different) ionisation times. In other scattering and ionising systems such as electron–ion scattering in the presence of infra-red radiation [22–24] or ionisation of the hydrogen atom in crossed magnetic and electric fields [26] the definition of ionisation times and time delays is more difficult and usually requires fitting trajectories to asymptotically free motion far from the interaction centre.

4. Results

Since all trajectories hit an axis in the \( r_1, r_2 \)–plane at least once and the system is symmetric in \( r_1 \) and \( r_2 \), it is sufficient to study trajectories starting on the \( r_1 \)-axis. Each trajectory can be identified by two parameters such as the initial position on the \( r_1 \)-axis and the initial momentum \( p_1 \) in \( r_1 \)-direction. This gives a complete survey of phase space. A given trajectory corresponds at least to one, usually to several, sets of initial conditions \( r_1, p_1 \). The requirement that the motion of both electrons should be bound initially restricts the domain of possible initial conditions to (fig. 4)

\[
\frac{Z-1}{r_1} - 1 < \frac{p_1^2}{2} < \frac{Z-1}{r_1}.
\]

We have calculated trajectories and corresponding ionisation times (for \( Z = 2 \)) by scanning the set of possible initial conditions in various regions of phase space and on different scales. Calculations were performed on a lattice of \( 500 \times 500 \) points. Fig. 5 shows results of four calculations on finer and finer scale, where each picture is a magnification of a small region of the preceding picture. The ionisation times are drawn in a colour representation in the \( r_1, p_1 \)-plane of initial conditions. Yellow points correspond to the shortest and blue points to the longest ionisation times. Trajectories which do not ionise in the duration of the calculation are attached to the maximum of calculated times. (The scattering time delay of a given trajectory characterised by initial parameters \( r_1, p_1 \) corresponds simply to the sum of its ionisation time and the ionisation time of the trajectory with the initial parameters \( r_1, -p_1 \).)

Fig. 5a displays that part of the allowed region (6) which is marked by the rectangle in fig. 4. Three successive magnifications (figs. 5b, 5c, 5d) show the self-similar structure very clearly. Sections at \( p_1 = 0 \) are given for the same regions of \( r_1 \) in fig. 6.
There is pronounced structure in the pictures. Stripes which consist of only one colour, i.e. trajectories with nearly the same ionisation time, alternate with variegated stripes which contain the same pattern again, and so on. Looking at the trajectories in the $r_1, r_2$-plane one understands how these structures arise. The shortest way to ionisation is along a trajectory which goes from the $r_1$-axis directly to the line $r_1 = r_2$ where energy transfer according to (3) gives a positive value for $E_2$. These trajectories form the yellow stripe on the left of fig. 5a. The next large yellow stripe around $r_1 = 3.7, p_1 = 0$ contains trajectories with one more collision with the $r_1$- or $r_2$-axis respectively. Every further big homogeneous stripe consists of trajectories with one more oscillation in $r_2$. In terms of the $(+, -)$-coding introduced above, this means one more "+"-sign in front of the code. So the stripes can be classified by the codes of the trajectories in it. The first yellow stripe on the left contains only trajectories which do not hit an axis before ionisation and is therefore classified by an empty code with no sign. The next stripe contains the two classes "+" and
Fig. 5. Ionisation times as a function of the initial values $r_1, p_1 (r_2 = 0)$ in atomic units. They are shown in a colour representation with yellow for the shortest and blue for the longest times. The precise meaning of the colours is specified in the spectrum at the bottom of (d). (b), (c) and (d) are in each case magnifications of small areas (marked by rectangles) in the preceding pictures.
Fig. 5. (cont.)
Fig. 6. Ionisation times as function of the initial value of $r_1$ with $p_1 = 0$. From top to bottom the panels correspond to cuts parallel to the $r_1$-axis of figs. 5a–d.

"-". In the third stripe (around $r_1 = 5$, $p_1 = 0$) we find the "++" and "+-", the next stripe contains "+++" and "++-", etc. Since the length of the trajectories increases with code length the colour of the stripes changes gradually.

The structures inside the inhomogeneous stripes can also be understood on the basis of this coding scheme. All inhomogeneous stripes show the same pattern and the codes of their trajectories depend on that neighbouring homogeneous stripe outside which has the longer code – on the right (left), say. The inhomogeneous stripes have the largest homogeneous structure near
the middle. There we find trajectories having an additional "-" before the last sign compared with the corresponding stripe outside. To the right and left there are stripes with decreasing width. Those to the left (right) get just another "+" before the last sign for each step to the left (right), those to the right (left) one "-" in the first step and then likewise one more "+" in every further step. Fig. 7 illustrates how this coding scheme works. The first column represents a horizontal cut through fig. 5a, as in the top row of fig. 6. The second column shows a magnification of the second inhomogeneous stripe (around $r_1 = 4.3$, $p_1 = 0$). The neighbouring homogeneous stripe with the longer code is the "++", "+-" stripe. Therefore the trajectories which

Fig. 7. Coding scheme of the unbound trajectories. Parts of the upper three panels of fig. 6 are represented schematically; the $r_1$-axis is rotated and goes from bottom to top. The open rectangles stand for the homogeneous stripes and the codes of the corresponding classes of unbound trajectories are given inside (or next to) them.
constitute the stripe around \( r_1 = 4.35 \), \( p_1 = 0 \) are coded by \( "++-" \) and \( "+++" \). Towards lower \( r_1 \) we have \( "++-++" \) and \( "++-+-" \), then \( "+-++-++" \) and \( "+-++--" \) etc. Towards higher \( r_1 \) the next stripe is \( "++-+-" \) and \( "+-++--" \) followed by \( "+-+++-" \) and \( "+-++-++" \) and so on.

