Stochastic ionization of surface-state electrons: Classical theory

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In recent experimental investigations of the ionization of Rydberg atoms in low-frequency electromagnetic fields, ionization rates have been measured which depend strongly on the intensity of the oscillating fields but only weakly on the frequency. In an attempt to understand this ionization mechanism, an analogous, one-dimensional system is considered consisting of a surface-state electron bound to the surface of liquid helium by its image charge. A complete classical analysis of the behavior of this nonlinear oscillator in a microwave field is presented which shows that above a critical field strength the electron dynamics become chaotic and the electron diffuses in energy until it ionizes. Analytic estimates for the classical thresholds and rates for stochastic excitation and ionization are determined as functions of the microwave-field amplitude and frequency. Since the microwave frequencies and field strengths required for stochastic ionization of this one-dimensional "hydrogen" atom are readily available, this system provides an opportunity to thoroughly explore the manifestations of classical chaos in a quantum system.

I. INTRODUCTION

Recently, deterministic classical systems with chaotic dynamics have been the subject of extensive research; however, little progress has been made on the question of whether the chaotic dynamics persist in a quantum-mechanical description of these systems. This question is important in a variety of problems, such as the calculation of the vibrational and rotational spectra of polyatomic molecules and the determination of the response of atoms and molecules to time-dependent electromagnetic fields.[1] Both the n-body problems and the driven oscillators correspond to nonintegrable, classical systems which exhibit chaotic behavior. Investigations of the quantum dynamics of these nonintegrable systems have significant applications in chemistry, laser isotope separation, and the development of short wavelength lasers. Experimental and theoretical studies of the time-dependent systems are especially important for an understanding of the variety of novel features exhibited by atomic and molecular physics in large-amplitude laser and microwave fields.

To date, the attempts to develop a quantum description of these systems remain incomplete and controversial.[2] These problems first arose in the early development of quantum mechanics. In 1917 Einstein[3] remarked that the semiclassical quantization procedure fails for nonintegrable classical systems such as the three-body problem. More recently, numerical investigations[4] of nonintegrable systems which exhibit chaotic behavior in the classical limit suggest that the quantum dynamics may also be stochastic. However, theoretical studies of some time-dependent Hamiltonians with discrete quasienergy spectra[5] indicate that the quantum dynamics of such systems are always quasiperiodic and never chaotic.

The question of whether a quantum system can be chaotic must, ultimately, be resolved by experimental investigations of real physical systems which are amenable to theoretical analysis. In the last ten years some very suggestive experiments on the microwave ionization of highly excited hydrogen atoms have been performed by Bayfield and Koch and their co-workers.[6–8] These observations of excitation and ionization rates which depend strongly on the *intensity* of the microwave fields and only weakly on the frequency provide strong evidence for stochastic behavior in a quantum system.

In their pioneering work Bayfield and Koch[6] measured the ionization rate of a beam of neutral hydrogen atoms, carefully prepared in highly excited Rydberg states (principal quantum number $n \sim 66$), as they passed through a microwave cavity. Although the microwave frequency (9.9 GHz) was only $\sim 40\%$ of the resonant frequency for single-photon excitation to the $n=67$ level and $\sim 1\%$ of the photon frequency for excitation to the continuum, they observed significant ionization above a critical microwave field of ~ 20 V/cm.

Thus far conventional quantum theory has been unable to account for these experimental observations. Since the microwave frequency is less than the classical orbital frequency of the electron, Stark ionization at the peak electric field is a prime candidate for the ionization mechanism. (The interaction with the magnetic field is much weaker and can be neglected.) However, estimates of the ionization due to tunneling in a static field, based on the quantum-mechanical calculations of Damburg and Kolosov[9] and the experimental measurements of Koch and Mariani,[10] give rates which are too slow to explain the experimental results. The microwave field required for significant tunneling during the beam transit time through the cavity ($\tau_{\rm tr} \sim 10^{-7}$ sec) is a factor of 2 larger than the observed critical field.[11] Another likely explanation of the ionization at low-field strengths is multiphoton absorption. However, since nearly 100 photons must be absorbed to reach the continuum, the theoretical investigation of this mechanism has been intractable, requiring 100 orders of perturbation theory and complicated by the effects of intermediate resonances.

The only successful theoretical analysis involved a classical treatment of the orbiting electron in a plane polarized, oscillating electric field. This approximation is justified by the large quantum numbers, $n \sim 66$, in the experiment. Leopold and Percival[12] integrated the classical equations of motion for this nonlinear oscillator in a time-dependent field and demonstrated the possibility of classical excitation and ionization of the electron for peak electric field strengths below those required for significant quantal tunneling. Furthermore, using a Monte Carlo model of Bayfield and Koch's experiment, they were able to calculate ionization rates which agreed well with the experimental measurements.[12]

The physical ionization mechanism evoked by this classical calculation for a driven, nonlinear oscillator is stochastic diffusion of the electrons in phase space. Ionization results when the chaotic trajectories wander into the continuum. Since the Kolmogorov-Arnol'd-Moser theorem[13] guarantees that for small microwave fields most of the trajectories will remain regular, the microwave fields must exceed a critical level before the orbits become sufficiently chaotic to cause significant ionization. Unfortunately, because the electrons move around in a six-dimensional phase space, detailed numerical investigations of the transition to global stochasticity are extremely time consuming. Moreover, analytical attempts to describe the transition to global stochasticity[14–16] due to resonance overlap[17] in the classical phase space of the electron have proven difficult and remain incomplete.

In an attempt to understand this stochastic ionization mechanism a simpler, one-dimensional quantum system was proposed[18] for theoretical and experimental study consisting of surface-state electrons (SSE) which are weakly bound to the surface of liquid helium by their image charge.[19] Spectroscopic studies of the unperturbed SSE by Grimes *et al.*[19] and by Lambert and Richards[20] found that the energy levels are given by the hydrogenic formula

$$E_n = -Z^2\mathscr{R}/n^2, \quad n = 1,2,3, \ldots , \tag{1}$$

where $\mathscr{R} = 13.6$ eV. These energy levels correspond to a one-dimensional, quantum-mechanical description of the SSE which assumes an attracting Ze^2/x potential due to the image charge and a repulsive barrier at the surface due to Pauli exclusion. In the classical limit the electron bounces back and forth between the helium surface and the one-dimensional Coulomb potential.

The proposed experiment consists of an investigation of the dynamics of this one-dimensional "hydrogen" atom in the presence of an oscillating microwave field with polarization normal to the helium surface. This driven, nonlinear oscillator provides an ideal system for the theoretical and experimental study of quantum chaos. First, the classical, semiclassical, and quantum analyses of the behavior of this one-dimensional system are much more tractable than those for the full three-dimensional hydrogen atom. Second, since liquid helium is a poor dielectric, the image charge is very small, $Z \sim 7.1 \times 10^{-3}$, and the binding energies and characteristic frequencies of the SSE are four orders of magnitude smaller than those for real hydrogen atoms. Therefore, available microwave sources can be used to investigate the dynamics of the perturbed SSE in both the classical ($n \gg 1$) and quantum ($n \sim 1$) limits. Consequently, this nonintegrable dynamical system provides a unique opportunity to test the predictions of the classical, semiclassical, and quantum theories with a real experiment which should resolve much of the controversy surrounding the problem of quantum chaos.

