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Circular Rydberg orbits in circularly polarized microwave radiation

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Using classical dynamics we analyze the ionization of the maximum angular-momentum circular Rydberg orbits of the hydrogen atom in strong circularly polarized microwave radiation. We find the ionization threshold generally higher than that for the static field, depending upon the direction of rotation, and in some cases completely different than the prediction based upon the above barrier escape. Below the ionization threshold the system returns to its initial state after interacting with the smooth microwave pulse.

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While interaction of atoms in Rydberg states with a linearly polarized microwave field has been studied in detail both theoretically [1] and experimentally [2], the corresponding process in a circularly polarized microwave field has, so far, received little attention. In a recent Letter [3] the first experimental measurement to our knowledge of the threshold field for ionization by a circularly polarized field was reported. The threshold field, higher than that for the linearly polarized field, scaled with the principal quantum number in a way characteristic of the classical dynamics and was very close to the value expected for the onset of the field ionization in a static field ($E=1/16n^4$). In a brief Comment [4], Nauenberg argued that a simple application of the constant of motion, characterizing the motion in the frame rotating with the field, predicts the threshold field dependence on the frequency and, contrary to the experiment, it quickly decreases with growing frequency. In another recent paper, using a classical phase-space averaging method [5], Griffiths and Farrelly [6] confirmed the experimental observation [3] that the departure from circular polarization towards elliptic polarization quickly decreases the threshold field. In Ref. [7] the existence of the approximate constant of motion has been demonstrated, which helps to label the eigenstates of the quantum Hamiltonian.

In the present paper we examine in some detail the classical dynamics of the Rydberg state in the circularly polarized microwave field. We confine our attention to the maximum angular-momentum circular orbits placed in the polarization plane. Hence, the problem is merely two dimensional. There were some earlier studies of the classical dynamics for this case [8]. For the most part the different points of the phase space of the orbit are equivalent and minimum attention needs to be paid to the phase-space averaging. And yet, even this simple problem has many unexpected features.

Our starting point is simply a Newton equation for the electron moving under the influence of the Coulomb force and the force of the microwave field of amplitude E and frequency ω. We use atomic units:

$$\ddot{x}=-\frac{x}{r^3}+E\cos\omega t\ ,\qquad \ddot{y}=-\frac{y}{r^3}+E\sin\omega t\ . \tag{1}$$

We then transform our equations to the noninertial frame rotating with the frequency of the field and in the same direction as the field,

$$\ddot{x}'=-\frac{x'}{r^3}+E+\omega^2x'+2\omega\dot{y}'\ ,\qquad \ddot{y}'=-\frac{y'}{r^3}+\omega^2y'-2\omega\dot{x}'\ . \tag{2}$$

These equations have an obvious constant of motion:

$$\mathcal{E}=\frac{(\dot{x}'^2+\dot{y}'^2)}{2}-\frac{1}{r}-Ex'-\tfrac{1}{2}\omega^2r^2\ . \tag{3}$$

Hence, in spite of the nonpotential Coriolis force, the initial conditions for which the constant $\mathcal{E}$ has its value below the saddle point of the potential $V(x',y')$,

$$V(x',y')=-\frac{1}{r}-Ex'-\tfrac{1}{2}\omega^2r^2\ , \tag{4}$$

generate trajectories which are confined to the interior of the potential and will never be ionized.

So far we just followed Ref. [4] but now we depart from that reference. Suppose we deal with a circular orbit of the Rydberg type, of principal quantum number n. Its energy is $E_n=-1/2n^2$. Transforming the initial condition, the constant $\mathcal{E}$ is

$$\mathcal{E}=E_n-\frac{\omega}{\sqrt{2|E_n|}}+\frac{E\cos\varphi}{2|E_n|}\ , \tag{5}$$

where φ is the difference between the initial phase of the field and the phase of the motion on the orbit at the initial moment. If the frequency of the applied field, ω, is positive, we deal with the electron moving in the same direction as the field. Negative ω describes the situation of the two motions in opposite directions. It is easy to

see why (for positive ω) the electron sinks deeper into the potential well (sign of the second term). This is because in the rotating frame it moves slower, hence its kinetic energy is smaller but its (negative) potential energy is the same since the distance from the nucleus has not changed. It makes the sum smaller.

In most cases, we introduce the excited Rydberg atom into the field. Hence we deal with smooth pulses. So in the initial moment the field amplitude is zero and the initial value of our constant of motion has no contribution from the last term in (5) and is independent of the point on the orbit. So the initial value of the constant characterizes the whole orbit.

