

## Localized chaos in one-dimensional hydrogen

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We calculate the response of hydrogen to the presence of both a strong dc electric field (necessary to isolate a nearly one-dimensional motion) and a strong radiation field of higher frequency than the binding energy of the system, a regime that has not previously been examined by theory or experiment. We determine the classical ionization threshold, the quantum-delocalization threshold, and the threshold of  $n$  mixing due to chaotic effects. The analysis indicates that the dc field can have a dramatic effect on the quantum localization of classically chaotic diffusion, changing the delocalization threshold by more than an order of magnitude. Moreover, this system provides a large spectral region in which quantum-mechanical localization inhibits classical chaotic diffusion. This theory is well suited to experimental testing.

The flourishing of the study of nonintegrable classical systems has stimulated interest in the equivalent quantum systems, especially because quantum mechanics was developed and tested from analogies with and observations of systems which classically are integrable.<sup>1</sup> A great deal of theoretical effort has been expended in an attempt to understand the quantum dynamics of systems which are classically chaotic.<sup>1-6</sup> The debate concerning the relevance of classical chaos to quantum systems has important implications to the foundations of quantum mechanics and its relation to classical mechanics.<sup>1</sup>

It has been recognized for quite some time now that because all closed, bounded quantum systems are quasiperiodic in time (can be fully described by a sum of discrete harmonic frequencies), they cannot exhibit true chaos (positive Lyapunov exponents).<sup>1</sup> This result has been extended to bounded systems with periodic Hamiltonians.<sup>7</sup> However, experiments and numerical simulations on such systems have shown some results that agree with predictions from classical chaos theory,<sup>8</sup> suggesting a quantum mimicking of classical chaos.

One of the quantum systems that received considerable interest in this regard is the highly excited one-dimensional hydrogen atom in the presence of intense low-frequency radiation (microwaves). The problem was studied extensively classically, quantum mechanically, and by experiment.<sup>2,9-14</sup> The intensity threshold for inducing chaos in the classical system was found to give the quantum-mechanical ionization threshold for radiation frequencies lower than the natural atomic frequency. However, the two thresholds were found to diverge from each other for radiation frequencies larger than the atomic frequency. In this regime, the classical threshold continues to drop at very high frequencies, while the quantum threshold rises with frequency.

The divergence of the quantum and classical results in the new frequency region was predicted by the theory of dynamical localization of chaos in quantum systems,<sup>15,4,2</sup> which was developed in conjunction with numerical experiments and predicts that the evolution of a quantum system will mimic its classically chaotic counterpart as

long as its quasiperiodic frequency components are not resolved. This theory was applied to both the quantum Chirikov map (or "standard map")<sup>4</sup> and the one-dimensional hydrogen atom<sup>2</sup> in an external ac field, for driving frequencies within a factor of 3 of the natural frequency of the system. This effect, often described as a dynamical version of Anderson localization in solids, limits the chaotic diffusion of the energy of the system for cases in which the external field has passed the chaos threshold but not a new and higher threshold, the delocalization threshold.<sup>2</sup> The atom-external-field system is said to be delocalized if it ionizes before its quasiperiodic frequency components are resolved. A number of very successful experiments have been performed measuring ionization thresholds and excitation rates for highly excited hydrogen in microwave fields, giving agreement with the delocalization theory.<sup>11-13,16</sup> However, these experiments only probed the edge of the regime of disagreement between classical and quantum-mechanical predictions, and were not able to conclude on the basis of experiment alone that the quantum localization of the classically chaotic diffusion actually occurs.<sup>16</sup>

We analyze here the implications of such effects on a system, a highly excited hydrogen atom in combined dc and ac electric fields (the ac field produced by a CO<sub>2</sub> laser) that is amenable to measurement and calculation of the localization effects. First, we have included the effect of a strong dc field on the delocalization and chaos threshold in general. This is important, because the dc field allows the separation of a nearly one-dimensional motion, amenable to theoretical treatment, from other motions present in the system. The effect of a strong dc field is drastic, and gives rise to changes of more than an order of magnitude in some of the chaos and delocalization thresholds. Because the dc field is a parameter under external control, this allows one to perform a larger class of experiments.

Second, we apply the localization theory to a new regime of very high frequency, where the frequency of the ac field is greater than the frequency associated with the binding energy of the system. In this case, the additional

frequency components (sidebands) induced by the ac field fall very far outside the width in energy (or frequency) of the region of phase space involved in the motion. The theory has been previously applied to cases in which the sidebands fall within the region of phase space involved in the motion (low to medium frequency).

We also introduce a criterion, based on the localization theory, for the appearance of large changes in the spectral structure caused by the ac field, although we are not able to predict the details of the line positions or strengths. This result may help to understand under what circumstances an intense spectroscopy laser might be expected to change the spectrum it is measuring.