The goal of this paper is to present a complete classical analysis of the dynamics of the SSE in a microwave field. This classical treatment, which is based on the resonance overlap criterion for the onset of a global stochasticity, shows that ionization results from stochastic diffusion of chaotic electron orbits and provides analytic estimates for the microwave-field threshold for stochastic ionization as well as an ionization rate.[18] These results should be valid for large quantum numbers, $n \gg 1$, corresponding to the classical limit. In addition, this calculation predicts the microwave powers and frequencies required for experimental investigations of the manifestations of classical chaos n the quantum limit ($n \sim 1$). A detailed description of the electron dynamics in the quantum regime, which requires the solution of a one-dimensional Schrödinger equation with time-dependent potential, will be pursued in future work. Although this partial differential equation with variable coefficients does not appear to be amenable to analytic solution, it can be studied, numerically, using efficient algorithms for the solution of partial differential equations. Some preliminary numerical results for bound electrons in one and two dimensions[21] indicate that the quantum-mechanical description of these systems also exhibits ionizing solutions.

In Sec. II the resonance overlap criterion is used to calculate the critical-field strength for the onset of global stochasticity. If the microwave frequency is greater than or comparable to the classical bounce frequency of the SSE, then the resonance overlap criterion gives a direct estimate for the critical field required for stochastic excitation and ionization. However, if the driving frequency is much less than the frequency of the nonlinear oscillator, then an additional criterion must be satisfied. Analytical and numerical calculations of the critical field for stochastic ionization in this limit of quasistatic perturbations are presented in Sec. III. Since most of the experiments with real hydrogen atoms[6–8] have been performed in this regime, these new results have direct bearing on the theoretical interpretation of these experiments.

Once the electron motion becomes chaotic, the electrons diffuse in energy until they reach the continuum. A statistical, random walk treatment of the electron dynamics can then be used to estimate the stochastic diffusion rates. In Sec. IV quasilinear calculations of the stochastic excitation and ionization rates are presented and compared with the results of numerical integrations of the classical equations of motion for this perturbed, nonlinear oscillator. Finally, Sec. V concludes with a discussion of the feasability of the proposed experiment and the possibility of extending the nonlinear classical analysis for the perturbed SSE to the problem of microwave ionization of three-dimensional Rydberg atoms.

II. RESONANCE OVERLAP CRITERION FOR THE PERTURBED SSE

Zaslavskii and Chirikov[17] have developed a simple, approximate description of the effects of a periodic perturbation on one-dimensional, nonlinear oscillators. For sufficiently large perturbations these nonintegrable dynamical systems exhibit a stochastic instability leading to chaotic motion. By examining the trajectories in the action-angle variables of the unperturbed oscillator, a resonance overlap criterion can be used to estimate the critical perturbation strength for the onset of global stochasticity. This prescription is easily applied to the classical description of the SSE in an oscillating electric field.

A. Action-angle variables for the unperturbed Hamiltonian

First, we consider the integrable dynamics of a classical electron in a one-dimensional $1/x$ potential with a repulsive barrier at the origin. The equations of motion in atomic units[22] are generated by the Hamiltonian

$$H_0(x,p,t)=p^2/2+\begin{cases}-Z/x, & x>0\\ \infty, & x\le 0\end{cases}. \tag{2}$$

(Note that in the dimensionless units used in Ref. 18 $Z=\frac{1}{8}$.) Then Hamilton's equations for this nonlinear oscillator are easily integrated to show that a bound electron with energy $-E$ bounces back and forth in the potential well between $x=0$ and $x=a\equiv Z/E$ (a.u.) with frequency $\Omega_0=(8Z/a^3)^{1/2}$ a.u.

Before considering the effects of the perturbation it is convenient to make a canonical transformation to the action-angle variables

$$I=\sqrt{Za/2}\ , \tag{3}$$

$$\Theta=\begin{cases}2[\sin^{-1}(\sqrt{x/a})-\sqrt{(x/a)(1-x/a)}], & p\ge 0\\ 2\pi-2[\sin^{-1}(\sqrt{x/a})-\sqrt{(x/a)(1-x/a)}], & p<0\end{cases} \tag{4}$$

which reduce the unperturbed dynamics to straight-line trajectories in action-angle space. The action, I, is simply defined to be the constant area enclosed by a complete phase-space orbit, $(1/2\pi)\int p\,dx$, and the angle Θ is the position variable canonically conjugate to I. The new, unperturbed Hamiltonian is easily derived from Eq. (4) and the definition of a,

$$H_0(I)=-Z^2/2I^2\ , \tag{5}$$

which gives a constant angular velocity

$$\Omega_0(I)=dH_0/dI=Z^2/I^3\ . \tag{6}$$

The quantum-mechanical energy levels for the unperturbed oscillator can be determined using the Bohr-Sommerfeld quantization rule. In atomic units this implies that the discrete energy levels correspond to integer values of the action I. The characteristic scale lengths, frequencies, and electric field strengths for the unperturbed SSE's with $Z\simeq 7.1\times 10^{-3}$ can then be estimated by considering the classical description of the SSE with energies

$$-E_0=-Z^2/2I_0^2 \text{ a.u.}$$
$$\simeq -6.8\times 10^{-4}/I_0^2 \text{ eV for } I_0=1,2,3,\ldots . \tag{7}$$

The maximum excursion and oscillation frequency of the corresponding classical electron are

$$a_0=2I_0^2/Z \text{ a.u.}\simeq 1.5\times 10^{-6}I_0^2 \text{ cm}\ , \tag{8}$$

$$\nu_0=\Omega_0/2\pi=(1/2\pi)Z^2/I_0^3 \text{ a.u.}\simeq 330/I_0^3 \text{ GHz}\ , \tag{9}$$

and the binding electric field at maximum excursion is

$$F_0=(\tfrac{1}{4})Z^3/I_0^4 \text{ a.u.}\simeq 450/I_0^4 \text{ V/cm}\ . \tag{10}$$

B. Effects of perturbing microwave fields

Since the standing wavelengths of the microwave radiation are long compared with the maximum excursion, a_0, of the SSE from the liquid-helium surface, the spatial variation of the perturbing electric fields can be neglected. In addition, for nonrelativistic electron velocities the interaction with the oscillating magnetic field is also negligible. We therefore consider a perturbation of the form

$$V(x,t)=xF\cos(\Omega t)\ , \tag{11}$$

where F and Ω are the electric field amplitude and angular frequency of the externally applied microwave fields.

For sufficiently small electric fields the Kolmogorov-Arnol'd-Moser (KAM) theorem[13] guarantees that most of the straight-line trajectories in action-angle space will be only slightly distorted by the perturbation. If we expand the perturbation in a Fourer series[17] in Θ, the perturbed Hamiltonian can be written as

$$H(\Theta,I,t)=H_0(I)+F\sum_{m=-\infty}^{\infty}V_m(I)\cos(m\Theta-\Omega t)\ , \tag{12}$$

where the Fourier amplitudes of the perturbation are defined by the integrals

$$V_m(I)=\frac{1}{2\pi}\int_0^{2\pi}d\Theta\, e^{im\Theta}x(\Theta,I)\ . \tag{13}$$

The maximum distortion of the orbits in action-angle space will occur at resonances where the phase, $m\Theta-\Omega t$, is stationary.[17] The resonant frequencies and actions are therefore determined by the relation[17]

$$m\Omega_0(I)-\Omega=0\ . \tag{14}$$

Then using Eqs. (6) and (14), the action resonant with the mth subharmonic of the perturbation is

$$I_m=(mZ^2/\Omega)^{1/3}\ . \tag{15}$$

For small perturbations the Hamiltonian can be approximated in the vicinity of each resonance by the Hamiltonian of a pendulum; and the electron trajectories near the resonances are confined in narrow island chains in action-angle space. Between the resonances the surviving Komolgorov-Arnol'd-Moser (KAM) surfaces prevent the orbits from wandering from one resonance to another. The electrons gain and lose energy as they ride the perturbation but no net change in the energy occurs. The per-