Now we may apply the saddle-point criterion. All orbits with constant $\mathcal{E}$ below the rim of the potential are confined. This way we get the dependence of the threshold field on the applied frequency represented by a solid line in Fig. 1. The threshold field at zero frequency is of course $1/16n^4$ (in figures we omit the Coulomb scaling dependencies on the principal quantum numbers, so the field should be divided by n^4 and the frequency by n^3). For positive frequencies it grows a little and then tends to zero at Kepler frequency $1/n^3$. For negative frequencies it goes to zero very quickly and reaches zero at the frequency which is only $\frac{1}{8}$ of the Kepler frequency. The resulting dependence differs from the one obtained in [4]. The difference is caused by the second term in the formula (5) which is present because we deal with the circular orbit.

But physics is somewhat more complicated since we deal with pulses. Fields are time dependent making $\mathcal{E}$ time dependent. To account for that we have performed a series of computer runs of the original Newton equations (1) for the smooth pulse with the envelope given by

$$f(t)=\sin^2\left[\frac{\pi t}{T}\right], \tag{6}$$

where T is the duration of the pulse. The results of these runs are indicated as black triangles in Fig. 1. The observations may be summarized as follows.

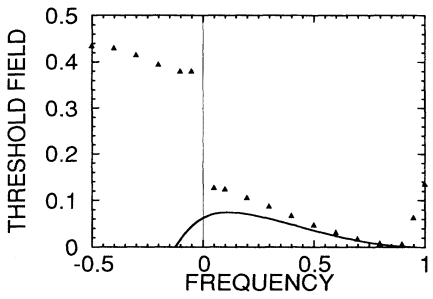

FIG. 1. Threshold amplitude of the microwave field for the ionization of the circular Rydberg orbit as a function of the frequency. The solid line represents the application of the saddle-point argument. The triangles mark break-up points for the sine square pulse. Note the dramatic difference between positive and negative frequencies. There are no data at $\omega=0$ because different points on the orbit ionize at different field in this case. There are also nonionizing trajectories for frequencies greater than 0.4 above triangles.

(i) For the whole range of frequencies studied, the threshold intensity for the smooth pulses is higher than predicted by the saddle-point estimate.

(ii) If the length of the pulse is sufficiently long, then the threshold field is independent of the initial point on the orbit and of the duration of the pulse. We tested it using up to 100 randomly chosen phases. The system behaves adiabatically. Even very short pulses are adiabatic. In fact almost all triangles in Fig. 1 correspond to pulses lasting only 100 Kepler periods. There are two exceptions. At the frequency -0.1 we needed 200 Kepler periods and at -0.05 we needed 400 Kepler periods to get the phase-independent result. At this frequency it is only 20 cycle pulses. The case of static field viewed from the point of our numerical experiment is fairly singular especially when approached from negative frequencies. Obviously the notion of an adiabatic pulse has to break down for sufficiently small frequencies. Indeed, at zero frequency we got different threshold fields from different points in the phase space. This case would need a genuine ensemble average. However, it corresponds to a separable Hamiltonian and has been studied in detail in the past [9]. Different points of the given energy manifold in the phase space indeed ionize at different field strengths with the highest threshold field equal to 0.38. Note that *outside* of the region of static field we find that *the whole* realizable classical orbits have the same escape thresholds [14] but also that for the negative frequencies this threshold *exceeds* the maximum threshold field for the static electric field.

(iii) We also studied in some detail the dependence of the threshold field on the pulse shape. For the positive frequencies we find the results hold also for the trapezoidal shape pulses lasting 100 Kepler periods with 10% rise time 80% of constant amplitude and 10% fall. However, for negative frequencies such pulses at the same length are not adiabatic enough and considerably lower thresholds are obtained.

The further analysis is very different for the positive and negative frequencies. To explain this we looked at the time dependence of $\mathcal{E}$. It is shown in Fig. 2. As we see, this "constant" changes smoothly and slowly for the nonionizing trajectories. In fact for positive frequencies $\mathcal{E}$ is pulled deeper into the potential. The broken lines

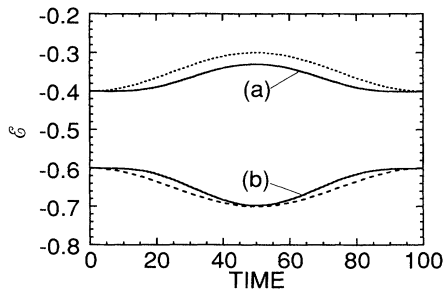

FIG. 2. Adiabatic change of $\mathcal{E}(t)$ for the sine square pulses. The duration of the pulse was 100 Kepler periods, the peak field was 0.1. Compared are cases of $\omega=\pm 0.1$: (a) the negative frequency, (b) the positive one. Note the opposite signs of the variation. Dashed lines represent the approximate formula (7).

represent a simple adiabatic approximation to $\mathcal{E}(t)$:

$$\mathcal{E}(t)=E_n-\frac{\omega}{\sqrt{2|E_n|}}-\frac{\omega}{|\omega|}\frac{Ef(t)}{2|E_n|}\ . \tag{7}$$

This neatly explains why the orbit can withstand stronger fields than that predicted by the saddle-point argument for the cw field. In fact the threshold field for pulses agrees with taking the minimum value of $\mathcal{E}$ just below the escape point and using it for the saddle-point argument. Consistent with the above is the finding that the threshold field for the square pulses is almost identical with the constant field predictions represented by the solid line in Fig. 1.