Let us begin by introducing the hypothesis of quantum localization of classical chaos.<sup>4,2</sup> The quasiperiodic quantum system follows the chaotic classical diffusive behavior until a time  $t_B$  long enough that the frequency differences between the periodic frequency components of its motion are resolved. We mean "follows" the classical behavior in the sense that the quantum levels denoted by a range  $\Delta n(t)$  of principal quantum number  $n$ , and only those levels, take an important part in the motion of the system, where  $\Delta n(t)$ , is now interpreted as a range of *classical action*  $n$  (Ref. 17) and denotes a region of *classical* phase space the classical motion is limited to at time  $t$ . We take advantage of the correspondence between classical action (in atomic units<sup>18</sup>) and principal quantum number, known before Schrödinger's equation,<sup>19</sup> and, since we will have no need here to distinguish between them, denote them both by the same symbol  $n$ . After  $t_B$ , the diffusive motion stops and the quantum system is localized to whatever region of phase space ( $\Delta n_B$ ) the classical chaotic system diffused to before the localization time  $t_B$ .<sup>4,2</sup>

To apply this principle, one must decide on a criterion for the resolution of the frequency differences. Direct numerical integration of Schrödinger's equation in several systems<sup>4,2</sup> has shown the following criterion to be relevant. The frequency difference that must be resolved is the local average of the nearest-neighbor spacings of the frequencies. Then, two chosen frequencies  $\omega_a$  and  $\omega_b$  are considered resolved when the phase difference  $\phi = (\omega_a - \omega_b)t$  is greater than  $2\pi$ .<sup>2</sup> It makes sense to take only those frequency components related to the region of phase space into which the system diffuses classically before  $t_B$ , because only those frequency components are excited by the motion. Since the frequency components one uses to calculate the extent of localization are dependent on the extent of localization, one needs to do a self-consistent calculation to find  $t_B$ . Sometimes this self-consistent equation has no solution. Then the system does not localize. In those cases, the quantum system follows the classical diffusive behavior forever, or at least as long as anyone has been able to measure, in either experiments or numerical simulations. This phenomenon, called "delocalization," appears to be responsible for the agreement between predictions of chaos thresholds and measurement of microwave ionization thresholds of highly excited states of hydrogen.

To apply the localization hypothesis as stated above, one needs to know what frequencies are present in the

motion, so we would like to briefly discuss the frequency spectrum of a bounded system with a Hamiltonian  $H(t)$  periodic in time with period  $T = 2\pi/\Omega$ . The Hamiltonian of the experiment described here is, in the dipole approximation, in atomic units,<sup>18</sup>

$$H(t) = \frac{p^2}{2} + \frac{1}{x} + F_{dc}x + F_{ac}x \cos\Omega t . \quad (1)$$

Although this system is not bounded, the following analysis has agreed with experiment and numerical simulation in the low to medium frequency,  $F_{dc} = 0$  case.<sup>2</sup> The reason for this agreement is not yet clear; perhaps only bound states are involved in the motion below the threshold of ionization, and so only a bounded motion is important to calculations of this threshold. In any case, we will assume the relevance of the bounded analysis to the current system.

Because the Hamiltonian is time dependent, energy is not conserved and there are not even any energy eigenstates. However, we can deduce certain qualities of the frequency spectrum which are useful in motivating the localization theory. The solutions  $\Psi(x, t)$  can be written<sup>7,20</sup> as

$$\Psi(x, t) = \sum_j c_j e^{i\omega_j t} \Psi_j(x, t) , \quad (2)$$

with quasienergies<sup>7</sup>  $\omega_j$ , and quasienergy eigenstates

$$\Psi_j(x, t + T) = \Psi_j(x, t) \quad (3)$$

forming a complete orthonormal set at all times.<sup>7</sup> For a constant Hamiltonian (that is, as the ac field goes to zero), the quasienergies are just the true energies and the quasienergy eigenstates just the energy eigenstates. Expand each periodic quasienergy eigenstate in a Fourier series

$$\Psi_j(x, t) = \sum_k a_{jk}(x) e^{-ik\Omega t} ,$$

and Eq. (2) becomes

$$\Psi(x, t) = \sum_j \sum_k c_j a_{jk}(x) \exp[-i(\omega_j + k\Omega)t] . \quad (4)$$

Equation (4) is saying that the frequency spectrum of the evolution of the system consists of the sum of the frequency dependencies of its component quasienergy eigenstates. The frequency components of a single quasienergy eigenstate are just its central quasienergy  $\omega_j$ , plus sidebands spaced by the externally imposed frequency  $\Omega$ . The values of the quasienergies are difficult to solve for, but the nature of the sidebands can be clarified by a simple example of sidebands in a two-level atomic hydrogen system.<sup>21</sup> Once prepared in a quasienergy eigenstate, the system will stay in it forever.