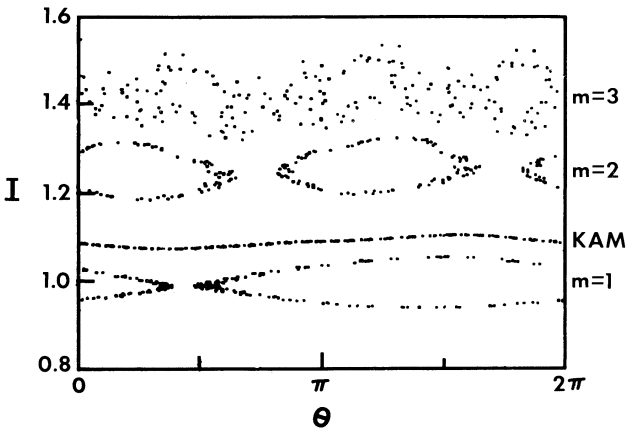

FIG. 1. Island chains for the $m=1,2,3$ resonances for a perturbation with $\Omega=Z^2$ a.u. and $F=0.0075Z^3$ a.u. The $m=3$ islands already exhibit large stochastic regions. Also shown is a confining KAM surface between the $m=1$ and $m=2$ resonances.

turbed trajectories in action-angle space can be graphically displayed by numerically integrating the equations of motion for x and p, then transforming to the action-angle variables, I and Θ, and plotting the location of the orbit at integer multiples of the perturbation period. The island chains corresponding to the three lowest resonances for a periodic perturbation of the form $V(x,t)=xF\sin(\Omega t)$ with frequency $\Omega=Z^2$ a.u.$\simeq$330 GHz and amplitude $F=0.0075Z^3$ a.u.$\simeq$13.5 V/cm are shown in Fig. 1. The island chains corresponding to the subharmonic resonances with a perturbation of the form of Eq. (11) are simply shifted in angle by $\pi/2$. Figure 1 also shows a typical KAM surface between the $m=1$ and $m=2$ islands. For weak perturbations these smooth curves fill the regions between the island chains and below the $m=1$ island confining the electron trajectories to narrow bands in action and energy.

As the perturbation increases, the islands grow wider in action. When the islands are sufficiently large the electron can diffuse in action (or energy) by wandering from one island chain to another. These transitions occur when the orbit of the electron is so distorted by one resonance that its oscillation frequency becomes resonant with another resonance corresponding to a subharmonic of the oscillating microwave field. In Fig. 1 the $m=3$ island chain is already beginning to break up due to its interaction with the $m=4$ resonance. This qualitative picture provides a means of estimating the size of the perturbation required to make the transition from regular to stochastic behavior. Roughly speaking, this occurs when the islands generated at the resonances overlap.

C. Resonance overlap criterion

The widths of the islands are determined by the corresponding Fourier amplitudes, V_m, of the perturbation. Following Ref. 17, we estimate the island widths by approximating the Hamiltonian in the vicinity of the resonance by the Hamiltonian for a pendulum. Near the mth resonance we need only consider the mth Fourier component of the perturbation. Then making a canonical transformation to the new variables $\Delta=I-I_m$ and $\xi=\Theta-(\Omega/m)t$ using the time-dependent generating function[23] $F_2(\Delta,\Theta,t)=(I_m+\Delta)[\Theta-(\Omega/m)t]$ and Taylor expanding the new, approximate Hamiltonian in small Δ and F, we get

$$K(\xi,\Delta)\simeq-(\tfrac{1}{2})\Omega_0'(I_m)\Delta^2 + FV_m(I_m)\cos(m\xi)+\text{const}\ , \tag{16}$$

where

$$\Omega_0'\,|_{I=I_m}=d\Omega_0/dI\,|_{I=I_m}=3Z^2/I_m^4\ . \tag{17}$$

Except for an irrelevant constant and a difference in sign, K is just the Hamiltonian of a simple pendulum; and the island width in action-angle space corresponds to the width of the trapping (libration) region[24] of the pendulum

$$W_m=4(FV_m/\Omega_0')^{1/2}\,|_{I=I_m}\ . \tag{18}$$

The Fourier components, V_m, of the oscillating microwave potential can be determined analytically by evaluating the integrals in Eq. (13) using a trick suggested by Landau and Lifschitz[25] for the calculation of the Fourier components of the dipole moment for a system of two charged particles,

$$V_m(I)=\frac{1}{2\pi}\int_0^{2\pi}d\Theta\, e^{im\Theta}x(\Theta,I)\equiv J_m'(m)I^2/Zm \sim 0.411I^2/(Zm^{5/3}) \tag{19}$$

for large m, where J_m' is the derivative of the ordinary Bessel function of order m. In Refs. 11 and 18 these integrals were evaluated numerically and were found to scale approximately as $m^{-3/2}$. However, a proper asymptotic analysis of the derivative of the Bessel function gives a $m^{-5/3}$ dependence as shown in Eq. (19).[26]

Combining Eqs. (15) and (17)–(19), the width of the mth resonance is

$$W_m=4I_m^3[FJ_m'(m)/3mZ^3]^{1/2} \tag{20}$$

and using Eq. (15), the separation of the m and $m+1$ resonances is

$$\delta_m=I_{m+1}-I_m=(Z^2/\Omega)^{1/3}[(m+1)^{1/3}-m^{1/3}] \sim I_m/3m \tag{21}$$

for large m. Then the zeroth-order islands overlap when the ratio of the island width to the separation is greater than one,

$$1<0.5(W_{m+1}+W_m)/\delta_m\sim 4.5F^{1/2}m^{5/6}/(Z^{1/6}\Omega^{2/3}) \tag{22}$$

for large m. This inequality provides an approximate criterion for the critical microwave field required to destroy the KAM surfaces between the m and $m+1$ island chains. Then the electron can wander from one classical resonance to the other. This island overlap criterion for the onset of global stochasticity requires that

$$F>F_c\sim 0.05Z^{1/3}\Omega^{4/3}/m^{5/3} \tag{23}$$

in atomic units. Finally, using Eqs. (10) and (15) the critical field can be expressed in terms of the initial action I_0, and Coulomb binding field, F_0, for the unperturbed SSE as

$$F_c \simeq 0.05 m^{-1/3} Z^3 / I_0^4 \simeq 0.2 m^{-1/3} F_0 \ . \tag{24}$$

This estimate for the critical field assumes that m is large. A more precise calculation of the island widths, Eq. (20), and separation, Eq. (21), for the $m=1$ and 2 resonances gives a result which is $\sim 40\%$ lower.

If the SSE has an initial energy corresponding to an action which lies between I_m and I_{m+1}, then the application of microwave fields in excess of the threshold defined by Eq. (24) will cause the electron to diffuse in action. Moreover, since the island overlap, Eq. (22), increases with m, once the microwave field exceeds the threshold for stochastic diffusion for electrons with action I_m, then the confining KAM surfaces will also be destroyed for larger actions. Since the stochastic region in phase space is bounded below and unbounded above the electrons will tend to diffuse to larger actions (or energies) until they ionize.

The resonance overlap criterion has been the subject of extensive numerical and analytical investigations[24,27,28] which indicate that it provides a good estimate (within a factor of ~ 2) of the perturbation strength required for global stochasticity. In actual fact secondary island chains are generated by the nonlinear interaction of the primary islands which accelerate the destruction of the confining KAM surfaces. Consequently, this estimate provides an upper bound on the critical-field strength for the onset of stochastic excitation and ionization of the electron.

III. QUASISTATIC IONIZATION CRITERION

If the frequencey of the perturbation is much less than the oscillation frequency of the nonlinear oscillator or, equivalently, if the electron has an initial action, I_0, which is smaller than $I_1=(Z^2/\Omega)^{1/3}$, then the overlap of primary resonances, $m=1,2,3,\ldots$, is not a sufficient condition for the onset of stochastic diffusion since some confining KAM surfaces always remain below the $m=1$ classical resonance. If $I_0 < I_1$, then the primary island in action-angle space, centered at I_1, must expand until it reaches I_0 in addition to overlapping the $m=2$ island chain before ionization can occur.

