

Hydrogen atom in circularly polarized microwaves: Chaotic ionization via core scattering

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(Received 19 November 1996)

The ionization of hydrogen atoms in a circularly polarized (CP) microwave field is studied using classical mechanics and simulations. We first focus on the ionization of circular orbits in a model two-dimensional atom for which there have been previous simulations and studies. It has been suggested that in the ionization of both circular and elliptical states collisions with the nucleus play only a minor role. However, the ionization of circular states, in particular, is known to be strongly dependent on the details of the microwave pulse shape. We explain this observation and show that, in some cases, essentially the entire initial ensemble of atoms is ionized during the rise time of the microwave field. Under these conditions, atoms are switched directly into unbound parts of phase space and promptly scatter to infinity without collisions with the nucleus—succinctly, the rise time of the pulse is responsible for the vast majority of ionization events. We identify the conditions leading to this situation based on analytical and numerical studies of the behavior of the Jacobi constant K (energy in the rotating frame) during the rise time of the field. A large number of classical simulations are presented not only for initially circular states but also for states of medium eccentricity. For orbits that survive the pulse rise time these computations support a model of ionization that involves collisions with the core. We argue that the complexity of the ionization of hydrogen atoms in CP microwaves can be ascribed to the fact that orbits of different initial eccentricity in the original ensemble are not only switched to different regions of phase space, but also to different values of the Jacobi constant, during the rise time of the pulse. [S1050-2947(97)02205-1]

PACS number(s): 32.80.Rm

I. INTRODUCTION

In pioneering experiments, Gallagher and co-workers [1] observed the ionization of (sodium) Rydberg atoms by circularly (CP) and elliptically (EP) polarized microwave fields (frequency $\omega = 8.5$ GHz) (see also [2] and [3]). These experiments showed a strong dependence of the ionization threshold on polarization but found that the CP threshold field F goes approximately as $1/16n^4$ (n is the principal quantum number) as in the static case—i.e., the Stark effect. Fu *et al.* [1] explained their CP results by going to a rotating frame and proposing that, in this frame, ionization proceeds in roughly the same way as for a static field, i.e., a static field has the same effect whether its coordinate system is rotating or not. Subsequently Nauenberg [4] argued that the ionization mechanism was substantially more complicated and that the effect of rotation on the ionization threshold must be taken into account. Ever since, the theory of the CP problem has been surrounded by controversy but, as with most such cases, it has also been a vehicle for the discovery of new phenomena. For example, it has been suggested that it might be possible to create nonstationary, nondispersive electronic wave packets in Rydberg atoms using CP fields [5], possibly in combination with magnetic fields [6,7]. These states are associated with equilibria that are created by the CP field and support stable motion in much the same fashion as gravitational equilibrium points in the restricted three-body problem of celestial mechanics [5,8,9]. The similarity is so close to the dynamics of the Trojan asteroids that the term “Trojan wave packet” has come to be associated with these states. Here is not the place for a comprehensive review of the CP

problem although, after the discussion and remarks of Zakrzewski, Gębarowski, and Delande [10], the time is ripe for an objective overview of the field.

In fact, the paper by Zakrzewski, Gębarowski, and Delande [10] brings into sharp focus a controversy that has been brewing for some time: simply put, must an electron collide with the nucleus to ionize in a CP field? Indeed, Ref. [10] is sprinkled with statements to the effect that core collisions are not relevant to ionization in the high-frequency CP problem and the paper concludes by asserting that “... the fact that states of medium eccentricity are easiest to ionize confirms that *collisions with the nucleus play a minor role* in the CPM (i.e., CP microwave) ionization of elliptical and circular states” (italics and parentheses ours). This clearly implies that not only are core collisions *not needed* for ionization but that they are *irrelevant* in the ionization of all except linear orbits. Similar conclusions are drawn in Ref. [11] and this view is shared by other researchers who, based on the Kepler map [12,13], also claim the existence of a new ionization process in which the electron in the CP problem picks up energy while it is remote from the nucleus and then ionizes directly [14–16]. It should be noted that these latter authors [14,16] do not, however, go so far as to propose that this channel is dominant. In this paper we adopt the usual definition of a collision, i.e., a particle is scattered by some scattering center while obeying conservation laws (e.g., angular-momentum barriers). As in conventional collision theory, such an encounter is considered to be a collision even if the distance of closest approach is not zero.

Of course, it is counterintuitive to imagine ionization

without collisions: e.g., in the linearly polarized (LP) microwave system core collisions are the dominant ionization mechanism [17–19] and conservation of angular momentum makes ionization of linear orbits particularly efficient. In contrast, in the planar CP problem, angular momentum (i.e., the quantum number m) is not conserved and linear, or nearly linear, orbits turn out to be relatively stable, despite frequent collisions with the nucleus. It should be noted that positing that collisions with the core are necessary for ionization does not imply that collisions, or close encounters, with the nucleus are *sufficient* to cause ionization. After all, linear orbits in the hydrogen atom itself collide with the nucleus without ionizing: actually, in the CP problem the linear orbits are strongly coupled to the nucleus and somewhat adiabatically decoupled from the CP field, hence their stability [20].

Some perspective can be gained by considering recent work using half-cycle pulses (HCP) that has only reemphasized the role of core collisions in the ionization of Rydberg atoms [21]. The point of HCP experiments is to craft a pulse that circumvents the need for core collisions as a mechanism for energy transfer. Unlike a laser pulse, the electric field of an HCP pulse does allow momentum transfer to an essentially free electron that is remote from the nucleus. In light of this widely grasped fact it seems surprising that an almost-free electron in a CP field can absorb energy and ionize even though the momentum impulse communicated to a free electron by the electric field $\mathbf{F}(t)$, i.e.,

$$\Delta \mathbf{p} = \int_{-\infty}^{\infty} \mathbf{F}(t) dt \quad (1)$$

is zero.

Other researchers, including ourselves, have maintained that collisions with the core are necessary for the atom to ionize [8,15]. Early insight into this problem came from an unexpected quarter: Mignard's [22] studies of an analogous problem in planetary physics, namely, dust orbiting a planet while simultaneously subject to radiation pressure. The similarity of the two problems can be seen most closely by consulting Deprit [23], who did not fail to recognize the connection with atomic physics and who discusses in some detail the nuances of Hamiltonian dynamics in a rotating frame [24]. Mignard's research highlighted collisions with the atmosphere as a means of depleting a planetary dust ring. The objective of this paper is to answer the question (for a hydrogen atom in a CP field): "Must an electron suffer collisions with the nucleus to ionize?"

At the outset we wish to make clear that the answer depends critically on the conditions under which the experiments or simulations are performed—a point emphasized by Kappertz and Nauenberg [25]. Regrettably, the issue has been somewhat clouded by a failure to take details of state preparation into account, especially a failure to distinguish the following situations [10]: (i) ionization of an electron that is continuously subjected to a CP field (the so-called continuous wave, or "CW," limit) and (ii) ionization during the CP field rise-time and turn-off phases of the experiment. Simulations that seek to mimic experiments must, of necessity, employ some rise time and turn off of the microwave field. This involves selection of the initial ensemble of states

followed by a decision about what shape of pulse to use, how long the pulse should be on, and the criteria used to decide if an electron has ionized. Indeed, for the long pulses typically used in CP experiments it is critical that ionization during the "flat-top" part of the pulse be understood, i.e., how does an electron ionize while the CP field is still "on", as distinct from ionization during the rise or fall of the pulse?

Another point of possible confusion concerns the relationship of initial orbital parameters to the orbits that are prepared at the end of the pulse rise time. Clearly orbits of the initial Kepler problem are linear, circular, or elliptical. Because angular momentum is not conserved when the CP field is turned on, it is no longer possible (for strong fields) to use the language of perturbation theory and refer to linear, elliptical, and circular orbits as if such orbits necessarily grow smoothly out of the Kepler limit. The topologies of orbits in the CP problem are, for the strong-field strengths and high frequencies used, determined by nonlinear mechanisms and by the pulse shape and may not respect the initial eccentricity of a Kepler orbit in the original ensemble. We argue that the complexity of the ionization of hydrogen atoms in CP microwaves can be ascribed to the fact that orbits of different eccentricity in the original ensemble are not only switched to different regions of phase space during the pulse rise time, but also, to different values of energy in the rotating frame (Jacobi constant). At the conclusion of the switching process each orbit emerges into phase space with its own Jacobi constant. Because the distribution of regular, chaotic, and scattering regions in phase space itself depends in a complex way on (i) the Jacobi constant, (ii) the field strength, and (iii) the field frequency, it is difficult to predict the ultimate fate of an orbit based on a knowledge only of its initial eccentricity. To backup these statements we map the parts of phase space to which the pulse takes the initial ensemble.

Before launching into the details, we wish to present our resolution to this problem in general terms. Any electron that survives the rise time of the pulse can only escape either (i) by colliding with the nucleus directly or (ii) by colliding with a barrier that shields the nucleus from direct hits. This barrier is much like a potential energy barrier, except it defines a region where the *mechanical velocities* rather than the momenta become imaginary, i.e., it is a barrier in a *zero-velocity surface*. In the latter case we find that the radius of the orbit can extend to many hundreds of times the distance of the closest approach between successive collisions before ionization finally occurs. The ionization mechanism is, thus, fundamentally collisional. How hard of a collision the electron needs to suffer depends, in principle, on how close it is to the ionization threshold. Unfortunately, in the CP problem there has been no prior analysis of what that threshold actually is. The presence of nonconserved velocity-dependent Coriolis forces rules out naively examining a potential energy to determine the threshold. A component of our argument is, therefore, to identify an asymptotic ionization threshold. If an electron lies well below this threshold it either needs to collide many times with the core or to undergo one or a few hard collisions to ionize. Electrons that, at the end of the rise time, are marginally below the threshold need only make a soft collision (i.e., a collision at relatively large distances) with the core to ionize. We find that, for the circular orbits and field parameters used in Ref. [11], the *bulk* of the atoms

are ionized directly during the pulse rise time and those that survive, in most cases, lie so close to the threshold that only a relatively soft collision with the nucleus is required. We pause to note that there is, of course, no paradox associated with an electron being ionized during the rise time (or turn off) of the pulse. After all, the pulse might switch an electron directly into the exit channel after which it has no choice but to ionize promptly. This, in fact, turns out to be a distinct possibility in the CP system where electrons can ionize so promptly that some orbits only rarely intersect a Poincaré surface of section (SOS, singular and plural).