Figs. 5 and 6 clearly show the self-similar fractal structure of phase space. There is an obvious resemblance to the standard Cantor set, where a certain fraction of a continuous interval is excluded in the first step and the same (or different) fractions of the remaining connected intervals are excluded in each successive step. The homogeneous stripes correspond to the intervals excluded in the construction of the Cantor set and the only non-ionising trajectories are those which in no step belong to such a homogeneous stripe. Thus the initial conditions of the non-ionising trajectories (including the periodic orbits) form the residual Cantor set. The borders of the homogeneous stripes consist of trajectories with arbitrarily high ionisation times. These are trajectories in which an encounter on the line \( r_1 = r_2 \) gives one electron an energy just below (but arbitrarily close to) zero. So this electron moves far away from the nucleus, but comes back after an arbitrarily long time.

Figs. 8a and 8b show the density \( n \) of trajectories starting with initial conditions in fig. 5b as function of ionisation time \( T \), calculated in intervals of 40 atomic units width. Fig. 8a has a linear scale in \( T \) and a logarithmic scale in \( n(T) \), in fig. 8b both scales are logarithmic. Except for the shortest times, \( n(T) \) shows a linear behaviour in the histogram 8b and therefore follows a power law. A fit of the data yields

\[
n(T) \propto T^{-1.82}
\]

for sufficiently large times. Note that \( n(T) \) remains bounded for short times. Because the exponent in this power law is between \(-1\) and \(-2\), the mean ionisation time

\[
\langle T \rangle = \frac{\int_0^\infty n(T) T \, dT}{\int_0^\infty n(T) \, dT}
\]

has a convergent denominator and a divergent numerator. Thus the mean ionisation time defined by (8) is infinite. This means that the average lifetime of classical s-wave helium with respect to autoionisation is infinite although most trajectories lead to ionisation.

In the construction of a Cantor set, the measure of the residual set decreases
The density of trajectories (plotted logarithmically) as a function of ionisation time $T$ (a), (b) and as a function of the number $w$ of encounters on the line $r_1 = r_2$ (c), (d). The scale of the abscissa is linear in (a) and (c) and logarithmic in (b) and (d).

We find such an exponential decrease of the number of trajectories as a function of the number $w$ of encounters on the line $r_1 = r_2$, i.e. the number of "-"-signs in the code. This is illustrated in Fig. 8c, where the density of trajectories is plotted logarithmically as a function of $w$ on a linear scale. (For comparison fig. 8d shows a doubly logarithmic plot.) A straight line fit to fig. 8c yields
The physical interpretation of this is as follows. In a chaotic system the long time behaviour of motion should not depend on the initial conditions. In our case this means that, after a number of encounters on the line \( r_1 = r_2 \), the probability that both electrons remain bound after the next encounter should approach a constant. This yields the exponential dependence of the density of trajectories on \( w \). The different behaviour of \( n \) in (7) and in (9) is related to the occurrence of trajectories with very long ionisation times but relatively few encounters on the line \( r_1 = r_2 \). Such trajectories contain at least one long interval between two successive encounters, i.e. at least one long interval during which the motion of the two bound electrons is uncoupled.

Investigations of other models [19–21] have identified the Cantor set structure of delay times and an exponential decrease of the measure of the set of initial conditions for which a trajectory has a delay time longer than a given value \( T \) (corresponding to an exponential decrease of \( n(T) \)) as characteristics of chaotic scattering [18,21]. The Cantor set structure is present in s-wave helium, but the exponential behaviour is seen if we characterise the trajectories by the number \( w \) of encounters of the two electrons rather than the delay time. As a function of the delay (or ionisation) time \( T \), the number of trajectories decreases via a power law, a behaviour also observed by Wiesenfeld [22] and Lai et al. [24] for Coulomb scattering in the presence of an oscillating field, and by Hillermeier et al. [25] in kicked hydrogen. In [24] the power law decay is attributed to the presence of islands of stability which surround stable periodic orbits, in [25] it is related to symbolic dynamics involving infinitely many symbols. Neither of these situations applies in the present case of s-wave helium, in particular, there are no stable periodic orbits. Whether we see exponential or power-law decay depends on the variable chosen to characterise the trajectories.