This limit of quasistatic perturbations is difficult to analyze because the microwave field required to ionize the electrons is usually large enough to cause strong interaction of a number of resonances and the simple, two-resonance perturbation theory fails. However, two cases are analytically tractable. First, for very low-frequency perturbations the classical adiabatic theory can be used to calculate the critical perturbation strength for static-field ionization.[29,30] Second, for perturbations nearly resonant with the $m=1$ classical resonance, the critical field can be determined from a calculation of the width of the $m=1$ island alone. In the intermediate regime the other Fourier components of the perturbation play an important role in generating secondary island chains below the $m=1$ primary island which extend the stochastic region in action-angle space to lower actions. In this case we resort to numerical integrations of the classical equations of motion for the perturbed oscillator to connect the adiabatic and near-resonance regimes.

A. Adiabatic theory, $\Omega \ll \Omega_0$

The potential energy of the combined Coulomb and microwave electric fields,

$$V(x,t) = -Z/x + xF\cos(\Omega t) \ , \tag{25}$$

is shown in Fig. 2 for $\Omega t = 2k\pi$, $(k+1/2)\pi$, and $(2k+1)\pi$ for $k=0,1,2,\ldots$. In the first case the electric field is directed away from the surface and the electron is pulled toward the surface by both fields, in the second the perturbing field vanishes, and in the third the field is pointed toward the surface and the two fields pull in opposite directions which lowers the top of the potential barrier. In a static or slowly varying electric field, $\Omega \ll \Omega_0$, this lowering of the potential permits bound electrons with sufficiently large energies or actions to escape from the surface. The height, V_t, and position, x_t, of the top of the potential are easily determined from Eq. (25),

$$V_t = -2(FZ)^{1/2} \ , \tag{26}$$

$$x_t = (Z/F)^{1/2} \ . \tag{27}$$

If the electric field is turned on slowly, the classical adiabatic theory can be used to calculate the critical-electric-field strength to dissociate an electron with initial action I_0 and energy $-E_0 = -Z^2/2I_0^2$. The analogous classical calculation of the critical fields for Stark ionization of hydrogen atoms has been carried out by Banks and Leopold.[29,30]

If the perturbation is sufficiently slow, then the true action of the perturbed oscillator will remain an adiabatic invariant even though the electron energy, E, decreases as the electric field becomes more negative. This invariant action, J, is defined by

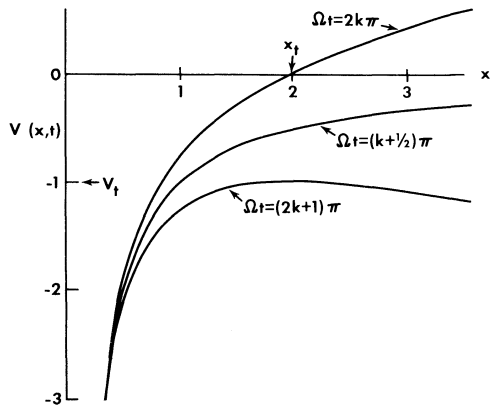

FIG. 2. The time-dependent potential for the SSE in an oscillating electric field at times corresponding to the maximum and minimum microwave fields is compared with the unperturbed Coulomb potential.

$$J=(\sqrt{2}/\pi)\int_0^{x_-}(Z/x+xF-E)^{1/2}dx\ , \tag{28}$$

where x_- is the classical turning point,

$$x_-=[E-(E^2-4FZ)^{1/2}]/2F\ , \tag{29}$$

which is one of the two roots of the integrand in Eq. (28). When $F=0$, Eq. (28) reduces to the definition of the action for the unperturbed oscillator. Since J is assumed to be an adiabatic invariant, it is equal to the initial action I_0 of the unperturbed SSE.

As the amplitude of the electric field is increased the classical turning point for an electron with action J increases until $x_-=x_t=(Z/F_c)^{1/2}$. For larger fields the electron escapes over the barrier. Since the two roots of the integrand in Eq. (28) coalesce at x_t, the critical field, F_c, for classical dissociation can be determined as a function of J by evaluating the integral

$$J=(\sqrt{2}F_c/\pi)\int_0^{x_t}(x_t-x)/\sqrt{x}\ dx$$
$$=(4\sqrt{2}/3\pi)Z^{3/4}/F_c^{1/4}\ . \tag{30}$$

This exact calculation gives a critical electric field strength for Stark ionization of the SSE of

$$F_c=(4\sqrt{2}/3\pi)^4Z^3/I_0^4$$
$$\simeq 0.130Z^3/I_0^4=0.52F_0\ , \tag{31}$$

expressed in terms of the initial action, $I_0=J$, and the binding Coulomb field, F_0, of the unperturbed oscillator. This threshold for classical ionization in adiabatically varying electric fields, $\Omega \ll \Omega_0$, is considerably larger than the critical field for island overlap predicted by Eq. (24).

B. Near-resonance theory, $\Omega \sim \Omega_0$

If the initial frequency of the nonlinear oscillator is approximately resonant with the microwave frequency, $\Omega \sim \Omega_0$, then for small perturbations the electron will be trapped in the resonance and the electron dynamics will lie on one of the closed, librating trajectories of the $m=1$ island. Figure 1 shows one of these trapped orbits centered at $I=1$. Below the separatrix of the $m=1$ island the electrons with $\Omega_0>\Omega$ correspond to untrapped (passing) particles. As the perturbation increases, the islands grow wider in action and the $m=1$ Fourier component of the microwave perturbation interacts more strongly with the other components resulting in the breakup of the island chains. The confining KAM surfaces between the resonances are destroyed and the librating and passing orbits mix chaotically. However, since KAM surfaces persist below the separatrix of the $m=1$ island, the near-resonance, passing trajectories may remain confined despite the onset of stochastic diffusion for electrons with larger actions and energies.

The critical-field strength required to stochastically excite and ionize these near-resonance electrons can be estimated by computing the perturbation strength required for the $m=1$ island centered at I_1 to trap an electron with action, $I_0<I_1$. As a first approximation we neglect the effects of the Fourier components of the perturbation with $m\neq 1$. Then the exact Hamiltonian in action-angle space, Eq. (12), reduces to

$$H_1(\Theta,I,t)=-Z^2/2I^2+FV_1(I)\cos(\Theta-\Omega t) \tag{32}$$

where

$$V_1(I)=J_1'(1)I^2/Z\simeq 0.325\ . \tag{33}$$

In addition we make a canonical transformation to a frame moving with the angular velocity Ω using the time-dependent generating function $F_2(I,\Theta,t)=I(\Theta-\Omega t)$. This transformation to the new angle variable, $\xi=\Theta-\Omega t$, defines a new time-independent Hamiltonian

$$K_1(\xi,I)=-Z^2/2I^2+\epsilon I^2\cos\xi-\Omega I\ , \tag{34}$$

where $\epsilon=0.325\ F/Z$. K_1 describes an integrable dynamical system with closed (librating) orbits for resonant actions and untrapped (passing) trajectories away from the resonance at I_1. Figure 3 shows both trapped and passing orbits which were calculated by numerically integrating the equations of motion generated by K_1.