A final issue relates to the integrability of this system: in a rotating frame the problem has two integrable limits, the Stark and linear Zeeman limits [26]. The transition between these extreme cases is quite complicated but at the end of the pulse rise time it is critical to understand, for whatever value, the Jacobi constant K is switched to by the pulse, whether one is in a region of chaos, close to one of the integrable limits, or in an intermediate regime. This can be understood by examining SOS at fixed K values. However, because different initial orbits (e.g., circular vs elliptical), and, possibly, even different points along the *same* orbit, end up with different K values, it is not possible to use a single SOS to describe the ultimate fate of the initial ensemble. By examining SOS for individual orbits (using their particular K values at the end of the pulse rise time) we are able to draw some general conclusions about the ionization of low, medium, and high eccentricity orbits under specific field conditions. We emphasize, however, that *no single set of all-purpose conclusions or rules of thumb* need apply in all situations. However, the theoretical framework and numerical recipes we present might prove useful as a “tool box” to use in understanding particular sets of experimental or numerical data.

The paper is organized as follows: Sec. II introduces the Hamiltonian and a consideration of the asymptotics of the ionization channel in the rotating frame. This leads to specific predictions as to when the atom will ionize. In particular, we show how, for circular orbits, to obtain analytical estimates for the energy in the rotating frame at the end of the pulse. This allows us to construct SOS that are salient to the explicitly time dependent simulations performed in the nonrotating frame. Section III is given over to a series of classical simulations that consider ionization caused by the way the CP field is turned on and off over a finite period. In particular, we analyze the times at which circular trajectories ionize in relation to the pulse duration using the model presented in Sec. II. These simulations show that collisions are necessary for ionization unless the pulse prepares atoms in direct-scattering regions of phase space. Again, the mechanism that we propose in Sec. II is used to explain our results which are then placed into perspective by comparison with extant computations. Although our emphasis is on the ionization of circular states in the high-frequency regime we also consider ionization of initial orbits of medium eccentricity. Conclusions are in Sec. IV.

II. HAMILTONIAN AND ASYMPTOTICS

In this paper we concern ourselves only with the relative motion for a hydrogen atom (in the limit of an infinitely

massive nucleus) subjected to a CP microwave field (field strength F and angular frequency ω). In atomic units ($m_e = \hbar = e = 1$) the Hamiltonian is given by

$$H = E(t) = \frac{1}{2} (p_x^2 + p_y^2 + p_z^2) - \frac{1}{r} + f(t)F(x \cos \omega t + y \sin \omega t), \quad (2)$$

where ω is the microwave frequency and $f(t)$ controls the rise time and turn off of the pulse and will generally be taken to describe a “flat-top” shape, i.e.,

$$f(t) = 1, \quad t_1 < t < t_2. \quad (3)$$

We will give explicit functional forms of $f(t)$ during the rise-time and turn-off stages later. In some situations we will assume that $f(t) = 1$ for all t . Throughout we work in scaled units $E \rightarrow E/n^2$, $F \rightarrow n^4 F$, $\omega \rightarrow n^3 \omega$, $t \rightarrow t/n^3$, but for convenience will not change the symbols for these quantities. The time dependence arising from the CP field [rather than from $f(t)$] in Eq. (2) can be eliminated by going to a frame that rotates at the constant angular velocity ω . This leads to the Hamiltonian

$$K = \frac{1}{2} (p_x^2 + p_y^2 + p_z^2) - \frac{1}{r} - \omega(x p_y - y p_x) + f(t)F x, \quad (4)$$

where, again for notational simplicity, x, y, z , etc. are now taken to refer to the rotating frame—Hamiltonians denoted by the letter K will always be in a rotating frame. K has dimensions of energy in the rotating frame. In the limit that $f(t)$ is constant (e.g. during the “flat-top” part of the pulse) K is a constant of motion analogous to the Jacobi constant in celestial mechanics [9]. In fact, the behavior of K is central to the ionization process. At this point we specialize to the planar limit $z = p_z = 0$ for a number of reasons: In several studies [8,10,25] the classical and quantum mechanics of H and K have been investigated in the x - y plane because: (i) a particle started out in the plane $z = 0$, with $p_z = 0$ will remain forever in that plane, (ii) the classical dynamics in the plane can be studied using SOS, and (iii) ionization might be expected to occur along the electric-field direction, as in the Stark effect [1]. In particular, the calculations in Ref. [11] are performed in the two-dimensional (2D) planar limit. We emphasize, however, that our simulations use a 3D code based on the technique of regularization of the 3D Hamiltonian in extended phase space [27]. The planar Hamiltonian is, in the rotating frame,

$$K = \frac{1}{2} (p_x^2 + p_y^2) - \frac{1}{r} - \omega(x p_y - y p_x) + f(t)F x \quad (5)$$

and our immediate task is to estimate the asymptotic ionization limit.

A. Ionization threshold

The calculation of an unambiguous ionization threshold is complicated by the fact that it is impossible to define a potential-energy surface in this problem because the Coriolis term in Eq. (5) is not conserved. In a sense to be explained, the threshold is a dynamical one that depends on the particu-

lar orbit involved. However, it is possible to deduce an asymptotic ionization limit. We begin with the problem in the rotating frame: consider Eq. (5) in cylindrical coordinates (ρ, ϕ) , where $\rho = \sqrt{x^2 + y^2}$, $\phi = \arctan(y/x)$, i.e.,

$$K = \frac{1}{2} \left(P_\rho^2 + \frac{P_\phi^2}{\rho^2} \right) - \frac{1}{\rho} - \omega P_\phi + F \rho \cos \phi \quad (6)$$

and the switching function $f(t)$ has been temporarily set to unity. Let us imagine an experiment in which, say, a circular orbit of the hydrogen atom is subjected to a CP field that rises from $t=0$ with some shape given by $f(t)$. At time $t=0$ the Jacobi constant K_0 will take a value defined by

$$E = K_0 + \omega P_\phi, \quad (7)$$

where E is the initial hydrogenic energy. During the rise time of the field both K and E will change with time until the CP field is fully switched on. At that point K will become a constant of the motion while E and P_ϕ will continue to vary. If the atom ionizes then $\rho \rightarrow \infty$ and the Hamiltonian, for finite P_ϕ , reduces to

$$K = -\omega P_\phi + F \rho \cos \phi, \quad (8)$$

which establishes a relationship between the Jacobi constant K , ω , F , and P_ϕ . Because P_ϕ is not conserved it seems impossible to tell if the atom will ionize or is even bound at the end of the pulse. A favorable ionization channel might be deduced by looking at the level curves of Eq. (8) which show that, along the line $\phi=0$ the angular momentum must rise roughly as fast as ρ to conserve K . However, along $\phi = \pi/2$ (the y direction) conservation of the Jacobi constant can be maintained as $\rho \rightarrow \infty$ simply by requiring that $P_\phi = -K/\omega$. Taking as an example $\omega=4$ and an electron initially on a circular orbit with $P_\phi=1$, then the initial Jacobi constant $K = -4.5$ ($E = -1/2$ in scaled units). Asymptotically the effective potential takes the value $-\omega P_\phi$. Because P_ϕ is not conserved when $F \neq 0$ this is only a rough guide to the ionization threshold and is strictly valid only in the zero-field limit; however, it indicates that once the Jacobi constant achieves the value $-\omega P_\phi$ or greater the electron might be asymptotically free. This analysis can be placed on a more rigorous footing by establishing a better estimate of the ionization threshold.

Consider again Eq. (5) in the asymptotic region where the Coulomb term can be neglected and $f(t)=1$

$$K = \frac{1}{2} (p_x^2 + p_y^2) - \omega (x p_y - y p_x) + F x. \quad (9)$$

The linear term in x can be removed by a canonical transformation

$$x = \xi + x_0, \quad p_x = p_\xi; \quad y = \eta, \quad p_y = p_\eta + p_0, \quad (10)$$

with $x_0 = F/\omega^2$, $p_0 = F/\omega$, giving

$$K = \frac{1}{2} (p_\xi^2 + p_\eta^2) - \omega (\xi p_\eta - \eta p_\xi) + \frac{F^2}{2\omega^2}, \quad (11)$$

and we may set the asymptotic constant of motion $P'_\phi = (\xi p_\eta - \eta p_\xi)$. Thus the ionization limit is given by

$$K = -\omega P'_\phi + \frac{F^2}{2\omega^2}. \quad (12)$$

In the original Cartesian coordinates

$$P'_\phi = (x p_y - y p_x) - \frac{F}{\omega^2} (p_y + \omega x) + \frac{F^2}{\omega^3}. \quad (13)$$

As soon as the quantity $Q = (K - F^2/2\omega^2)/P'_\phi > -\omega$ the electron is above the asymptotic ionization threshold and may, in principle, ionize directly. The time it actually takes to escape will depend on the precise value of K and the region of phase space that the electron finds itself in when the pulse enters its flat-top stage. At the end of the pulse rise time some atoms might fall in regular parts of phase space where the Kolmogorov-Arnol'd-Moser (KAM) tori abound, others in chaotic regions, and yet others in scattering regions for which the particle simply departs to infinity (just like starting out an electron in the hydrogen atom with energy $E > 0$). It stands to reason, however, that not all *initial* conditions will be bounded nor will all of them ionize: because the initial state is not prepared in the asymptotic region an *instantaneous* value of $Q > -\omega$ may not mean that the orbit will ultimately ionize because Q can oscillate wildly in the inner region. All of these points are best illustrated by actual examples.

To make a comparison with Ref. [11], we consider initially circular orbits with $\omega=4$ and $F=4.5$. The pulse used is as defined in Ref. [11], i.e.,

$$f(t) = \begin{cases} \sin^2(\pi t/2\tau) & \text{for } 0 < t < \tau \\ 1 & \text{for } \tau = t_1 < t < T - \tau = t_2 \\ \cos^2\{\pi[t - (T - \tau)]/2\tau\} & \text{for } T - \tau < t < T \\ 0 & \text{elsewhere} \end{cases} \quad (14)$$

although in some simulations, for which the flat top is long, we do not actually turn the pulse off. Further, this pulse shape may not always correspond as closely with the experimental situation as one might desire, but we adopt the same pulse as a previous work [11], again for comparison. The suite of pictures contained in Fig. 1 shows various orbit parameters for a trajectory that is deliberately chosen to be as similar as possible to the one in Fig. 5(c) of Ref. [11], i.e., an orbit that ionizes, apparently without colliding with or coming close to the nucleus. In this case, following Ref. [11], the pulse is not a flat top since $\tau=25$ and $T=50$ Kepler periods (Kepler periods will be assumed whenever values of τ and T are given). Figure 1(b) shows how the Jacobi “constant” [the function $K(t)$ will, from here on, be referred to as the “Jacobi function,” tempted as we are to call it the Jacobin] changes from its initial value as a function of time while Fig. 1(c) shows the radius of the orbit. Because the pulse has no flat top K never achieves a constant value. The main point is that the electron ionizes roughly at the top of the pulse. To clarify the situation we repeated the calculation using a flat-top pulse. This case is illustrated in Fig. 2 which shows the behavior of Q with time indicating that (i) it achieves constant asymptotic value, as predicted, and (ii) $Q > -\omega$ asymptotically for an ionizing orbit. This latter finding will be substantiated further in Sec. III. As shown in Fig. 1(c), upon