We now briefly present the physical argument leading to the equation describing the localization. In order to find the quantum-mechanical limitation on classically chaotic diffusion, we need to use a quantum-mechanical quantity closely related to the classical diffusion. Since, for simplicity, the classical diffusion calculation starts (see below) with the distribution function  $f(n, t)$

$=\delta(n-n_0)$ , concentrated at an initial (zero ac field) action (all values of the classical angle variable  $\theta$  are included equally), it makes sense to use the frequency components of the system prepared in the (zero ac field) energy eigenstate denoted by  $n_0$  to apply the localization condition limiting classical diffusion from an initial state. This is true whether the actual initial state excited in the experiment is the energy eigenstate denoted by  $n_0$ , as it has been in ionization threshold experiments, or some quasienergy eigenstate. It can be shown, assuming a correspondence between occupation of a region of the classical phase space and occupation of the quantum energy eigenstates of that same region of  $n$ , that the energy eigenstate  $n_0$  can be expressed as an expansion of approximately  $\Delta n_B$  quasienergy states, where  $\Delta n_B$  gives the width of the region of phase space to which the classical diffusion is localized.<sup>2,4</sup>

Given the number of quasienergy eigenstates involved in the initial state  $n_0$ , we can motivate the self-consistent equation for localization. Equation (4), because it associates a set of frequencies with each quasienergy eigenstate, can give a set of frequencies associated with a set of quasienergy eigenstates. If only one quasienergy eigenstate is involved ( $\Delta n_B < 1$ ), then the density of discrete frequencies is simply  $1/\Omega$ , one level in each space of width  $\Omega$  given by the sidebands associated with that single quasienergy eigenstate. If  $\Delta n_B$  quasienergy levels are involved in the motion, as described above, the sidebands of all of these levels must be included, and then the density of discrete frequencies becomes  $\Delta n_B/\Omega$ . Then the average nearest-neighbor spacing of discrete frequencies involved in the motion is  $\Omega/\Delta n_B$ . The localization hypothesis is that the average nearest-neighbor spacing be resolved by a  $2\pi$  criterion at the localization time  $t_B$ , or  $(\Omega/\Delta n_B)t_B = 2\pi$ . This gives the self-consistent equation

$$\Delta n(t_B) = (\Omega/2\pi)t_B. \quad (5)$$

The localization "length"  $\Delta n_B$  is defined as  $\Delta n(t_B)$ . Equation (5) was applied to the hydrogen atom in a microwave field with no external dc field.<sup>2</sup> This led to a prediction that localization would only occur when the microwave frequency exceeded the natural frequency of the initial atomic state, in agreement with experiment and numerical simulation.<sup>2</sup>

Given the already demonstrated importance of high-frequency external fields, we now extend this theory to the very high external frequency (high- $\Omega$ ) regime, where the spacing of the sidebands associated with one quasienergy eigenstate is much larger (a factor on the order of 100 for the physical system discussed here) than the average spacing of the quasienergies, which is the same<sup>2</sup> as the spacing of the original energy eigenstates labeled by  $n$ . If this is the case, and only a small range  $\Delta n_B$  of states are excited, the spacing of the frequency components has an unusual quality not seen in the low or medium frequency cases. It is a small  $\Delta n_B$  set of closely spaced levels, followed by a very large gap to another set of closely spaced sidebands, followed by another large gap and set of closely spaced sidebands, etc. It is possible that, in this case, the correct frequency difference to use in the locali-

zation criterion is the frequency difference between the original levels, excluding the sidebands, which is smaller than the  $\Omega/\Delta n_B$  used above, making localization more difficult and increasing the localization time  $t_B$ . This hypothesis leads to the self-consistent equation

$$\Delta n(t_B) = n_{\text{orbit}}(t_B) - n_0, \quad (6)$$

where  $n_{\text{orbit}}(t)$  is the function relating the orbital time of the classical system to the classical action.

Considering a conceivable limitation of Eq. (6) leads to a third possibility which combines Eqs. (5) and (6). Consider the situation with  $\Delta n_B < 1$ . Then the frequency components are just the sidebands of one quasienergy level, and it seems inappropriate to use the spacings of other levels not involved in the motion, which led to Eq. (6). For  $\Delta n_B < 1$ , the frequency components are always resolved at  $t = 2\pi/\Omega$ , and so one might expect the diffusion to stop right then and never reach the region described by Eq. (6) where other closely spaced levels are involved in the motion. This equation,  $t_B = 2\pi/\Omega$ , is just Eq. (5) but with  $\Delta n_B = 1$ . This reasoning leads to the following proposal. Perhaps we should first compute  $\Delta n_B$  from Eq. (5). If it is less than one, the system localizes before any other  $n$  levels have a chance to be involved in the motion. If it is greater than one, then we should apply Eq. (6) to get the true  $\Delta n_B$  and  $t_B$ .

The above analysis involves the criterion  $\Delta n_B > 1$ , which may also be important for other reasons. It is the condition for more than one quasienergy eigenstate to be involved in the expansion of one energy eigenstate, which is the same as the condition for more than one energy eigenstate to be involved in the expansion of one quasienergy eigenstate.<sup>2</sup> This means that it is simply a condition for mixing of the original levels. As such, it may very well be the condition for large effects due to chaos to appear in the spectrum of the system.