As the perturbation is increased the width of the trapping region, defined by the location of the separatrix, grows wider in action. Then the field, F_t, required to trap an electron with action I_0 in the $m=1$ island can be estimated by determining the field strength required for the separatrix to reach down to I_0. Since K_1 is independent of time, the trajectories of the librating and passing particles lie on a family of curves defined by the equation

$$K_1(\xi,I)=k\ , \tag{35}$$

where each orbit is labeled by the continuous parameter k. The separatrix trajectory which separates the trapped and untrapped orbits corresponds to a particular value of $k=k_s$ which is a function of the perturbation frequency, Ω, and strength, F. Once k_s is determined, then Eq. (35)

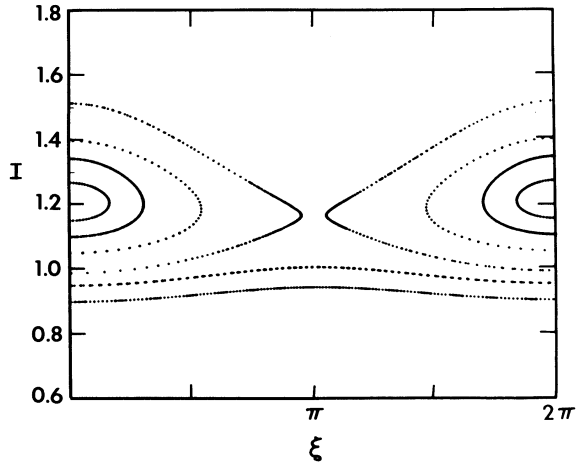

FIG. 3. Both trapped and passing trajectories are shown for the approximate Hamiltonian, Eq. (34), with $\Omega/\Omega_0=0.6$ and $FI_0^4/Z^3=0.04$. In this case the separatrix of the $m=1$ island located at $I_1=1.19$ has just succeeded in trapping some trajectories with initial action $I_0=1.0$.

can be solved to determine the width of the separatrix as a function of Ω and F.

Although K_1 can be approximated by the Hamiltonian of a pendulum, Eq. (16), for small F, estimates of F_t based on the width of the trapping region for the pendulum will tend to underestimate the critical field since the pendulum Hamiltonian fails to account for the asymmetry in the island growth which is illustrated by the trapped trajectories in Fig. 3. A more accurate estimate of the trapping field, F_t, for larger perturbations requires an explicit calculation of separatrix width for the Hamiltonian K_1.

Since the separatrix trajectory passes through the hyperbolic fixed point at $\xi=\pi$, the label for the separatrix orbit, k_s, can be calculated by evaluating the left-hand side of Eq. (35) at the fixed point. Since $d\xi/dt=\partial K/\partial I=0$ at the fixed point, the value of the action, I_s, at the hyperbolic fixed point is determined by the positive real root of the quartic equation

$$2\epsilon I_s^4+\Omega I_s^3-Z^2=0 . \tag{36}$$

Unfortunately, the exact expression for I_s in terms of the field strength and frequency is too complex to be useful. However, for near-resonance electrons we can approximate $I_s\simeq I_1=(Z^2/\Omega)^{1/3}$. Then using the fact that the separatrix reaches the lowest action at $\xi=0$, the minimum-field strength required to trap an electron with action I_0 is determined by

$$K_1(0,I_0)=k_s=K_1(\pi,I_s) . \tag{37}$$

The solution of this equation for F_t as a function of I_0 and Ω gives

$$F_t\simeq 3.08(Z^3/I_0^4)[(\tfrac{1}{2}+\delta-3\delta^{2/3}/2)/(1+\delta^{-2/3})] \tag{38}$$

measured in atomic units, where $\delta\equiv\Omega/\Omega_0\equiv\Omega I_0^3/Z^2$.

This estimate for the field strength required to trap an orbit with initial action I_0 in the single $m=1$ resonance should be valid for $\delta>0.25$. In the opposite limit the trapping field, F_t, can be calculated by expanding the equations for the separatrix width in small δ. In this case we get

$$F_t\simeq(0.265-0.49\delta)Z^3/I_0^4 . \tag{39}$$

The exact numerical evaluation of the single $m=1$ island width confirms the validity of Eq. (38) for $\delta>0.25$ and Eq. (39) for $\delta<0.25$. However, the low-frequency estimate for F_t exceeds the critical-field strength for static-field ionization for $\delta<0.28$. Consequently, the neglected Fourier components are expected to significantly modify microwave-field threshold for stochastic ionization in this limit.

C. Intermediate frequency regime

In the intermediate regime between the adiabatic and near-resonance limits, numerical integrations of the exact equations of motion for the perturbed SSE with a variety of microwave-field strengths and frequencies were used to estimate the critical field for the onset of stochastic dynamics. If the electron orbits were observed to exhibit stochastic behavior in a reasonable time (typically ~ 100 bounce periods), then the perturbation strength was assumed to exceed the critical threshold. If the electron orbits appeared to lie on passing (KAM) trajectories in action-angle space, then the field strength was assumed to be below critical. The results of these numerical simulations are shown in Fig. 4 along with a comparison of the analytical predictions in the adiabatic and near-resonance limits.

For $\delta>0.7$ the single-resonance theory provides an excellent description of the field, F_t, required to trap an electron in the $m=1$ resonance. However, in these cases the field threshold for the overlap of the $m=1$ and 2 island chains given by Eq. (24) is larger than F_t. Consequently, the critical field for stochastic excitation and ionization is determined by the overlap criterion alone. For $\delta<0.7$ the $m=1$ and 2 resonances overlap before the $m=1$ island reaches down to I_0. In this case the critical field is determined by the extent of the stochastic region in action-angle space. For δ between 0.5 and 0.7 the location of the $m=1$ separatrix predicted by the single-resonance theory, Eq. (38), provides a good estimate of the critical field for the onset of stochastic diffusion.

For microwave frequencies corresponding to $\delta<0.5$ the strong interaction of the other Fourier components of the perturbation generate secondary islands below the location of the $m=1$ separatrix. Some of these island chains are illustrated in Fig. 5 for a perturbation of the form $V(x,t)=xF\sin(\Omega t)$. These trajectories were calculated by numerically integrating the equations of motion for several initial conditions with action near $I=1$ in an oscillating electric field with a frequency corresponding to $\delta=\Omega/\Omega_0=0.55$. As the perturbation strength is increased

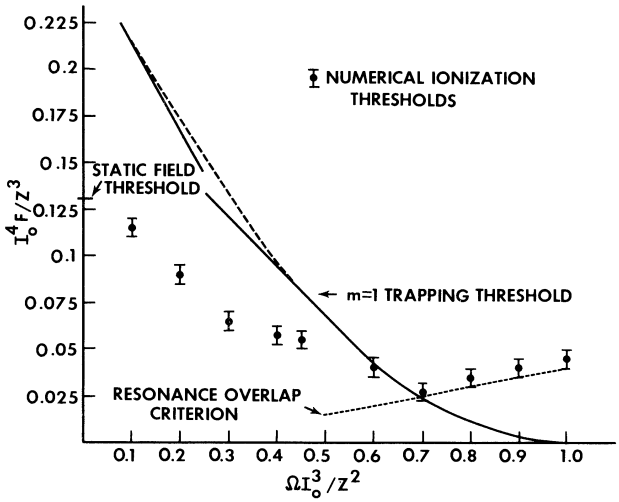

FIG. 4. Numerical calculations of the critical field for stochastic ionization in the quasistatic regime, $\Omega<\Omega_0$, are compared with the analytic predictions based on the resonance overlap criterion, Eq. (24), the $m=1$ trapping width, Eqs. (38) and (39), and the adiabatic theory, Eq. (31). The estimates for the critical field based on the large and small δ approximations for the $m=1$ trapping width are connected by exact numerical calculations indicated by the dashed curve.