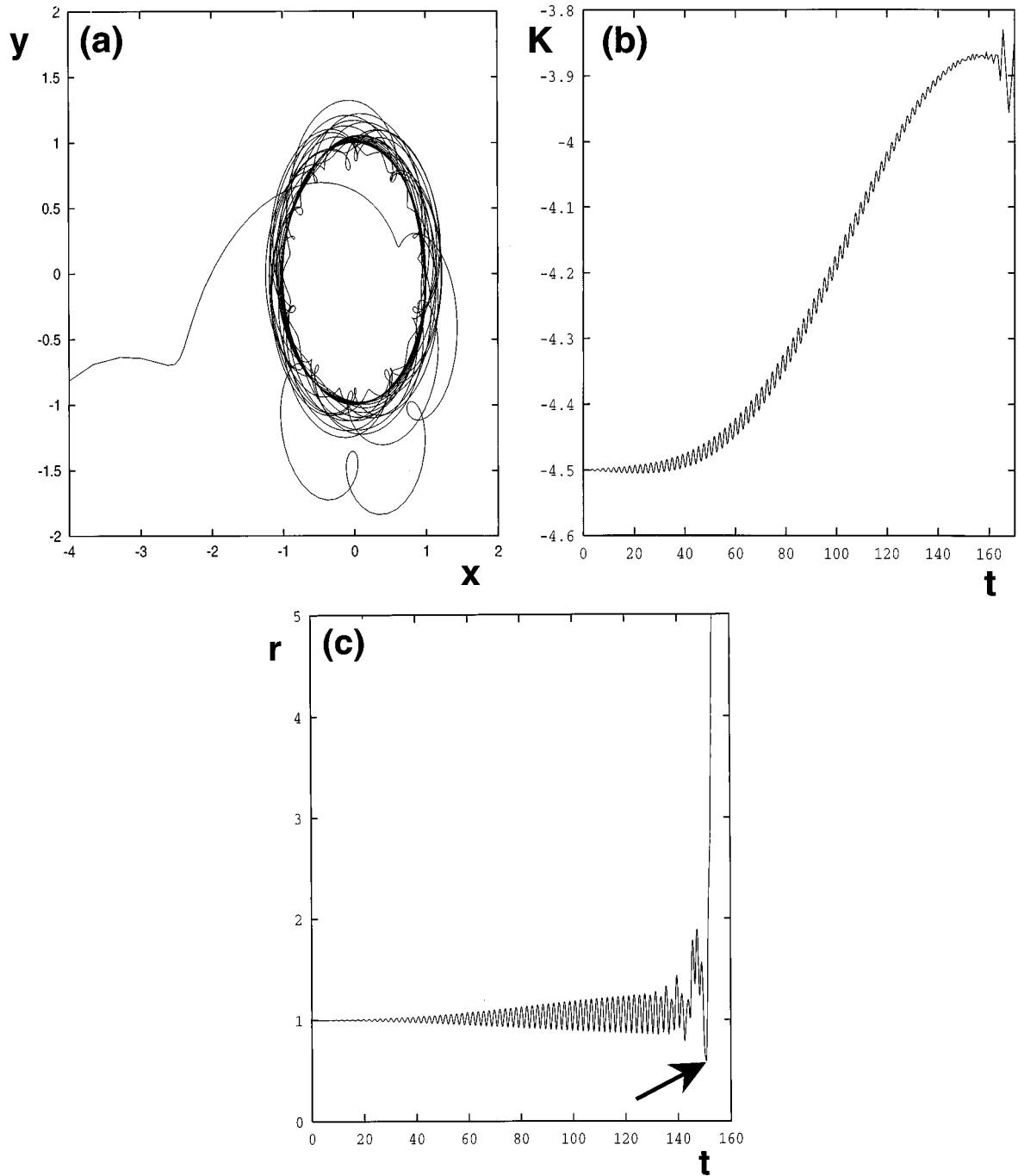


FIG. 1. Time evolution of orbit parameters with $\omega=4$, $F=4.5$, $\tau/2\pi=25$, $T/2\pi=50$ for an initially circular orbit ($P_\phi=1$); (a) coordinate space projection of the orbit, (b) Jacobi function, (c) the radius of the orbit. The pulse achieves its maximum value at $t \approx 157$ (scaled units).

ionization, the electron's radius increases smoothly after a final oscillation of the radius $r(t)$. The “time of ionization” we will use throughout the study is the time at which the electron experiences its closest approach to the nucleus immediately prior to ionization. This point is shown by an arrow in Fig. 1(c) and is characteristic of all ionizing trajectories. We obtain this “last point-of-closest approach” by back integrating a trajectory after it has ionized (i.e., entered the asymptotic region). The next issue, dealt with in Sec. II B, is the prediction of the value that K achieves at the end of the rise-time period.

B. Kapitza averaging: the Jacobi function $K(t)$ for circular orbits

It is apparent from an examination of Fig. 1(b) that the Jacobi function, at least in the high-frequency regime, has a shape quite similar to that of the pulse, and apart from high frequency oscillations it changes smoothly during the rise time of the pulse. This section shows how Kapitza's method [28] can be used to estimate the value the Jacobi constant will take at the peak of the pulse. In the rotating frame we may write

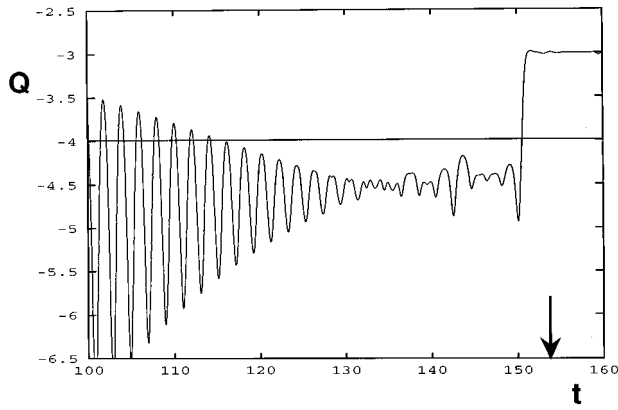


FIG. 2. Time evolution of the quantity Q for the same orbit as Fig. 1. The horizontal line is the asymptotic value as explained in the text.

$$K(t) = \frac{1}{2} \left(P_\rho^2 + \frac{P_\phi^2}{\rho^2} \right) - \frac{1}{\rho} - \omega P_\phi + f(t) F \rho \cos \phi \quad (15)$$

or

$$K(t) = K_0 + f(t) F \rho \cos \phi. \quad (16)$$

For a circular orbit we assume that $\phi \approx \Omega t$ where, in the rotating frame and in the high-frequency limit ($\omega \gg \omega_K$, with ω_K being the Kepler frequency) $\Omega \approx -\omega$. In scaled units the radius of the initial orbit $\rho = 1$. A straightforward application of the Kapitza averaging method to the Hamiltonian

$$K(t) = K_0 + f(t) F \rho \cos \Omega t, \quad (17)$$

where $f(t)$ is assumed to be slowly varying results in the expression for $K(t)$

$$K(t) = K_0 + \frac{F^2}{2\omega^2} f(t)^2. \quad (18)$$

Figure 3 compares this estimate with actual simulations for both a high- and low-frequency case assuming an initially circular orbit. The comparison is excellent for the high-frequency case, and, as expected, the approximation deteriorates at low frequencies. Actually, this approximation works rather well even for initially eccentric orbits in the high-frequency limit assuming their frequencies are well approximated by the mean motion. Armed with confidence in this result (in the high-frequency limit) we can, therefore, use the fact that, at the end of the rise time $f(t_1) = 1$ to arrive at the final value of K , i.e., $K(t_1) = K_0 + F^2/2\omega^2$. Since the ionization limit is given by $K = -\omega P'_\phi + F^2/2\omega^2$ then we can estimate the ionization threshold in terms of the initial value of the Jacobi constant K_0 , i.e., $K_0 = -\omega P'_\phi$. Unfortunately, it is not possible to estimate the value of P'_ϕ at the end of the pulse since this is not a well conserved quantity except in the asymptotic regime. Let us take an opposite point of view: asymptotically the ionization threshold is given by Eq. (12) and therefore the threshold value of P'_ϕ needed to ionize is $P'_\phi < -K_0/\omega$, which for the parameters $\omega = 4$ and $F = 4.5$ gives $P'_\phi < 1.125$. The very rough estimate $P'_\phi \approx P_\phi = 1$ indicates that *all* the initial conditions along a circular

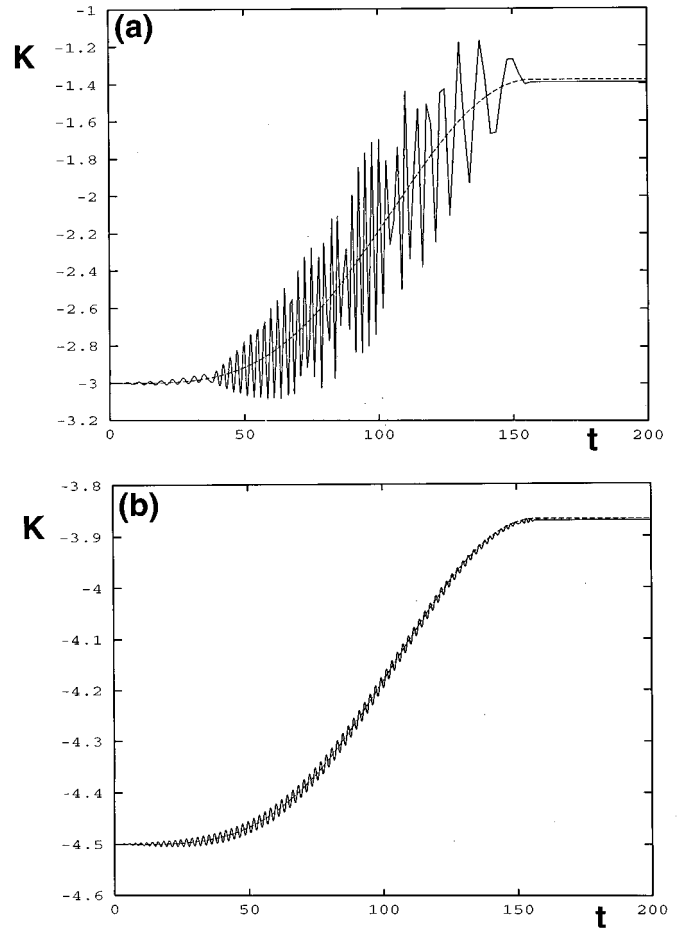


FIG. 3. Comparison of the change in the Jacobi function with time (oscillatory curve) with the estimate obtained using Eq. (18) (smooth curve) for (a) $\omega = 2.5$, $F = 4.5$, $\tau/2\pi = 25$ and (b) $\omega = 4$, $F = 4.5$, $\tau/2\pi = 25$.

orbit are being switched directly into an unbound region of phase space and thus all will ionize directly. This picture contrasts strongly with the conventional expectation that high angular momentum, initially circular states might be especially stable. In fact, their high angular momentum is part of the reason for their instability: without additional constraints these orbits essentially spiral out to infinity as we now discuss.