We now discuss the procedure for solving Eqs. (5) and (6). We need to know the function  $\Delta n(t)$ , which requires some classical chaos theory. Of course,  $\Delta n(t)$ , the region of chaotic diffusion, will be zero if there is no chaos and no diffusion. Therefore, one must first calculate the threshold of global classical chaos, including the effect of a strong dc field. This threshold is the threshold of overlap of the nonlinear resonances.<sup>22</sup> Recently, these effects were calculated by a number of methods.<sup>9,14,10</sup> The chaos thresholds for the case of an ac field of frequency  $2.82 \times 10^{13}$  Hz (CO<sub>2</sub> laser) and dc fields of  $-2, 0$ , and  $10$  kV/cm are given in Fig. 1, along with other data. The exact procedure used here, with detailed explanations of the theory and results, can be found in a previous publication.<sup>10</sup> The chaos thresholds in the region we are interested in here ( $n = 15$  to  $25$ ) are much lower than the intensity regime we will be interested in, and so, in the following, we will assume that the system is already above the threshold of global chaos. One should keep in mind, however, that the following analysis is only valid above the threshold of classical chaos as given in Fig. 1.

The irregular motion of the electron above the threshold of classical chaos leads to a diffusion in the size of the region of phase space within which it travels. This classi-

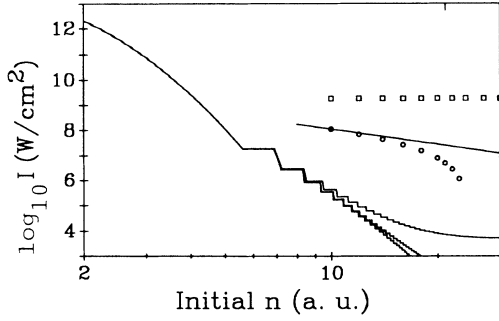


FIG. 1. Thresholds of classical chaos, in laser intensity  $I$ , are plotted as a function of the initial principal quantum number, or classical action,  $n$ , for a one-dimensional hydrogen atom in a combined dc and CO<sub>2</sub> laser field. Some thresholds of quantum-mechanical delocalization, calculated from Eq. (5), are presented for comparison. From lower left to upper right:  $F_{dc} = -2$  kV/cm, 0, 10 kV/cm chaos thresholds,  $F_{dc} = -2$  kV/cm (circles), 0, 10 kV/cm (squares) delocalization thresholds.

cal chaotic diffusion can be described by the Fokker-Planck equation

$$\frac{\partial}{\partial t} f(n, t) = \frac{\partial}{\partial n} \frac{D(n)}{2} \frac{\partial}{\partial n} f(n, t), \quad (7)$$

where  $f(n, t)$  is the probability distribution of the system over some variable or variables representing the phase space. In this case,  $f(n, t)$  is a probability distribution over the classical action (averaging over the classical angle  $\omega$ ), which is approximately equal to the principal quantum number  $n$ , so we label it  $n$ . The diffusion parameter  $D(n)$  can be derived from the quasilinear theory<sup>23</sup> under the assumption that correlations decay

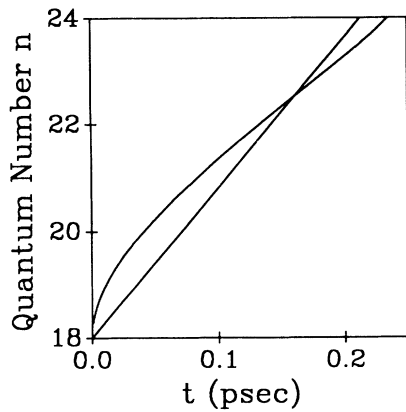


FIG. 2. A plot of the curves  $\Delta n(t)$  as calculated from Eq. (10) and  $\Delta n = (\Omega/2\pi)t$ , to solve the self-consistent equation (5). The localization time and length,  $t_B$  and  $\Delta n_B$ , are given by the intersection of the two curves. The  $y$  coordinate is labeled by  $n = n_0 + \Delta n$ . Here  $n_0 = 18$ ,  $F_{dc} = -2$  kV/cm, and the laser intensity is  $1.4 \times 10^7$  W/cm<sup>2</sup>.

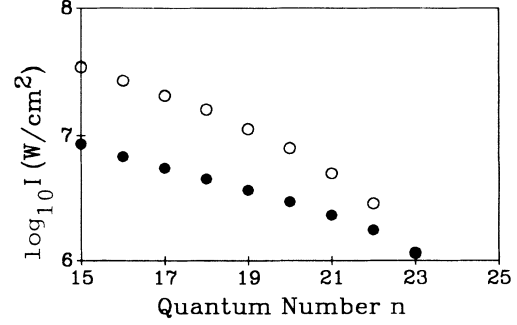


FIG. 3. Delocalization and  $n$ -mixing thresholds  $I_d$  (open circles) and  $I_{dn1}$  (solid circles) for the  $F_{dc} = -2$  kV/cm case, for principal quantum numbers between 15 and 25.

due to chaos, a procedure that gives agreement with numerical simulations in the case  $F_{dc} = 0$ .<sup>9</sup> The result is

$$D(n) = \pi F_{ac}^2 V_m^2 m^3 / \Omega, \quad (8)$$

where  $m$  is the index of the Fourier component of the oscillatory motion of the system most nearly resonant with the external frequency  $\Omega$ , and  $V_m$  is a parameter representing the strength of the driving force at that Fourier component.<sup>9</sup> The parameters  $m(n)$  and  $V_m(n)$  were computed using a numerical integration procedure for the cases with nonzero dc field; this procedure was described in detail previously.<sup>10</sup> For simplicity, the initial probability distribution is taken as  $f(n, t=0) = \delta(n - n_0)$ . The system will be chaotic only above a certain  $n = c$ , giving the boundary condition  $(df/dn)_c = 0$ .<sup>2</sup>