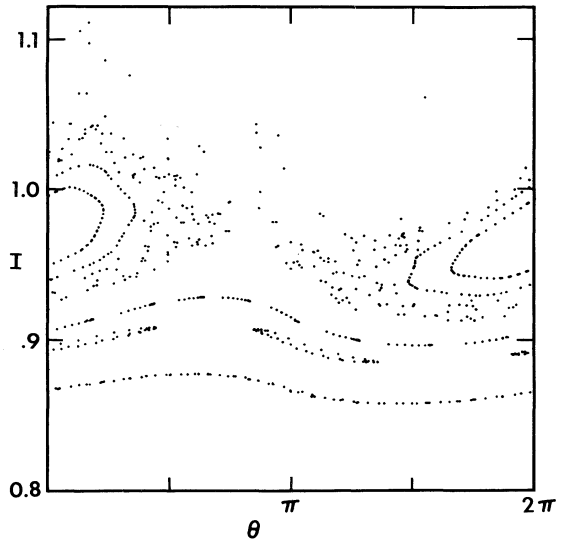

FIG. 5. Secondary islands generated by higher-order interactions of the primary subharmonic resonances are shown for a perturbation with $\delta=\Omega/\Omega_0=0.55$ and $FI_0^4/Z^3=0.06$. Island chains with 1, 2, and 8 islands as well as an apparent KAM surface are clearly visible well below the $m=1$ primary island which is located at $I_1=1.22$.

or δ is decreased the overlap of these secondary resonances causes the stochastic region in phase space to extend well below the location of the single, $m=1$ separatrix. Additional numerical studies of the equations of motion, generated by approximate action-angle Hamiltonians with four resonances corresponding to the $m=\pm1$ and ±2 Fourier components of the microwave perturbation, reveal that the biggest distortion of the $m=1$ separatrix towards lower action is due to the influence of the $m=-1$ Fourier component. The remaining resonances serve primarily to break up any lingering islands and KAM surfaces. As a consequence, the estimates of the critical field for the onset of stochastic diffusion obtained from the numerical simulations are significantly lower than those predicted by the single-resonance theory.

Finally, as δ is decreased further the numerical simulations indicate that the critical field for stochastic excitation and ionization connects smoothly onto the adiabatic prediction for static-field ionization, Eq. (31). Both the analytic and numerical estimates of the threshold fields shown in Fig. 4 provide upper bounds for the microwave fields required to ionize the classical SSE for quasistatic perturbations, $\delta<1$. These estimates should be useful in designing experiments to study the microwave ionization of SSE's. Furthermore, since the microwave ionization experiments with real hydrogen atoms[6–8] have been performed in this quasistatic regime, $\delta<0.5$, these new results can also be applied in the interpretation of these experiments.

D. Comparison of the quasistatic theory with experiment

In Refs. 6, 7, and 8, 9.9-GHz microwave fields were used to ionize highly excited hydrogen atoms with principal quantum numbers ranging from $n=26$ to 66, corresponding to frequency ratios of $\delta=0.026$ to 0.43. Although all of these experiments lie in the quasistatic regime significant differences were observed in the qualitative dependence of the ionization rate on the peak microwave field.[11] In the near-resonance limit, $\delta\sim0.4$, substantial ionization was observed for field amplitudes, $n^4F\sim0.05$ a.u., well below the classical threshold for static-field ionization of $n^4F\sim0.13$ a.u. predicted by Leopold and Banks.[29,30] Whereas, no ionization was observed for field strengths below $n^4F\sim0.11$ a.u. in the experiments in the adiabatic limit with $\delta\sim0.03$.

In an attempt to explain the experimental results and the classical, Monte Carlo simulations,[12] Meerson *et al.*[14,15] and Zaslavskii[16] have developed a classical description of the electron dynamics by approximating the Hamiltonian for the highly excited hydrogen atom in a microwave field with a Hamiltonian for a one-dimensional perturbed oscillator which is very similar to the one describing the perturbed SSE. However, Zaslavskii[16] used the resonance overlap criterion to estimate the critical field for the onset of stochastic diffusion which is not appropriate in this quasistatic regime. Moreover, in the absence of detailed numerical calculations Meerson *et al.*[14] used a heuristic estimate for the quasistatic perturbation strength required for stochasticity which gave a critical field of $n^4F_c\sim1/27\simeq0.037$. Although this prediction is only slightly lower than the near-resonance $n\sim66$ results, it significantly underestimates the threshold field in the adiabatic limit.

The analytical and numerical calculations of the quasistatic field thresholds for stochastic ionization, illustrated in Fig. 4, provide a qualitative explanation for both experimental limits. For $\delta\sim0.4$ the critical field for the onset of stochastic diffusion in the oscillating perturbation is a factor of 2 smaller than the static-field threshold, while the critical field approaches the static-field threshold for $\delta<0.1$ If we identify the initial action I_0 with the principal quantum number n and set $Z=1$, then the results for the perturbed SSE are also in quantitative agreement with the measured ionization thresholds for the highly excited hydrogen atoms.

IV. STOCHASTIC EXCITATION AND IONIZATION RATES

Once the critical-field strength for the onset of stochastic diffusion is exceeded, the motion of the perturbed SSE appears to be governed by a random process. In addition, the equations of motion exhibit "extreme sensitivity" to initial conditions[17] such that the orbits of classical electrons with nearby initial conditions evolve very differently. Since it is impossible in practice to specify the initial position and momentum of an electron with infinite precision, either classically or quantum mechanically, this chaotic or mixing[24] system is most conveniently analyzed using a statistical description. Simple random walk arguments applied to the motion of test particles in fluctuating fields can then be used to estimate the stochastic diffusion rate. Since the stochastic region in action-angle space is bounded from below, the electrons will tend to diffuse to

larger actions and energies until they ionize.

A typical trace of the time history of the position of a SSE in an ionizing microwave field with $\Omega/\Omega_0=0.6$ and $FI_0^4/Z^3=0.1$ is shown in Fig. 6(a). The electron bounces with irregular amplitude and period until it succeeds in escaping from the surface at $t\sim240$ which corresponds to ~23 microwave periods. Escape occurs after the electron has diffused to sufficiently high energies that it can slip over the peak of the potential barrier of height $V_t=-2(FZ)^{1/2}$ which is located at $x_t=(Z/F)^{1/2}=1.58a_0$ with sufficient kinetic energy to escape the Coulomb potential. Then the electron moves steadily away from the surface while oscillating in the microwave electric field with a mean velocity that is determined by the kinetic energy that the electron carries over the potential barrier.

Since the total energy of the SSE in the oscillating microwave potential can become positive without dissociating the electron, the compensated energy,[12] defined by

$$E_c=\tfrac{1}{2}\left[p+\int_0^t(\partial V/\partial x)dt\right]^2-Z/x\ , \tag{40}$$

must be used to determine whether the electron has gained enough energy to ionize. This definition removes the oscillatory contribution to the energy due to the time-dependent electric field. When E_c becomes positive and remains positive the electron has been successfully dissociated from the liquid-helium surface. Figure 6(b) shows the time history of the compensated energy for the ionizing trajectory illustrated in Fig. 6(a). At first the compensated energy wanders over a range of negative values. However, once E_c becomes positive, it quickly approaches a constant value corresponding to a free electron in the oscillating electric field.