C. Zero-velocity surface and ionization threshold

In a previous publication we have examined in some detail the properties of the zero-velocity surface (ZVS) that governs much of the dynamics [8]. It is useful to review briefly the key points: the mixing of coordinates with momenta in Eq. (5) precludes the construction of a potential-energy surface in the usual sense. Nevertheless, it is possible to compute zero-velocity curves for this problem [9] defined by

$$\begin{aligned} \Xi(x, y) &= K - \frac{1}{2}(\dot{x}^2 + \dot{y}^2) \\ &= -\frac{1}{r} - \frac{\omega^2(x^2 + y^2)}{2} + Fx, \end{aligned} \quad (19)$$

where $\dot{x} = p_x + \omega y$ and $\dot{y} = p_y - \omega x$. It is instructive to consider first the case $F=0$ but in the rotating frame. Figure 4(a) shows a section through the ZVS along the x axis for the hydrogen atom. The ZVS has been likened to the caldera of a volcano and consists of an interior region (the “caldera”) and an exterior region. The top of the caldera is a circular rim. To make contact with the more familiar concept of the effective potential contained in Eq. (5), superimposed on the ZVS is a plot of $V(\rho)$ at fixed $P_\phi=1$ along the x axis for $\omega=1/2$. Note how the effective potential has a minimum that is contained inside the caldera. This minimum defines the location of circular orbits of the hydrogen atom in the rotating frame. In Figs. 4(b) and 4(c) are shown the corresponding plots with $\omega=1$ and $\omega=3$, respectively. Note how the minimum in $V(\rho)$ coincides exactly with the top of the rim in the ZVS when $\omega=1$ and has moved exterior to the caldera when ω is increased past the value $\omega=1$. This illustrates nicely the difference between the low- and high-frequency regimes. In particular, circular orbits of the hydrogen atom are transformed to the exterior of the ZVS when $\omega>1$. All of this is for the unperturbed atom viewed in a rotating frame of reference. The potential $V(\rho)$ is clearly not conserved when the electric field is turned on but these pictures do provide a rough guideline that explains certain features in the dynamics with nonzero fields.

With increasing F , the function $\Xi(x,y)$ develops two critical (equilibrium) points along the x axis with x being given by the two real solutions of the cubic equations: $\omega^2 x^3 - Fx^2 \pm 1 = 0$. The two equilibria are found to lie along the positive and negative x axis and are a maximum (M , associated with the Trojan states) and a saddle point (S) both of which are shown in Fig. 4(d). Any orbits that are switched to the interior to the ZVS cannot escape until the Jacobi constant achieves its value at the saddle point. As shown in [8] and discussed above it is also possible for orbits to exist *exterior* to the ZVS which may be stable or unstable. Initially circular orbits in the unperturbed hydrogen atom will tend to be switched to the exterior regions of the ZVS if $\omega>1$. It is now clear why these orbits may ionize directly: unless they are trapped by a dynamical potential such orbits may ionize by spiralling out to infinity. In some cases the minimum in the potential $V(\rho)$ can localize such states even in the presence of a nonzero field F . In essence, $V(\rho)$ persists as an approximate, adiabatic potential. Section III describes in more detail numerical simulations of the ionization process with an emphasis on such circular orbits.

III. NUMERICAL SIMULATIONS

The first 3D classical simulations of ionization of hydrogen atoms by CP and EP fields were performed by Griffiths and Farrelly using the Kustaanheimo-Stiefel transformation (KS) [27,29]. In this method the problem is regularized using new variables (involving a scaling of the real time by the radius r) to deal with the Coulomb singularity. The explicitly time dependent switching function $f(t)$ complicates the transformation, but this problem can be avoided by passing to an extended 10D phase space as done in [27]. The approach used in [27] is built on an earlier study of the quadratic Zeeman effect (QZE) by Saini and Farrelly [30] who performed adiabatic switching in regularized 2D *parabolic*

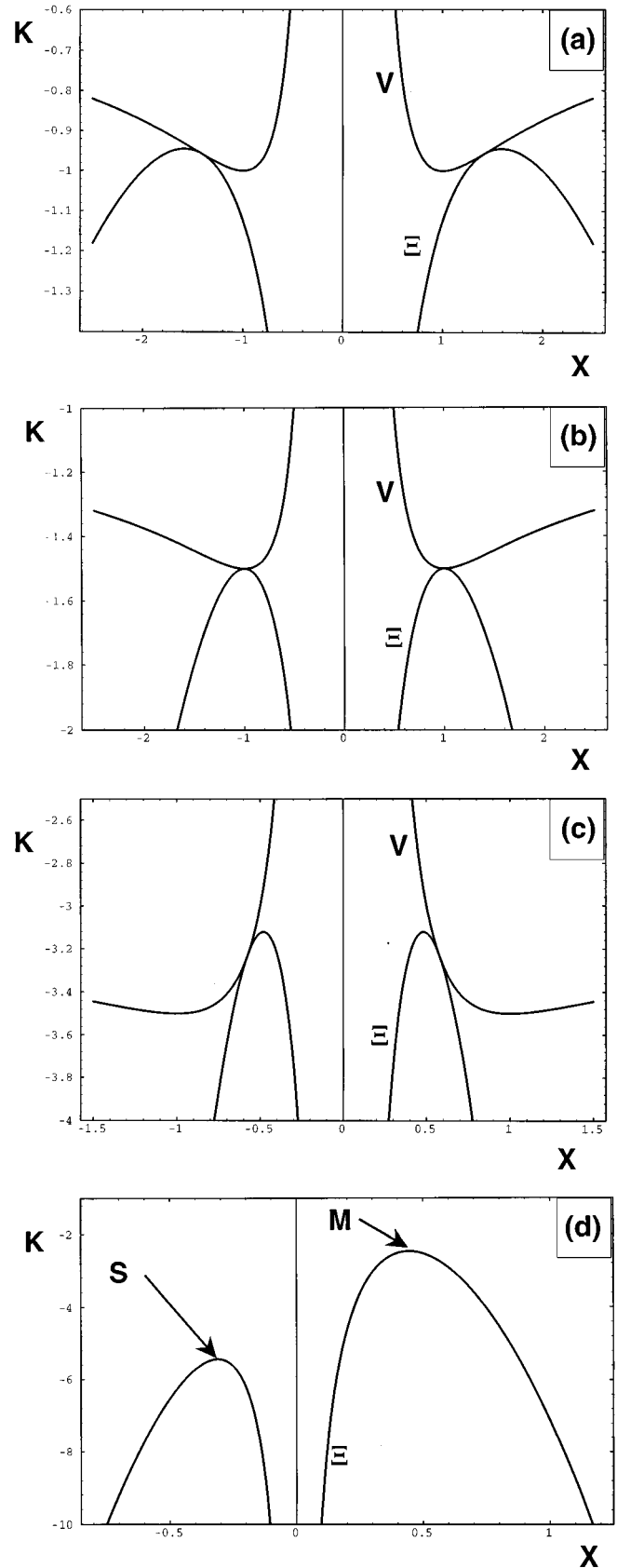


FIG. 4. Sections through $\Xi(x,y)$ in scaled units also showing in (a)–(c) the effective potential $V(x,y)$ for (a) $\omega=1/2$, (b) $\omega=1$, (c) $\omega=3$. In (d) the saddle (S) and the maximum (M) in the ZVS are illustrated for a nonzero electric field.

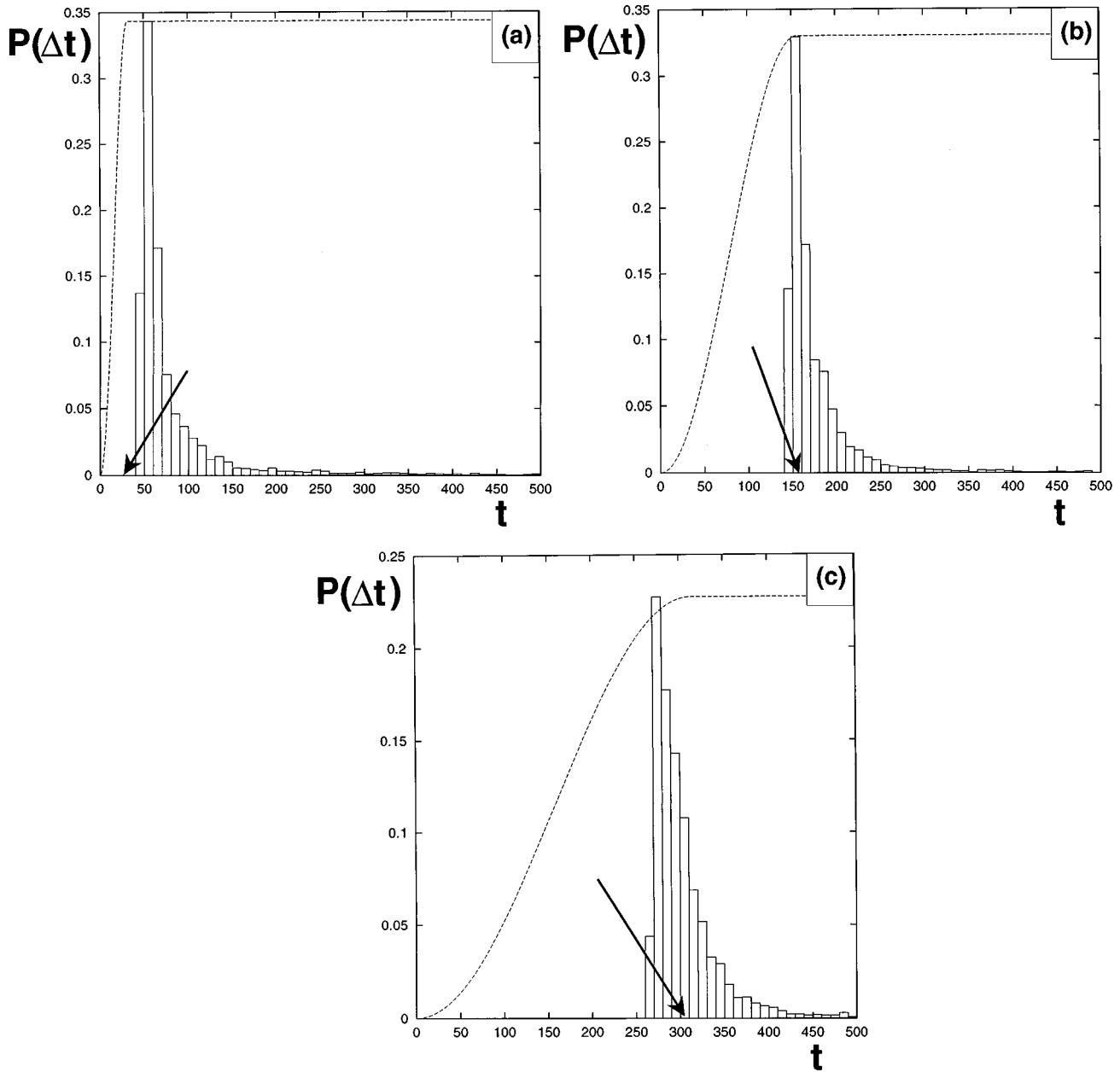


FIG. 5. Histograms showing the fraction P of trajectories ionized during consecutive intervals during the pulse for 5000 trajectories for field and pulse parameters as in Fig. 2 and (a) $\tau/2\pi = 5$, (b) $\tau/2\pi = 25$, (c) $\tau/2\pi = 50$. All initial conditions are chosen along a circular orbit. The time-box interval (Δt) is 10 scaled units. Superimposed is the pulse shape $f(t)$ which has been scaled so that its maximum coincides with the peak of the histogram. Its actual value at the end of the rise time is unity. The arrow indicates the time at which the rise time of the pulse is complete.