The quantity required from this analysis is  $\Delta n(t)$ , which is some approximate measure of the width of the distribution  $f(n, t)$ . In the case of zero dc field, it was possible to solve the Fokker-Planck equation in closed form, and then the rms deviation was given in terms of an expansion, and used for  $\Delta n$ .<sup>2</sup> In the present case, an analytical formula for  $D(n)$  is not available, only numerical values, and a different procedure is indicated. We use a heuristic method to define and compute  $\Delta n(t)$  which gives the same parameter dependence and similar (smaller by a factor of 0.8) absolute value of the delocalization threshold as the method described above for the case of

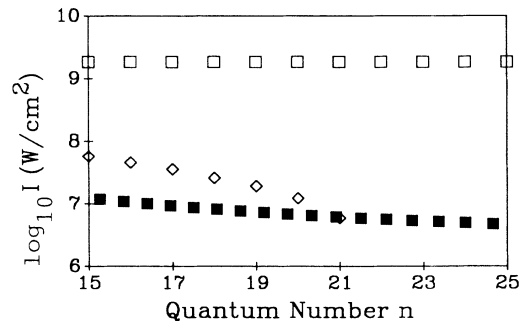


FIG. 4. Thresholds  $I_d$  (open squares),  $I_{E0}$  (open diamonds), and  $I_{dn1}$  (solid squares) for the  $F_{dc} = 10$  kV/cm case.

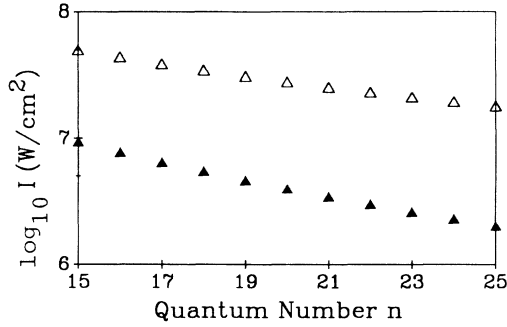


FIG. 5. Thresholds  $I_d$  (open triangles) and  $I_{dn1}$  (solid triangles) for the  $F_{dc}=0$  case.

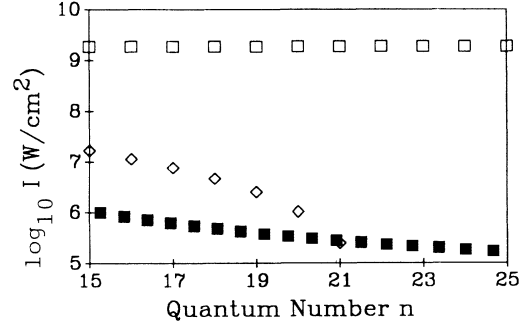


FIG. 7. Thresholds  $I_{dhf}$  (open squares),  $I_{E0hf}$  (open diamonds), and  $I_{dn1hf}$  (solid squares) for the  $F_{dc} = 10$  kV/cm case.

zero dc field, and is amenable to numerical solution, which allows an extension of the theory to the case of nonzero dc field. This approach can be motivated by a particular definition of  $\Delta n(t)$  and making some approximations to the Fokker-Planck equation. Since the width of  $f(n,t)$  is bounded below  $n_0$  by the chaos threshold  $c$ , we use the width above  $n_0$ , with some additional factors for simplicity, to define  $\Delta n(t)$ , given by

$$\Delta n = (\pi^{1/2}/2) \int_{n_0}^{\infty} (n - n_0) f(n,t) dn, \quad (9)$$

the mean difference between  $n$  above  $n_0$  and  $n_0$ , or the average of  $n$  above  $n_0$  minus  $n_0$ , times the fraction of  $n$  above  $n_0$ , with a factor of  $\pi^{1/2}/2$  to simplify the result. For large  $\Delta n$  the fraction of  $n$  above  $n_0$  is nearly one, because the diffusion is bounded from below but not from above, and this definition of  $\Delta n$  simply reduces to the average of  $(n - n_0)$ , with an extra factor of  $\pi^{1/2}/2$ . We then multiplied both sides of the equation by  $(n - n_0)$  and integrated from  $n_0$  to infinity, approximating the  $D(n)$  on the right-hand side by an average diffusion constant  $D(n_0 + \Delta n(t))$  (making it a function of time rather than space), and the  $f(n,t)$  on the right by  $f(n,t)$  for the problem with a constant  $D$  equal to this average  $D$ . This gives

$$\frac{d\Delta n}{dt} = \frac{1}{2} [D(n_0 + \Delta n(t))/t]^{1/2}, \quad (10)$$

which was solved by a numerical technique which involved taking the exact solution for constant  $D$  in small

intervals within which  $D$  was approximately constant, allowing the use of  $D(n)$  from a table rather than an analytical formula, but still overcoming the divergence at  $t=0$ .