The picture gets more complicated in two regions: for the field frequency approaching the Kepler frequency, the threshold field rapidly grows exhibiting the classical resonance. This region is likely beyond the validity of the classical picture and need not exhibit the same properties in quantum simulations. Much more surprising, however, is the dramatic increase of the threshold field or even very small negative frequencies. Moreover, $\mathcal{E}(t)$ is *growing* during the pulse, as shown in Fig. 2, so the threshold field in this case has nothing to do with the saddle-point argument. Also for negative frequencies this argument does not predict correctly the behavior of the system interacting with the square pulse. This is all at frequencies much smaller than the Kepler frequency, where the classical equations are generally considered to describe correctly the dynamics of the atomic system.

To get more insight into the dynamics we looked at the dependence of the final state on the peak amplitude of the field. The typical results are those in Figs. 3(a) and 3(b) where we have plotted the final energy and the final distance of the electron for the frequencies ± 0.1 and somewhat longer pulses of 400 Kepler periods or 40 field periods. Below the threshold the initial circular orbit is regained with a remarkable accuracy. Note that both threshold amplitudes are the same as in Fig. 1, where we have used a shorter pulse. They are both considerably higher than for the static field. The one for negative frequency is more than a factor of 5 larger. It means that the atom withstands without any effect a short pulse with the peak strength of about 40% of the Coulomb field. The very precise return to the initial state agrees with the conjecture [3] that according to quantum mechanics, below the threshold the system goes during the evolution adiabatically over a single eigenstate of the coupled Hamiltonian in the rotating frame.

To summarize, we find that, unlike in the case of the linearly polarized field (and also the static field), all points of the circular orbit of the Rydberg level become unstable at the same field amplitude of the circularly polarized low-frequency smooth pulse. Defined this way threshold amplitude depends dramatically on the angular momentum of the orbit. On an example of the maximum angular-momentum circular orbits we showed that if the electron rotates in the direction opposite to the field, the simple saddle-point argument does not work. The ionization threshold in this case is much higher if the ionizing pulse is sufficiently smooth. In fact, it exceeds even the highest threshold amplitude obtained for the static field. We also showed that below the ionization threshold the system returns exactly to its initial circular orbits.

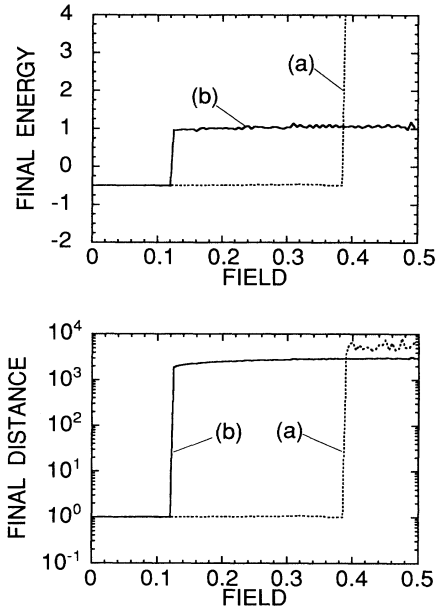

FIG. 3. Final-state energy (a) and final distance from the nucleus (b) for the interaction of circular orbit with the 400 Kepler periods with long pulses of varying peak amplitude. Note that below the ionization threshold the initial values (-0.5 and 1) are reproduced with great accuracy. (a) is the negative frequency (above the threshold is numerically unstable), (b) is the positive frequency.

Triggered by the present results, quantum-mechanical calculations have just been completed [10]. They yield results which are qualitatively similar to the classical ones. In particular the strong asymmetry between the positive and negative frequencies is observed with much higher threshold for the negative frequencies.

It would be interesting to test the above predictions in the experiment. In fact several schemes have been proposed to produce the circular Rydberg states [11] and some of them were even successfully implemented [12].

It is also worth noting the similarity of the results of this paper to the results obtained for the dependence of the, so-called, appearance intensities on the frequency for a multiphoton ionization of atoms by a powerful circularly polarized laser field [13].

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