coordinates, also in an extended phase space (axial symmetry of the QZE avoids the need for the full KS treatment). Subsequently, Rath and Richards [31] performed calculations using the *full* KS transformation in extended phase space, for the same system as [30]—an account of the procedural details can be found in [31].

Following all of this work, although apparently unaware of it, Gębarowski and Zakrzewski [11] adopted the identical approach to that used in [27,29] in their studies of ionization by CP microwaves. Of course, these types of simulation, while valuable for providing a database, do not, in themselves, constitute a theory. We emphasize that our simulations are designed to test our analytical predictions and to

uncover a physical mechanism for ionization. This section is divided into two parts: the first studies ionization of circular orbits in the high-frequency regime using generally the same field parameters as Ref. [11], while the second is a study of the ionization of medium and low eccentricity orbits and a numerical investigation of the classical dynamics of the regular and chaotic regions of phase space.

A. Ionization of circular orbits

Figure 5 is a series of histograms showing the fraction $[P(\Delta t)]$ of orbits that ionize at particular times along the pulse (in intervals of Δt) for an ensemble of initially circular

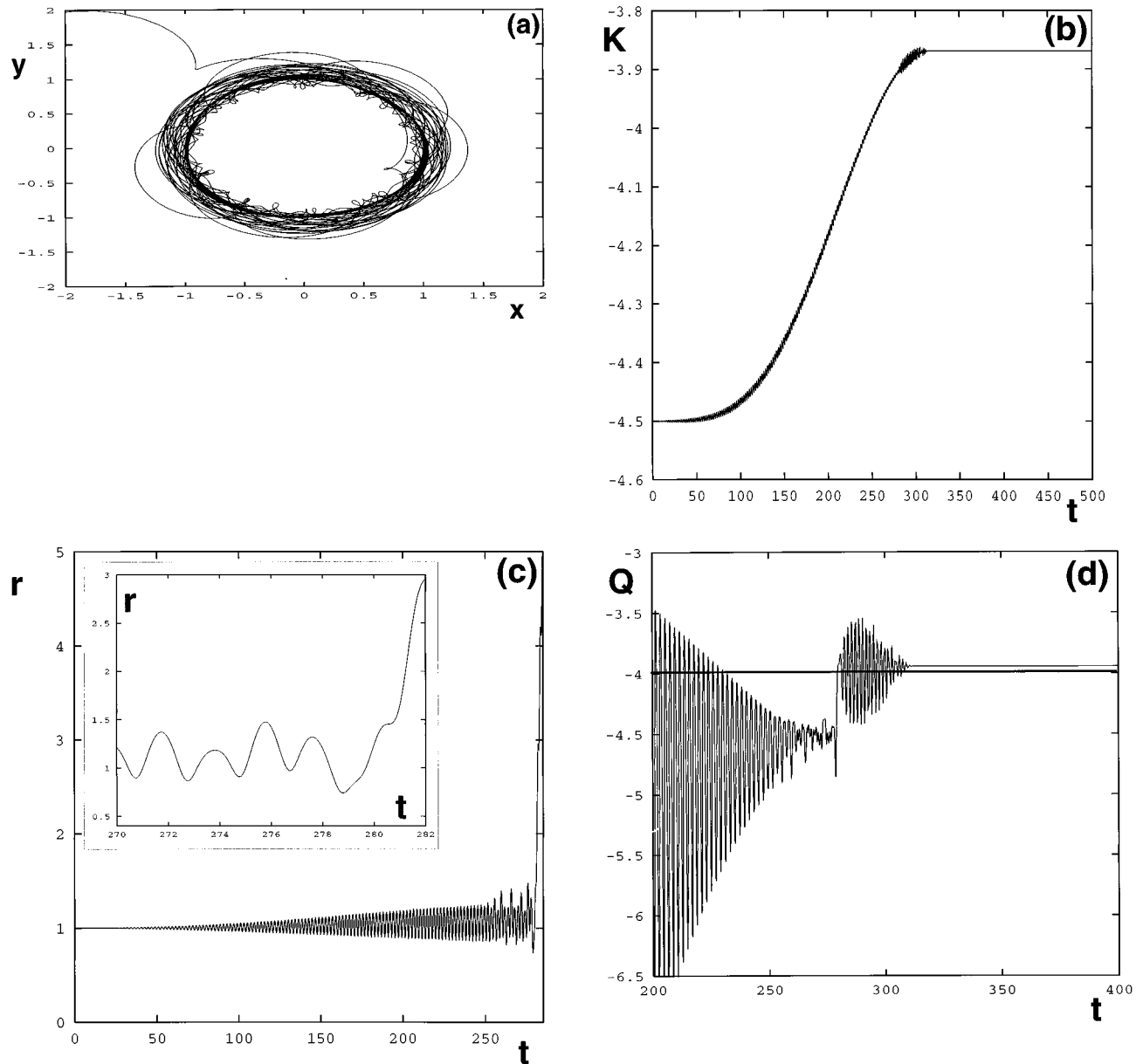


FIG. 6. Time evolution of orbit parameters for one of the outlying orbits in Fig. 5(b): (a) coordinate space projection of the orbit, (b) Jacobi function, (c) the radius $r(t)$. The detail in panel (c) is magnification of the radius at the time of ionization, (d) the quantity Q . The horizontal line is the asymptotic value as explained in the text.

orbits for the same field parameters as Fig. 1. Each of the panels corresponds to a different rise time, ranging from fast to slow switching. The “ionization time” was obtained as follows: each trajectory was integrated until it had clearly ionized. Ionization was diagnosed by monitoring (i) the radius $r(t)$ which, at ionization, increases rapidly, and, apart from high-frequency oscillations, almost monotonically, (ii) the quantity Q which assumes a constant value asymptotically [compare Fig. 2], and (iii) the quantity $r_p(t) = \sqrt{p_x(t)^2 + p_y(t)^2}$ which also, asymptotically, assumes a constant value. After it is decided that ionization has occurred the trajectory is back integrated to its last point of closest approach to the nucleus—see Fig. 1(c)—and this establishes the time of ionization. Superimposed on Fig. 5, and on all subsequent histograms, is the pulse shape (note that

the flat top, in this case, is not turned off). It is remarkable that, for essentially all of the orbits in Fig. 5 ionization occurs during or very shortly after the pulse rise time. This is not surprising given the initial distribution of Q values that is prepared by the pulse.

Nevertheless, some of the orbits clearly do not ionize during the rise time and so we examined the time evolution of one of the outlying orbits for the medium switching time in Fig. 5(b): various orbital quantities are shown as a function of time for this trajectory in Fig. 6. It is clear from Fig. 6(c) and the detail [which is a magnification of the radius $r(t)$] that immediately before ionization the orbit experiences a soft collision with the nucleus that causes it to ionize. The detail in Fig. 6 is, in fact, typical of the behavior of these orbits immediately prior to ionization. Note that the fairly

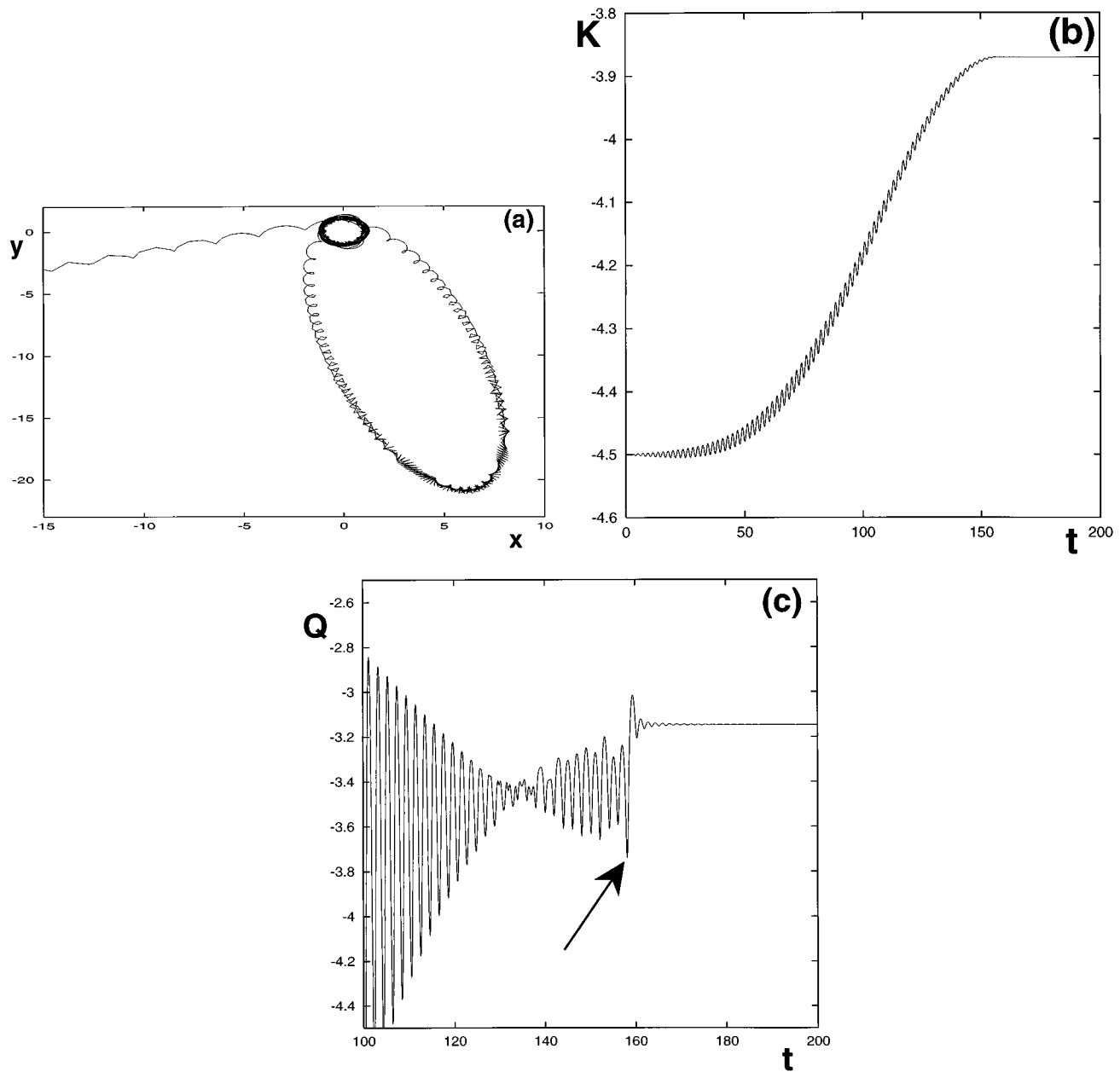


FIG. 7. Time evolution of orbit parameters for one of the orbits corresponding to Fig. 5(a): (a) coordinate-space projection of the orbit, (b) Jacobi function, (c) the quantity Q .