The self-consistent equation (5) was then solved by plotting the function  $\Delta n(t)$  from Eq. (10) on the same graph as the straight line  $\Delta n = (\Omega/2\pi)t$ . The coordinates of the crossing point give the localization time  $t_B$  and length  $\Delta n_B$ . Such a plot is given in Fig. 2. One can see that  $\Delta n(t)$  initially follows the behavior expected for constant  $D(n)$ , namely  $\Delta n(t) = (Dt)^{1/2}$ , and so is always initially higher than the straight line  $\Delta n = (\Omega/2\pi)t$ , then curves over to meet it at the localization time  $t_B$ . Then, at larger times, it no longer follows the constant  $D(n)$  behavior, and in fact even curves upward. At lower ac field strength ( $F_{ac}$ ),  $D(n)$  is smaller [see Eq. (8)], the curve is lower, and  $t_B$  is shorter. At higher  $F_{ac}$ ,  $D(n)$  is larger, the curve is higher, and  $t_B$  is longer.

If the ac field strength  $F_{ac}$  is high enough, then the curve of Fig. 2 curves up before it meets the straight line, there is no  $t_B$ , and therefore no localization. The atom ionizes, and delocalization has occurred. So one varies  $F_{ac}$ , and the value  $F_d$  for which the two curves first stop meeting is defined as the delocalization threshold. The value of  $F_d$  is expected to be the threshold in ac field strength of ionization due to chaos.

Solving Eq. (6) involves exactly the same procedure, except that the function  $\Delta n(t)$  is compared with the function  $[n_{orbit}(t) - n_0]$ , instead of the straight line

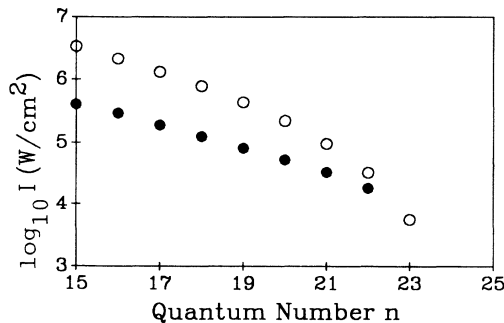


FIG. 6. Delocalization and  $n$ -mixing thresholds  $I_{dhf}$  (open circles) and  $I_{dn1hf}$  (solid circles) for the  $F_{dc} = -2$  kV/cm case.

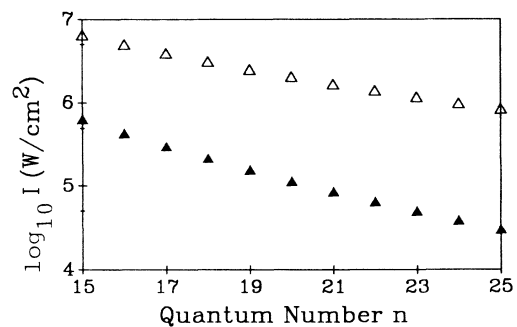


FIG. 8. Thresholds  $I_{dhf}$  (open triangles) and  $I_{dn1hf}$  (solid triangles) for the  $F_{dc}=0$  case.

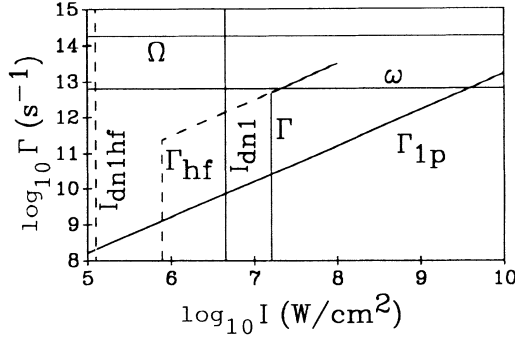


FIG. 9. Comparison of ionization rates  $\Gamma$ ,  $\Gamma_{hf}$ ,  $\Gamma_{1p}$ , external angular frequency  $\Omega$ , characteristic atomic angular frequency  $\omega$ , and thresholds  $I_{dn1}$  and  $I_{dn1hf}$  for the  $F_{dc} = -2$  kV/cm case, with  $n_0 = 18$ .

$\Delta n = (\Omega/2\pi)t$ . This gives rise to another delocalization threshold  $F_{dhf}$  corresponding to the high-frequency hypothesis that led to Eq. (6). Comparison of  $F_d$  and  $F_{dhf}$  with experiment may allow one to determine which interpretation of the localization idea is correct in the high-frequency case, which would further illuminate the physics underlying the localization effect. Likewise, one can solve for  $F_{dn1}$ , the threshold of  $\Delta n_B = 1$ ,  $n$  mixing, using the same method with either of the Eqs. (5) and (6), giving  $F_{dn1}$  and  $F_{dn1hf}$ , respectively. One simply reads out the  $y$  coordinate  $\Delta n_B$  of the point at which the appropriate curves cross, and adjusts  $F_{ac}$  to make it equal to one. Under this condition,  $F_{ac} = F_{dn1(hf)}$ .