5. Conclusion

In this paper we have presented a comprehensive account of the classical dynamics of s-wave helium at negative total energy. Periodic orbits are all unstable and proliferate exponentially, so the system is fully chaotic. Non-periodic trajectories can be assigned an ionisation time or a scattering time delay, which is the unambiguously defined time between an initial and a final encounter at \( r_1 = r_2 \). The binary coding developed for periodic orbits (and initially introduced by Wintgen and collaborators [9,13] in the collinear approximation of real helium) can also be adapted to classify the non-periodic
trajectories. The ionisation times impose a fractal structure on the phase space and the elements of this structure can be fully understood as classes of (non-periodic) trajectories with given binary codes. The initial conditions of the orbits which do not ionise after a finite time (or finite code length) form a Cantor set. The density of trajectories decreases exponentially as a function of the number \( w \) of encounters at \( r_1 = r_2 \), but its dependence on the ionisation times is given by a power law. The mean duration of bound motion, i.e. the mean lifetime with respect to autoionisation, is infinite, even though almost all trajectories ionise.

Acknowledgements

We thank C. Hillermeier for helpful discussions. This work was supported by the Deutsche Forschungsgemeinschaft, Az: IIC4-Fr 591/3-3.

Appendix

If there is bound motion \( (E_1 < 0, E_2 < 0) \) of both electrons, then this bound motion persists between an initiating and a terminating crossing of the line \( r_1 = r_2 \) in the \( r_1, r_2 \)-plane at which one electron looses respectively acquires positive energy\(^1\). Before the initiating and after the final crossing there are no other encounters on the line \( r_1 = r_2 \) and the energy of the outer electron is always positive.

To see this it is sufficient to show that a crossing leading from bound motion of both electrons to a positive energy of one electron remains the final encounter with the line \( r_1 = r_2 \). From time-reversed arguments it then follows that an initial crossing leading from positive energy of one electron to bound motion of both electrons must have been the first crossing.

Let us assume that a crossing at \( r_1 = r_2 = s \) leads from bound motion of both electrons in region 2 \( (r_1 < r_2) \),

\[
E_1^{(b)} < 0, \quad E_2^{(b)} < 0, \quad E_1^{(b)} + E_2^{(b)} = E = -1,
\]

(A.1)

to unbound motion of electron 1 (in region 1, \( r_1 > r_2 \)),

\[
E_1^{(a)} = E_1^{(b)} + 1/s > 0, \quad E_2^{(a)} = E_2^{(b)} - 1/s < -1.
\]

(A.2)

First we show that \( p_1^2 \) must be larger than \( p_2^2 \) at such a crossing. Just before the

\(^1\) Unless it persists without interruption for an infinite time.
encounter the energies of the two electrons are

\[ E_1^{(b)} = \frac{1}{2} p_1^2 - \frac{Z}{s}, \quad E_2^{(b)} = \frac{1}{2} p_2^2 - \frac{Z - 1}{s}. \tag{A.3} \]

Just after the crossing the energies of the electrons are

\[ E_1^{(a)} = \frac{1}{2} p_1^2 - \frac{Z - 1}{s} > 0, \quad E_2^{(a)} = \frac{1}{2} p_2^2 - \frac{Z}{s} < -1. \tag{A.4} \]

From the first inequality (A.4) and the second equation (A.3) we have

\[ \frac{1}{2} p_1^2 > \frac{Z - 1}{s} = \frac{1}{2} p_2^2 - E_2^{(b)} > \frac{1}{2} p_2^2, \tag{A.5} \]

where the last inequality follows from the fact that \( E_2^{(b)} \) is negative (bound motion before crossing).

Thus for the crossing at \( r_1 = r_2 = s \) we have

\[ \frac{1}{2} p_1^2(s) > \frac{1}{2} p_2^2(s). \tag{A.6} \]

\( p_1(s) \) must be positive, since the trajectory moves into region 1 after crossing. Since \( E_1^{(a)} \) is positive, \( r_1 \) must increase monotonically until a possible further crossing at \( r_1 = r_2 = s' > s \). Just before such a postulated second crossing we have

\[ \frac{1}{2} p_1^2(s') - \frac{1}{2} p_2^2(s') = \left( E_1 + \frac{Z - 1}{s'} \right) - \left( E_2 + \frac{Z}{s'} \right) \tag{A.7} \]

compared with

\[ \frac{1}{2} p_1^2(s) - \frac{1}{2} p_2^2(s) = \left( E_1 + \frac{Z - 1}{s} \right) - \left( E_2 + \frac{Z}{s} \right) \tag{A.8} \]

just after the aforementioned crossing at \( r_1 = r_2 = s \). Since \( s' > s \), comparing (A.7) and (A.8) gives

\[ \frac{1}{2} p_1^2(s') - \frac{1}{2} p_2^2(s') > \frac{1}{2} p_1^2(s) - \frac{1}{2} p_2^2(s) > 0. \tag{A.9} \]

However, a trajectory in region 1 (\( r_1 > r_2 \)) with \( p_1 > 0 \) can only approach the line \( r_1 = r_2 \) if \( p_2 > p_1 \), because the angle to the \( r_1 \)-axis must be larger than 45°. Hence the postulated second crossing cannot occur; the trajectory moves monotonically to larger and larger values of \( r_1 \) after the crossing at \( r_1 = r_2 = s \), which thus terminates the bound motion.
References