periodic motion of $r(t)$ is interrupted by a collision with the nucleus that causes the orbit to ionize. Such collisions or close encounters with the nucleus only *seem* unimportant because the pulse prepares most of the orbits above or very slightly below the asymptotic ionization threshold.

For a short rise time most of the orbits ionize very early in the flat-top part of the pulse rather than during the rise of the pulse—see Fig. 5(a). This occurs because the rise time is so fast: Fig. 7 shows the typical behavior of an orbit using a short sine-squared pulse: the electron almost ionizes [note, in Fig. 7(a) its large excursion from the nucleus] but again returns to the vicinity of the nucleus. Premature adjudication of its status might have led one to think the electron had ionized earlier than it did. If the pulse starts dying off during this excursion, thereby lowering the Q value of the orbit below the ionization threshold, ionization may be prevented.

This illustrates that the time scale of the pulse is faster than the time scale for ionization and accounts for the shift of the distribution into the flat top.

Figure 5(c), on the other hand, shows the limit of a much longer pulse rise time than that used in Fig. 5(b). The pattern is very similar to Fig. 5(b) except that an even larger fraction of the orbits ionize *during* the pulse rise time. Figures 5(a), 5(b), and 5(c) provide a striking overview of how the pulse rise time affects the time of ionization. Yet, in each case most of the ensemble is ionized during the pulse rise time or very shortly thereafter.

In these simulations all of the initially circular orbits achieve the same K value at the end of the pulse. For the present set of parameters this gives $K = -495/128 \approx -3.87$. Figure 8(a) is a composite SOS computed in the rotating frame by integrating a random set of initial conditions at

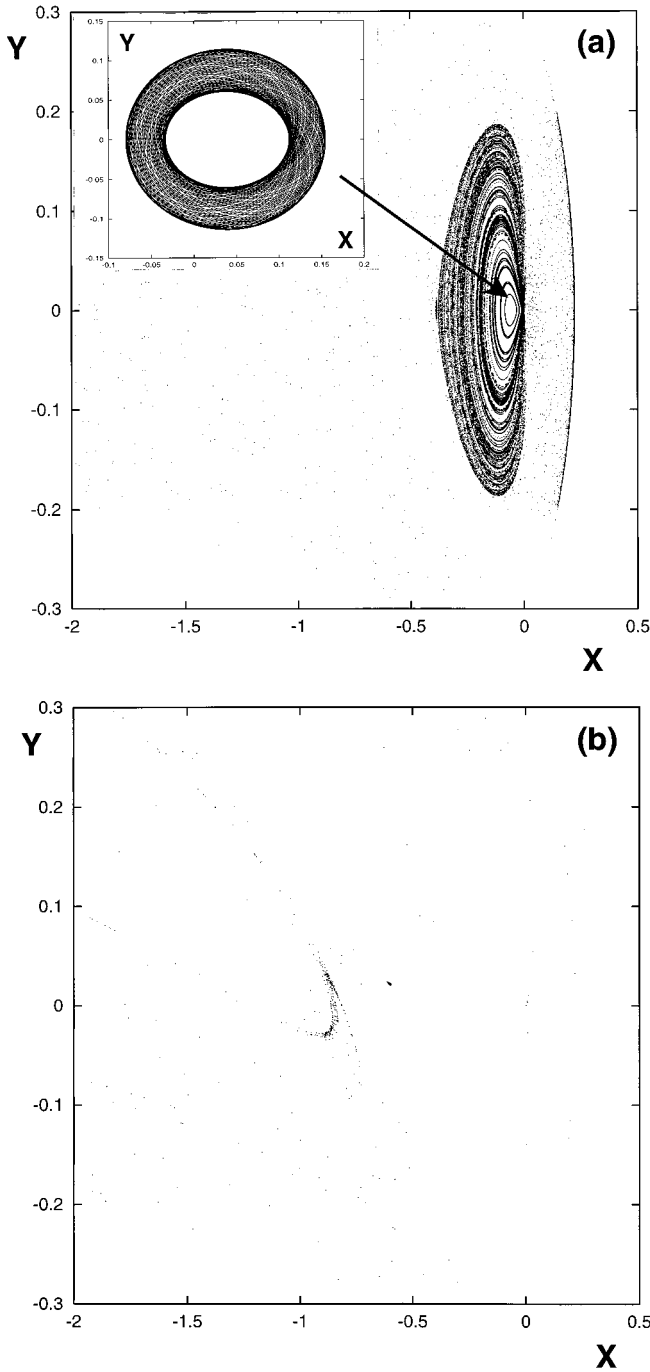


FIG. 8. (a) Combined Poincaré SOS for 200 randomly chosen initial conditions, calculated, as described in the text, at constant K , for $\omega=4$, $K_0 = -4.5$, $K=K_0+F^2/2\omega^2$, and $F=4.5$; (b) as (a) but for a set of initial conditions lying on an initially circular Kepler orbit taken from the ensemble used to compute Fig. 5(b). The SOS is computed in the rotating frame during the flat-top part of the pulse.

fixed K and computing their intersection with the plane $\dot{x}=0$. The Cartesian coordinate plane was then plotted. Because the coordinate plane is unbounded (orbits can go to infinity) the SOS is cut off close to the nucleus. The advantage of plotting the x - y SOS is that it provides information on where a particle is allowed to visit. The K value that the circular orbits are switched to is well *above* the saddle-point

value. We emphasize that the set of initial conditions used in generating Fig. 8(a) contains states of all eccentricities. The only orbits relevant to the hydrogen atom are those which originate from the initial ensemble of circular orbits and Fig. 8(b) shows their SOS.

The phase space for the parameters in Fig. 8(a) is divided between a highly regular region and a chaotic sea that contains relatively few points. The orbits confined to KAM curves close to the origin are very low angular-momentum states, i.e., highly eccentric, almost linear orbits (in the rotating frame initially linear orbits appear to rotate as shown in the inset. For details see [8]). It should be noted that the argument that collisions are necessary for ionization *does not* imply that collisions or close encounters with the nucleus are *sufficient* to cause ionization. For the present set of parameters only almost linear orbits are stable. Most of the points that pepper the rest of the SOS are prompt ionizers, and inhabit the region where the initially circular orbits of Fig. 5(b) are prepared. Because the SOS is computed in the x - y plane it is easy to see that orbits of radius $\rho=1$ are highly unstable and are, in fact, scattering orbits in most cases. Note that the point $x=-1$, $y=0$ is contained in the chaotic sea and this is roughly the region to which the initial ensemble is switched to by the pulse rise time. To verify this we compute in Fig. 8(b) SOS for a selection of trajectories selected from the initial ensemble. These orbits were switched as in Fig. 5(b) and at the top of the pulse their initial conditions were transformed into the rotating frame where they were integrated using the K value they were switched to by the pulse. Figure 8(b) should be compared to Fig. 8(a) and shows clearly where the initially circular orbits lie. Their ionization is obviously due to the details of the pulse rise time and that they do not experience hard collisions with the nucleus is irrelevant because they were switched, in most cases, directly into the ionization channel. Section III B considers the ionization of initially eccentric orbits.

B. Collisions with the nucleus as generators of chaos

In Ref. [10], for a different set of parameters, it was found quantum mechanically that orbits that are initially of medium ellipticity are the easiest to ionize as compared to linear or circular states. This led those authors to conclude that collisions are unimportant for ionization. For the parameters used in Fig. 8 of [10] we computed times of ionization for (a) initially circular orbits and (b) elliptical orbits for which initially $P_\phi=1/2$. During the time of integration it is significant that *none* of the initially circular orbits ionized. Figure 9 is a histogram for the elliptical orbits and in this case ionization occurs well into the flat-top part of the pulse. The integration was truncated during the flat top before all the orbits ionized. The main point is that ionization takes a relatively long time to occur for the elliptical orbits and does not occur *at all* for the circular orbits.

In fact, all the circular orbits were found to be switched directly into the resonance island centered roughly at $\{x=-1, y=0\}$ in the SOS shown in Fig. 10(a). This island is associated with the minimum of the effective adiabatic $V(\rho)$ potential lying exterior to the ZVS—compare to Fig. 4(c). Actually, as noted earlier, the island arises from an adiabatic decoupling of the fast angular frequency from the

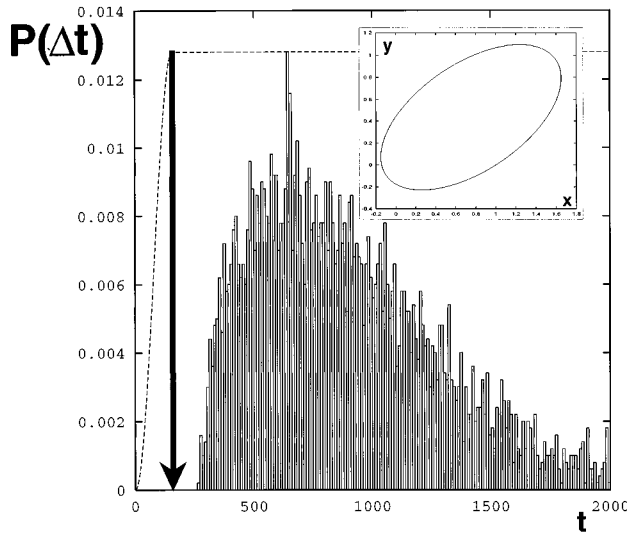


FIG. 9. As Fig. 5(b) except for $\omega=2.1$, $F=0.05$ and initial conditions chosen along an elliptical orbit (also shown) with initial eccentricity $e^2=3/4$ ($P_\phi=1/2$).

slow radial frequency of a circular orbit. The K value that the circular orbits are switched to, in this case $K \approx -2.6$, lies slightly *below* the saddle-point value of $K \approx -2.49$. The SOS (not shown) computed for orbits taken from the initial ensemble indicates that all of them end up rather tightly in the main resonance island [indicated by the top arrow in Fig. 10(a)]. This shows, again, that low eccentricity orbits are quite stable (see the large KAM curves close to the origin). In Fig. 10(a) there is a barrier (the white region) separating the chaotic sea from the nucleus. This is an example of a forbidden region and corresponds to the barrier in the ZVS (see Fig. 4). Howard [24] pointed out the existence of forbidden regions in this problem although their relevance was discounted in [10]. Similar forbidden regions are well known in the restricted three-body problem [9].