We define another threshold of ionization which may be useful in interpreting experiments, in addition to the delocalization threshold. Define the sign of the dc field so that positive dc fields are clamping fields, giving a one-dimensional potential well, while negative dc fields are unclamping fields, giving a potential barrier.<sup>10</sup> Then for positive dc fields, the one-dimensional atom can never ionize, because it has an infinite potential well. However, for the real three-dimensional hydrogen atom, there is another threshold of ionization  $F_{E0}$  [or  $F_{E0hf}$  using Eq. (6)], which is reached when  $\Delta n_B$  reaches the energy of the zero-field ionization threshold ( $E=0$ , which occurs at

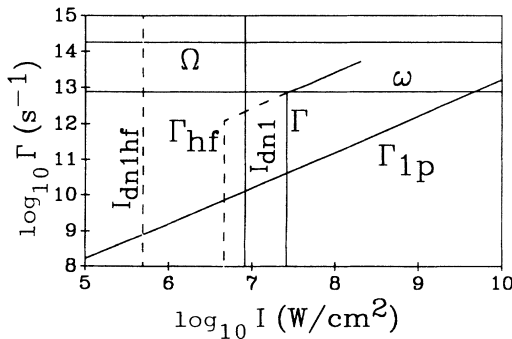


FIG. 10. Comparison of ionization rates  $\Gamma$ ,  $\Gamma_{hf}$ ,  $\Gamma_{1p}$ , external angular frequency  $\Omega$ , characteristic atomic angular frequency  $\omega$ , and thresholds  $I_{dn1}$  and  $I_{dn1hf}$  for the  $F_{dc} = 10$  kV/cm case, with  $n_0 = 18$ .

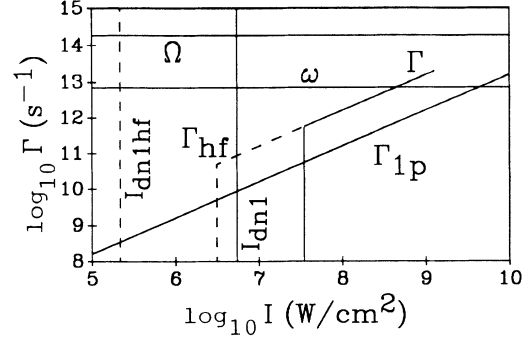


FIG. 11. Comparison of ionization rates  $\Gamma$ ,  $\Gamma_{hf}$ ,  $\Gamma_{1p}$ , external angular frequency  $\Omega$ , characteristic atomic angular frequency  $\omega$ , and thresholds  $I_{dn1}$  and  $I_{dn1hf}$  for the  $F_{dc} = 0$  case, with  $n_0 = 18$ .

$n = 22$  for 10 kV/cm), where a physical atom, approximating a one-dimensional atom, will ionize quickly due to its motion in the additional dimension which has been ignored in the one-dimensional approximation.<sup>24,25</sup>

Figures 3–5 give numerical results for the relevant thresholds for the hydrogen atom in dc fields of  $-2$ ,  $10$ , and  $0$  kV/cm, calculated from Eq. (5), and using an ac frequency of  $2.82 \times 10^{13}$  Hz, equivalent to the  $10.6 \mu\text{m}$  radiation from a  $\text{CO}_2$  laser. The value of the thresholds is given in terms of the intensity  $I$  of the  $\text{CO}_2$  laser radiation, with  $I = F_{ac}^2 / 2\mu_0 c$ , instead of ac field strength  $F_{ac}$ , so we have  $I_d$ ,  $I_{dn1}$ , and  $I_{E0}$ , corresponding to  $F_d$ ,  $F_{dn1}$ , and  $F_{E0}$ , respectively. One sees that the thresholds are similar, and that they decrease with  $n$ , the  $-2$  kV/cm thresholds decreasing sharply as they get closer to  $n = 24$ , the ionization threshold. Figures 6–8 give the same thresholds calculated from Eq. (6). One can see that these thresholds exhibit a similar behavior but are lower by about an order of magnitude, making the principles leading to Eqs. (5) and (6) clearly distinguishable by experiment. The range of intensities needed to study these effects, about  $10^4$ – $10^8$  W/cm<sup>2</sup>, is easily available. The delocalization threshold for the  $+10$  kV/cm case is a formal threshold, included for the sake of comparison, but the “chaos to  $E=0$ ” threshold is expected to be more relevant to experimental situations.

In an experiment, ionization by chaos will have to compete with ionization by single-photon absorption in the very-high-frequency case we are studying. However, single-photon absorption will not be as strong as one might expect, because the single photon is not nearly resonant with the ionization threshold but rather reaches deep into the continuum. We present some brief comparisons of the rate of chaotic ionization as compared with one-photon ionization, which may be useful in the design of experiments.

Photoionization cross sections were not available for hydrogen in an external dc field, or for one-dimensional hydrogen. To get an idea of the order of magnitude of single-photon ionization, we used the maximum (over all values of the orbital quantum number  $l$ ) cross section for a given  $n$  in the three-dimensional zero dc field case<sup>26</sup> to estimate the one-photon ionization rate  $\Gamma_{1p}$ .