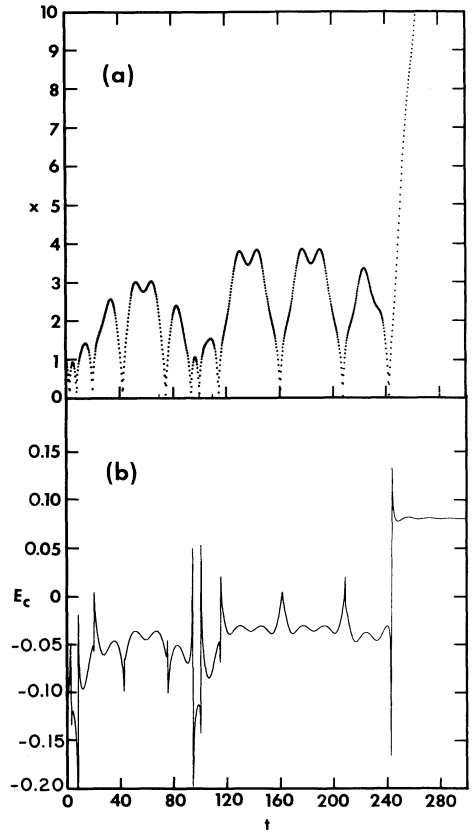

FIG. 6. (a) The excursion, x, in units of a_0 is shown as a function of time for a classical SSE in the presence of an oscillating electric field with $\Omega/\Omega_0=0.6$ and $FI_0^4/Z^3=0.1$. (b) The corresponding compensated energy, Eq. (40), is plotted as a function of time. Ionization occurs at $t\sim240$.

Since the Coulomb field is dominated by the microwave field for $x>x_t$, electrons which slip over the potential barrier will stop diffusing in action and energy. Consequently, if the mean velocity past the barrier is too low to ultimately escape from the Coulomb potential, then the electron will eventually return to the liquid-helium surface for another try. In Fig. 6(a) the electron is briefly trapped outside for $x>x_t$ on three occasions (indicated by the multiple peaks) before it finally escapes. In such cases the compensated energy can remain at a nearly constant negative value for long periods. If the microwave field is turned off while the electron is outside of the potential barrier, these slowly escaping electrons will be left in highly excited states. This stochastic excitation to "extremely" highly excited states has also been observed in numerical simulations of the three-dimensional hydrogen atom in a microwave field.[12]

Numerical integrations of the particle orbits for the perturbed SSE also reveal that some electron trajectories, for initial conditions near residual stable fixed points, remain regular (bounded in action and energy) for microwave fields in excess of the ionization threshold. However, these exceptional orbits, corresponding to Percival's "invariant tori,"[12] as well as the stable, extremely highly excited, "EHE" (Ref. 12) states are rare except for microwave-field strengths near threshold.

A. Quasilinear calculation of stochastic diffusion

When the critical field is exceeded and the islands overlap, most initial conditions lead to chaotic trajectories in phase space. A quasilinear[31,32] treatment of the evolution of the distribution of trajectories in action, $F(I,t)$, leads to a Fokker-Planck type diffusion equation,

$$\frac{\partial F}{\partial t}=\frac{\partial}{\partial I}D(I)\frac{\partial}{\partial I}F\ . \tag{41}$$

This procedure is valid as long as the perturbation is not too large. For very large microwave fields, $F\geq F_0$, the electron can be dissociated in one wave period. In this case the electron ionizes in a single jump rather than a sequence of small, random steps.

The diffusion coefficient in action, $D(I)$, can be calculated by considering the motion of a test particle in action-angle space under the influence of the many Fourier modes of the microwave perturbation. This test particle diffusion coefficient is defined by

$$D(I)=\lim_{t\to\infty}\frac{1}{2t}\int_0^t dt'\int_0^t dt''\langle\dot{I}(t')\dot{I}(t'')\rangle\ , \tag{42}$$

where $\dot{I}(t)\equiv dI/dt$ is evaluated along the particle orbit determined by the equations of motion in action-angle space,

$$\frac{d\Theta}{dt}=\Omega_0(I)+F\sum_{m=-\infty}^{\infty}\frac{\partial V_m}{\partial I}\cos(m\Theta-\Omega t)\ , \tag{43}$$

$$\frac{dI}{dt}=F\sum_{m=-\infty}^{\infty}mV_m\sin(m\Theta-\Omega t)\ , \tag{44}$$

and the average,

$$\langle\cdots\rangle\equiv\frac{1}{2\pi}\int_0^{2\pi}(\cdots)d\Theta_0\ ,$$

is taken over an ensemble of initial angles (phases), $\Theta_0=\Theta(0)$.

Using the approximate solution of Eq. (43), $\Theta(t)\simeq\Theta(0)+\Omega_0 t$, we can evaluate the integrand in Eq. (42) along the free-streaming trajectories. Then the average over random phase, Θ_0, gives

$$\langle\dot{I}(t')\dot{I}(t'')\rangle\simeq F^2/4\sum_{m=-\infty}^{\infty}m^2\,|\,V_m\,|^2e^{-im\Omega_0\tau}\times2[\cos(\Omega\tau)+\cos(2\Omega T)]\ , \tag{45}$$

where we have introduced $\tau\equiv(t'-t'')$ and $T=(t'+t'')/2$. In terms of these new variables the test particle diffusion coefficient can be written as

$$D(I)=\lim_{t\to\infty}\frac{1}{2t}\int_{-t}^{t}d\tau\int_{\tau/2}^{t-\tau/2}dT\langle\dot{I}\dot{I}\rangle(\tau,T)\ . \tag{46}$$

Since the integral over $\cos(2\Omega T)$ remains bounded as $t\to\infty$ these terms will not contribute to the diffusion coefficient. The remaining terms are independent of T. Therefore, the integral over T simply gives a factor of $t-\tau$. Moreover, if we assume that the autocorrelation function, $\langle\dot{I}\dot{I}\rangle(\tau,T)$, decays to zero as $\tau\to\infty$ due to nonlinear mixing,[32] then the τ integral can also be performed which reduces Eq. (46) to the standard quasilinear result[31]

$$D(I)=F^2/4\sum_{m=-\infty}^{\infty}m^2\,|\,V_m\,|^2\,2\pi\delta(m\Omega_0-\Omega)\ . \tag{47}$$

Finally, if we approximate $\Omega\simeq m\Omega_0$ and replace the infinite sum by an integral over m, then using Eq. (19) we get a simple expression for the local diffusion coefficient for $I\simeq I_m$ in terms of the amplitude and frequency of the perturbing microwave field,

$$D(I)\simeq(\pi/2)F^2m^2\,|\,V_m\,|^2/(\Omega Z^2)\simeq0.27I^3F^2/(\Omega Z)^{4/3} \tag{48}$$

measured in atomic units for large m.

B. Stochastic excitation rate

This local diffusion coefficient can be used to calculate the characteristic time, $\tau_d=\delta_m^2/2D(I_m)$, required for an electron to diffuse a distance, $\Delta I=\delta_m\equiv I_{m+1}-I_m$, in action. Since small excursions up and down occur with equal probability, the characteristic time required for an electron to be excited to larger actions, from the m to the $m+1$ classical resonance, is $2\tau_d$. Consequently, the excitation rate from the m to the $m+1$ resonance is given in atomic units by

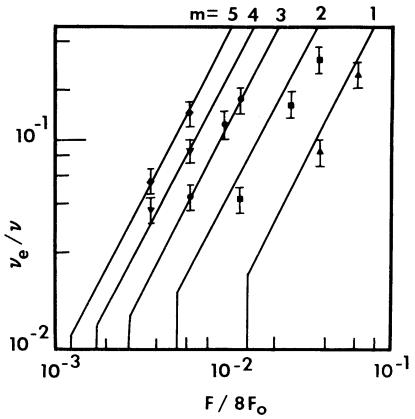

FIG. 7. Theoretical, Eq. (49), and numerical excitation rates from the m to $m+1$ resonances are plotted as a function of the electric field strength for the $m=1$–5 subharmonic resonances with a microwave perturbation at $\Omega=320$ GHz. The five different symbols for the numerical data points correspond to the different values of m. The error bars represent an estimate of statistical errors in the Monte Carlo calculation. No excitations were observed, numerically, for $F<F_c\simeq0.2F_0$ as predicted by Eq. (24).

$$\nu_e=D/\delta_m^2\sim2.4F^2m^{7/3}/(\Omega^{5/3}Z^{2/3}) \tag{49}$$

measured in atomic units for large m. Since the local excitation rate is a rapidly increasing function of m, the stochastic diffusion to larger actions accelerates rapidly once the electron has succeeded in diffusing past the $m+1$ resonance. It therefore provides a convenient order of magnitude estimate for the stochastic ionization rate.