The insets in Fig. 10(a) show two typical orbits, a circular orbit from the 1:1 resonance island and an orbit from the chaotic sea. The circular orbit does not ionize while the chaotic orbit does: the chaotic orbit collides multiple times with the ZVS before ionizing and its distance of closest approach is defined by the ZVS. However, this orbit does not correspond to an orbit of the hydrogen atom. The only orbits in Fig. 10(a) that have any physical meaning (in the sense of originating from the initial hydrogenic ensemble) are those that lie in the 1:1 resonance island. These are the initially circular orbits that are switched to the value $K = -2.6$. Orbits in the chaotic sea for this value of K can be thought of as representing states already containing contributions from the hydrogenic continuum. We estimated, and have confirmed numerically, that orbits having an initial angular momentum $P_\phi \approx 0.9477$ will be switched during the pulse turn on precisely to the saddle-point K value. A SOS for these orbits is shown in Fig. 10(b). Note that these orbits now lie just *outside* the resonance island and these orbits can now cross the saddle point and collide with the nucleus prior to ionizing. The inset shows the radius of such an orbit with time and it is apparent, as we argued in [8], that this trajectory undergoes many close encounters with the nucleus prior to ioniza-

tion. The chaotic orbits shown in these figures obviously (a) undergo close encounters with the nucleus (compare their inner and outer turning points prior to ionization) and (b) ionize after being bounced from the core one last time.

Figure 10(c) is a histogram of ionization times, cut off at $t=2000$: note the very long ionization times compared to Fig. 5. This suggests many collisions prior to ionization as confirmed by the insets in Fig. 10(b). As one moves to more elliptical orbits the collisions with the nucleus become “harder” and fewer are needed for ionization to occur. Using a numerically determined Jacobi constant for an orbit initially having $P_\phi=1/2$ (i.e., for the orbits in Fig. 9) we computed the SOS in Fig. 10(d). In this figure $K = -1.54$ which is well above the saddle point. Again much of phase space is regular close to the origin, but there is a sizeable chaotic sea corresponding to elliptical orbits. Significantly, the resonance island at $\rho \approx 1$ in Fig. 8(a) has moved even farther from the nucleus. SOS for orbits coming *only* from the initial ensemble are shown in Fig. 10(e) together with a typical ionizing orbit. Again, note the multiple collisions with the nucleus prior to ionization.

It is now obvious why the states originating from an initially elliptical orbit are much easier to ionize than the circular orbits which are switched, in this example, into a regular part of phase space by the pulse. Thus there is a simple classical explanation for the results of the 2D quantum simulations of Ref. [10]: the circular orbits are switched into KAM regions, as are the initially linear orbits. The states of medium eccentricity, for these field values, are switched directly into the chaotic sea. The difference in propensity for ionization can be understood not so much in terms of whether the ionizing states are initially elliptical or circular because these concepts have no meaning when the CP field is actually “on.” What matters are (i) the Jacobi constant(s) achieved by the ensemble at the end of the pulse rise time, and (ii) where these orbits emerge in phase space. In the case of circular orbits each point along the orbit is switched into a KAM island and essentially the same Jacobi constant K , i.e., the distribution of K values is very tight as given by Eq. (18). For electrons moving on the initially elliptical orbit shown in Fig. 9 the final distribution of Jacobi constants is also quite tight but clustered around a larger (less negative value) that lies above the saddle point in the chaotic sea.

In Fig. 11 we show an orbit for different field parameters that is switched to slightly *below* the saddle point and exterior to the ZVS together with its SOS [Figs. 11(a) and 11(b)]: Fig. 11(c), on the other hand, shows the region of phase space into which essentially the entire ensemble emerges at the end of the pulse rise time. These orbits eventually ionize after a remarkable sequence of hard collisions with the ZVS exemplified by Fig. 11(a). Note especially the *huge* values the radius of the orbit achieves prior to its last collision with the ZVS. These orbits are relatively rare since a slight decrease in initial eccentricity leads to the ensemble being switched above the saddle point when direct collisions with the nucleus dominate. It is hard to reconcile the statements in [11] that collisions with the core are irrelevant with this figure.

Finally we arrive at the mechanism by which chaos is generated in this problem. Figure 11(d) is a composite that shows the behavior of the radius $r(t)$, the angular momen-

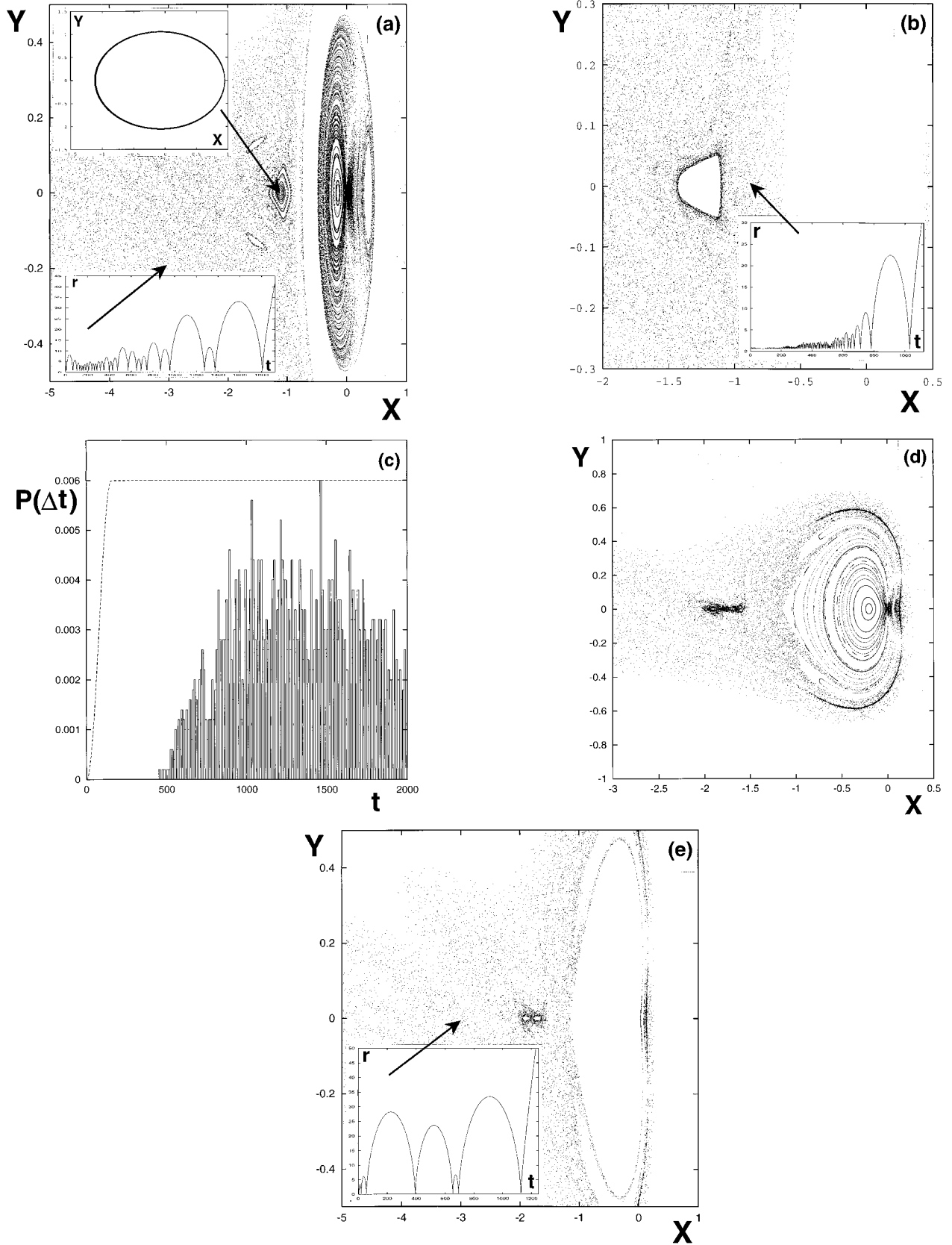


FIG. 10. (a) Combined SOS for 100 randomly chosen initial conditions, calculated as described in the text at constant K , for $\omega=2.1$, $K_0=-2.6$, $K=K_0+F^2/2\omega^2$, and $F=0.05$. The arrows relate typical orbits to the SOS. The top inset is an x - y plot of an orbit taken from the regular region, while the bottom inset is a radius (r) vs time (t) plot for an orbit taken in the chaotic region. (b) SOS for elliptical orbits with $P_\phi \approx 0.9477$ that are switched to the saddle-point value of K ; (c) histogram showing ionization times for the ensemble in (b) cutoff at $t=2000$; (d) composite SOS for $K=-1.54$; (e) SOS for elliptical orbits with $P_\phi=1/2$ that are switched to a value of $K=-1.54$ that is above the saddle-point value.

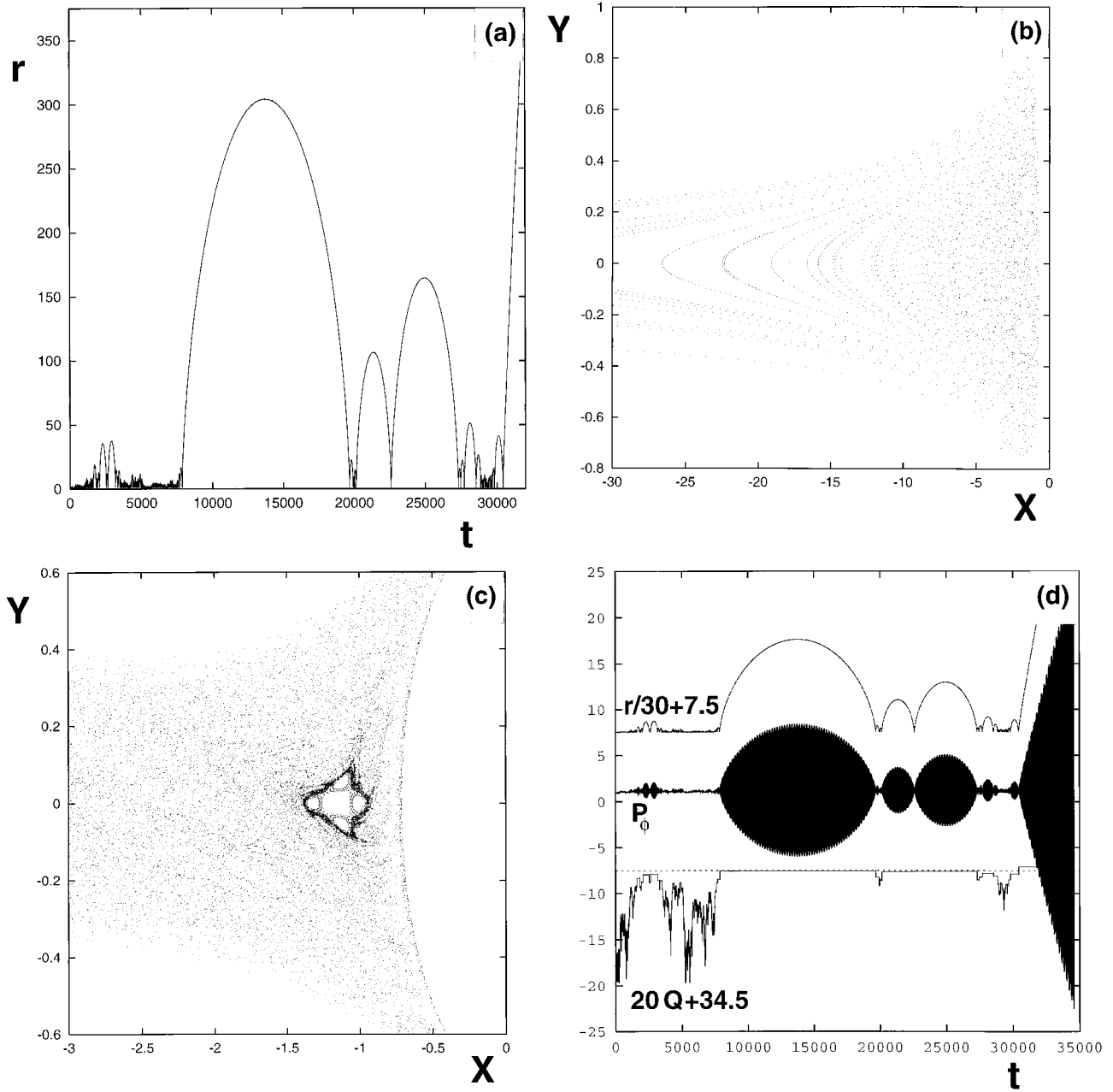


FIG. 11. (a) $r(t)$ for an orbit with initial $P_\phi = 0.98$, $K_0 = -2.55$, $\omega = 2.1$, and $F = 0.05$. The saddle-point energy is $K = -2.49$ for these parameters. Note the sequence of large excursions from the core followed by violent collisions with the ZVS. Its Poincaré SOS is shown in (b) while the SOS for an initial ensemble of 50 trajectories chosen with $P_\phi = 0.98$ is represented in (c). Note how different parts of the initial eccentric orbit are switched to different parts of phase space, as described in [8]. Frame (d) compares the behavior of the radius, $r(t)$, the angular momentum, $P_\phi(t)$, and $Q(t)$ for the orbit in (a). The quantities $r(t)$ and $Q(t)$ have each been scaled and then offset by a constant amount for clarity. The horizontal line shows the asymptotic ionization threshold for Q .

tum $P_\phi(t)$, and $Q(t)$ for the orbit in Fig. 11(a). During each collision the angular momentum is roughly conserved while, for large excursions from the nucleus, the angular momentum oscillates wildly (after ionization, going as $\rho \cos \theta$). On the other hand, the asymptotic quantity Q is best conserved (as expected) when the electron is far from the nucleus. Note how ionization occurs after Q has exceeded the asymptotic ionization threshold. Collisions with the core thus serve to “pump up” Q until ionization occurs. The correlation between $r(t)$, $P_\phi(t)$ and Q is remarkable.

Figure 12 is a SOS computed for field parameters for which a large chaotic sea is present. The parameters are cho-

sen to allow comparison with Ref. [15]: we use $\omega = 4$, $F = 0.01$, $K = -1.52$. At issue is the behavior of orbits started out in the chaotic sea. In Ref. [15] the point is made that orbits started out in the chaotic sea undergo repeated collisions with the nucleus and this may eventually lead to their ionization. Note that these simulations refer to a CW situation which may be relevant to the case of an ultrahigh molecular Rydberg state. Further the SOS in Fig. 12 relate to an ensemble of initial conditions with the same Jacobi constant. Figure 12 shows a typical orbit that repeatedly collides with the nucleus before ionizing—in this case there is no ZVS barrier to scatter from, i.e., the rotating frame energy is far

above the saddle-point energy. In fact the situation is quite reminiscent of chaotic scattering of atoms from a corrugated surface [32]. Note also the resonance islands apparent in the SOS: a typical orbit is also shown in Fig. 12 and it is clear that this orbit does *not* collide with the nucleus, hence its stability. To the extent that the CP problem is important in relation to issues of “quantum chaos” then to the same extent are collisions necessary to generate chaos in this problem. Any diffusive excitation mechanism [10,11], i.e., any mechanism that assumes diffusive transport through phase space, must, therefore, involve collisions. Elsewhere we will provide a more thorough account of chaotic scattering in this system [33] and simulations for 2D and 3D ionization.

IV. CONCLUSIONS

We have examined the question of how hydrogen atoms in circular polarized microwave fields become ionized using classical mechanical methods and simulations. Most of our simulations were for the case of initially circular orbits in a 2D model of the atom, i.e., motion confined to the plane of polarization. Kapitza’s method was used to estimate the value of the Jacobi constant K induced by the shape and time of the pulse rise time and this allowed us to explain the strong pulse dependence of ionization probabilities for circular and low eccentricity states.

The complexity of the ionization of hydrogen atoms in CP microwaves can be ascribed to the fact that orbits of different eccentricity in the original ensemble are not only switched by the pulse rise time into different regions of phase space, but also, effectively, to different energies in a rotating frame. Further, the phase space of the CP problem is obviously very different from the unperturbed hydrogen atom and depends strongly on the particular CP field strength and frequency used. Perturbative arguments such as “an orbit is circular in the hydrogen atom, is of high angular momentum and, therefore, cannot collide with the nucleus,” simply do not work if angular momentum is destroyed as a constant of the motion. The CP problem turns out to be tunable between two integrable limits and the transition can proceed smoothly or through transitions to chaos, depending on the particulars of the field used [26]. The sensitive way that the system depends on the pulse shape, the field parameters and the orbits in the initial ensemble essentially rules out simplistic, all-embracing explanations. Ionization depends on (a) the value the Jacobi constant achieves at the end of the rise time and (b) the part of phase space that the ensemble finds itself switched to by the pulse. The regularity or chaoticity of phase space itself depends on the Jacobi constant and the particular field parameters.

Arguments that collisions are unnecessary for ionization in the CP problem were also examined in some detail: we conclude that, unless the electron is ionized during the rise time of the pulse, ionization can proceed only through collisions. The “hardness” of the collision and distance-of-closest approach obviously depend on how close to the asymptotic ionization threshold the electron is at the start of the flat top. Since, for the field parameters and pulses used in some previous simulations [10,11], we demonstrated that

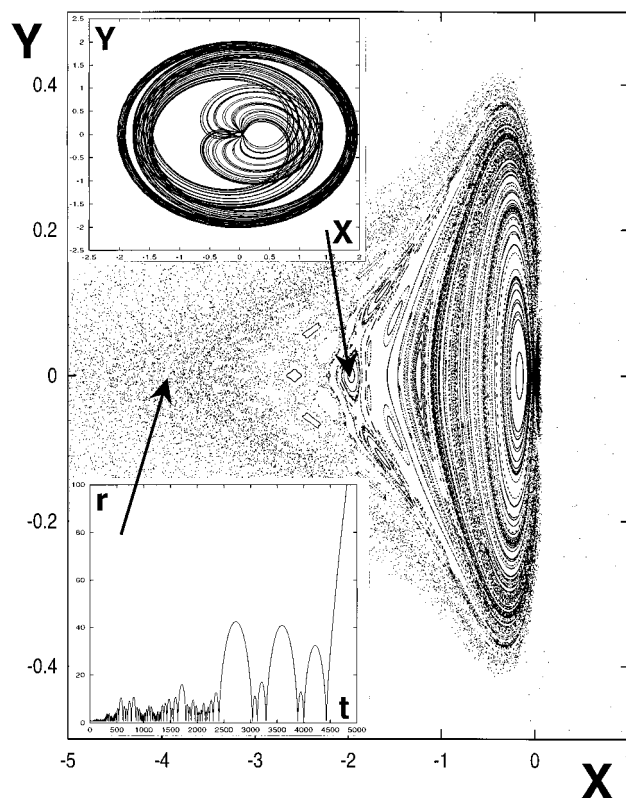


FIG. 12. Combined Poincaré SOS for 200 randomly chosen initial conditions, calculated as described in the text at constant $K = -1.52$, for $\omega = 4$, and $F = 0.01$.

most of the atoms are already ionized before the flat top, it is an oversimplification to conclude that circular orbits, in general, ionize without colliding with the nucleus. This conclusion is misleading and provides little insight into the nonlinear dynamics of how a Rydberg electron interacting with a CP microwave field finds its way into the exit channel (unless the pulse rise time places it there from the start). Weakly bound electrons need only experience soft collisions with the nucleus. Importantly, such weakly bound electrons live at the very periphery of a chaotic sea and are too fragile to undergo many such collisions without ionizing. As one penetrates farther into the chaotic sea, actually, one is sampling orbits that are more tightly bound and, therefore, must collide more often and more frequently with the nucleus if they are to ionize. In reality, the chaotic sea exists precisely because of a chaotic sequence of collisions between the electron and either a barrier in the ZVS or the nucleus. At some point the electron is simply knocked out of the atom by scattering from the core, and, in most cases by direct collisions with the nucleus itself.

ACKNOWLEDGMENTS

We are grateful to Professor Charles Jaffé and Ernestine Lee for valuable discussions and comments. Partial support of this research by the National Science Foundation and the American Chemical Society (donors of the Petroleum Research Fund) is gratefully acknowledged.

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