Our analytic estimates for the stochasticity threshold, Eq. (24), and the excitation rate, Eq. (49), have been verified by numerical integrations of the perturbed equations of motion. For small electric fields the electrons remained confined near their initial action (see Fig. 1); however, as the field was increased above the threshold, the trajectories spanned several resonances indicating the breakup of confining KAM (Ref. 13) surfaces. The numerical results for both the stochastic threshold and excitation rate are compared with the analytic predictions as functions of microwave field in Fig. 7 for initial actions near the first five classical resonances, $m=1$–5. Although the resonance overlap criterion is only expected to be accurate to within a factor of 2,[24] the quasilinear diffusion estimate is very reliable once the dynamics have become fully chaotic. This is illustrated by the excellent agreement between the theoretical and the numerical excitation rates.

V. DISCUSSION

The classical description of the nonlinear dynamics of a surface-state electron in a microwave electric field predicts a transition from regular to stochastic behavior when the electric field strength exceeds a critical threshold. By combining numerical simulations with simple analysis, we have derived convenient estimates for the threshold field as a function of the initial action or energy

of the SSE and the frequency of the perturbation. This classical treatment of the perturbed SSE fills in the details of a previous calculation[18] of the critical field based on the resonance overlap criterion for the onset of global stochasticity[17] and provides new results for the critical field for ionization in quasistatic perturbations. When the threshold field is exceeded, the SSE can diffuse to higher energies until it ionizes. Using a statistical, random walk description of the chaotic electron dynamics, we have also calculated the stochastic excitation and ionization rates as a function of the initial action of the SSE and the strength and frequency of the perturbation.

The perturbed SSE promises to be an ideal system for the investigation of quantum chaos. Since the system is one dimensional the theoretical analysis of this nonlinear oscillator is greatly simplified. In addition, the low binding energies and frequencies of the SSE permit experimentalists to probe the lowest quantum levels with available microwave sources. Our classical calculations of field thresholds and ionization rates then provide estimates for the microwave frequencies and powers required to experimentally explore the manifestations of classical chaos in this quantum system. If the classical stochasticity of this driven oscillator persists in the quantum regime, experimentalists should observe both enhanced line widths for the quantum levels and measurable ionization rates which increase as functions of microwave power. Moreover, since the quantum mechanical and semiclassical treatments of this one-dimensional hydrogen atom may also prove to be tractable, the predictions of these theories can also be tested with real experiments.

The feasibility of the proposed experiment can be assessed by calculating the critical field and ionization rate for the stochastic ionization of the ground-state SSE with initial action and quantum number $I_0=n=1$. According to Eqs. (7)—(10) the ground-state SSE has a binding energy of $E_0\simeq -6.8\times 10^{-4}$ eV, a natural oscillation frequency of $\nu_0\simeq 330$ GHz and a binding electric field at the maximum classical excursion of $F_0\simeq 450$ V/cm. If the frequency of the applied microwave field is $\nu\simeq 330$ GHz, then $I_0\simeq I_1$ and stochastic diffusion occurs when the $m=1$ and $m=2$ island chains overlap. In this case Eqs. (24) and (49) predict that a peak microwave electric field of $F\simeq 115$ V/cm is sufficient to classically ionize the ground-state SSE at a rate $\nu_e\simeq 14$ GHz. This peak field corresponds to a microwave intensity of $I_c\simeq 35$ W/cm^2.

If the experiment is performed in a microwave cavity, then the critical intensity is reduced by a factor equal to the Q of the cavity. Moreover, Eq. (24) predicts that the critical field decreases for higher-frequency perturbations since the initial action of the SSE, I_0, then corresponds to a classical resonance with $m>1$. However, according to Fig. 4, the critical field increases for microwave frequencies much less than ν_0 and approaches the static field, $F_c\simeq 230$ V/cm, required to classically dissociate the electron in the adiabatic limit.

Although microwave sources which can deliver 330 GHz at intensities >10 W/cm^2 are not yet readily available, the microwave intensity required to ionize the excited states of the SSE are significantly lower. Consider, for example, an experimental configuration similar to that used in Grimes's[19] original experimental studies of the quantum levels of the SSE's consisting of a microwave cavity half filled with liquid helium at 1.2 K. The SSE's are created by charging up the liquid-helium surface. If the cavity is designed to resonate with microwaves at a frequency of $\nu\simeq 120$ GHz, corresponding to the frequency for the single-photon excitation to the $n=2$ level, then the application of low-intensity microwaves will populate the first excited state. The classical binding field for the $n=2$ level, $F_0\simeq 28$ V/cm, is $n^4=16$ times smaller than the binding field of the ground state. In addition, the classical bounce frequency is $\nu_0\simeq 40$ GHz which implies that the initial action is resonant with the $m=3$ subharmonic of the microwave perturbation. Therefore, if the microwave intensity is slowly increased, then the classical theory, Eq. (24), predicts that the $n=2$ states will start to ionize when the microwave fields exceed $F_c\simeq 2.7$ V/cm. This critical field corresponds to a microwave intensity of only $I_c\simeq 2\times 10^{-2}$ W/cm^2. For high-Q cavities this source intensity is further reduced by the cavity Q.

The classical treatment of this one-dimensional hydrogen atom in a microwave field gives ionization thresholds and rates which are in qualitative agreement with the experimental measurements of the microwave ionization of real hydrogen atoms performed by Bayfield and Koch.[6–8] This suggests that the physical ionization mechanism in the experiments[6–8] and the numerical simulations[12] is also stochastic diffusion due to overlap of classical resonances in action-angle space. The same procedure used to analyze the nonlinear classical dynamics of the SSE can also be extended to the Hamiltonian,

$$H(\vec{r},\vec{p},t)=p^2/2-Z/r+xF\cos(\Omega t) \tag{50}$$

measured in atomic units which describes the dynamics of a classical electron orbiting the nucleus in the presence of an oscillating electric field. Although the classical, six-dimensional, action-angle phase space can be reduced to four dimensions by exploiting the cylindrical symmetry about the direction of the electric field, the analysis is considerably more complicated than that for the perturbed SSE because of the interplay of the additional degrees of freedom. For example, in higher dimensions the KAM surfaces no longer confine the phase-space trajectories. So the electron can always wander to higher actions or energies by "going around" these two-dimensional surfaces in the four-dimensional space. Consequently, there is no well-defined threshold for the onset of stochastic diffusion. However, this "Arnol'd diffusion,"[24] is usually extremely slow and rapid ionization due to stochastic diffusion will not occur until most of the KAM surfaces between the classical resonances are destroyed.

Meerson *et al.*[14,15] and Zaslavskii[16] have attempted to calculate the electric field threshold for the destruction of KAM surfaces due to island overlap by transforming the classical equations of motion to appropriate action-angle variables and using a multiple time-scale analysis to reduce the problem to one dimension. Although a number of details remain to be worked out, rough estimates of

the stochasticity threshold when the microwave frequency is close to the oscillator frequency are in good agreement with the experimental results for hydrogen atoms in the $n \sim 66$ level. However, these preliminary calculations are not adequate to explain the experimental results for the quasistatic experiments ($n \sim 29$). More extensive theoretical and experimental studies of Rydberg atoms in oscillating electric fields are currently being pursued to further explore the dynamics of these perturbed, nonlinear oscillators.

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