We will use the inverse of the ionization time for a typ-

ical atom as a rough estimate of the ionization rate for purposes of comparison with the one-photon process. We do not mean to imply that the chaotic ionization takes place at a constant rate, with the number of atoms ionized proportional to the duration of the laser pulse, as in the (unsaturated) single-photon ionization. For short times, there may be no chaotic ionization at all, for example. The “rates” we give for chaotic ionization are, unlike the single-photon ionization rate, rates only in the sense that, after a time equal to the inverse of the rate, but not for very long before that time, one expects most of the atoms to be ionized.

Because of a scaling relationship between  $D$  and  $t$  in Eq. (10), the rate of chaotic ionization is proportional to the diffusion parameter  $D$  which is proportional to the intensity [see Eq. (8)]. Notice that the chaotic ionization time, the timescale relevant to experiments in the delocalization regime, decreases with increasing intensity, which is opposite to the behavior of the localization time, the time scale relevant to experiments in the localization regime, which increases with increasing intensity, as discussed above.

Below the delocalization threshold, there is no ionization due to chaos, and the predicted chaotic ionization rate drops directly to zero. Above delocalization, the rate depends only on the chaotic ionization time, which is a purely classical quantity, and so the only difference between the rate  $\Gamma$ , calculated using the original interpretation of the localization idea, and the rate  $\Gamma_{\text{hf}}$ , calculated using the high-frequency interpretation, is the position of the delocalization threshold.

Due to the physical differences, the chaotic ionization rates are estimated by different criteria in the three dc field cases. For negative, or unclamping, dc fields the opposition of the external dc electric field and the internal electric field from the proton creates a potential barrier, and a classical ionization threshold at finite  $n$  ( $n = 24.07$ , for example, when the dc field is  $-2$  kV/cm). The ionization rate is just the inverse of the time it takes the diffusion (see Fig. 2) to reach the top of the potential barrier. In the other two cases, the one-dimensional atom does not ionize at finite  $n$ , and other estimates of the ionization time must be used. For positive (clamping) dc fields, the inverse of the time for the diffusion to reach the first state above  $E = 0$ , where a physical atom, only approximating a one-dimensional atom, will ionize quickly due to its motion in the additional dimension,<sup>24,25</sup> is used as an estimate of the chaos ionization rate. For zero dc field, an estimate already exists which proved successful in the low-to-medium frequency regime,<sup>2</sup>  $\Gamma_{\text{(hf)}} = D(n_0)/n_0^2$ . Physically, one might think of this as the inverse of the time it would take the diffusion to go as far as the initial quantum number (i.e.,  $\Delta n = n_0$ ), if the diffusion parameter did not change. Then, the actual increase of the diffusion parameter approximately makes up the remaining distance to ionization.

Figures 9–11 give rates and thresholds for the  $-2$ ,  $10$ , and  $0$  kV/cm cases respectively, with initial quantum number  $n_0 = 18$ . For high enough intensities so that the delocalization border has been crossed and ionization by chaos occurs, the chaotic rates are considerably higher

than the one-photon rate, showing that these effects should be observable experimentally without being at all obscured by the unavoidable presence of one-photon ionization. This is a surprising result, because in previous analyses of the hydrogen atom in an external ac field, it has been assumed<sup>2</sup> that one-photon ionization would dominate whenever the external frequency was high enough that the single photon had enough energy to ionize. There are two new circumstances in the current problem which contribute to the surprising relative strength of chaotic ionization with respect to one-photon ionization. First, the one-photon ionization is weakened by the fact that the external frequency is so high that a single photon reaches deep into the continuum, as compared to the binding energy of the initial state. This lack of near resonance with the ionization threshold suppresses the one-photon ionization. Second, the chaotic ionization rates are strongly enhanced by the addition of a dc field, as can be seen by a comparison of Figs. 9 and 10 with Fig. 11. This is because the dc field produces quick ionization once the diffusion has reached a certain degree, rather than requiring the diffusion to excite all the way to  $n = \infty$  to ionize. The strong enhancement of the chaotic ionization rates gives an example of the drastic effect of the dc field in this problem.

It is important to show that the intensity at which chaos spectral effects are predicted to appear is not so high that the lifetime of the atom with respect to one-photon ionization is short enough to change the original spectrum of the atom into a featureless continuum which will not be able to exhibit spectral changes. If the lifetime of the atom is short compared to its characteristic frequency  $\omega$ , then the width of its states in energy will be larger than their separation, and individual states will not be distinguishable as such any more. Figures 9–11 show that the proposed threshold of spectral effects (or “chaos  $n$  mixing”) is always much lower than the intensity at which the one-photon ionization becomes faster than the characteristic atomic frequency  $\omega$ , and therefore the spectrum will not be all washed out at the point at which these spectral effects are expected to occur. This is true whether the original interpretation of delocalization or the high-frequency interpretation is used.

The longest chaotic ionization time, from the inverse of the rates given in Figs. 9–11, is on the order of 20 ps. This is about 500 periods of the oscillatory field. For laser pulses shorter than this, one might not be able to tell if the chaotic diffusion stops because of the finite time of the pulse or the localization effect, and so pulses longer than 20 ps are desirable for experimental tests of the localization effect. Laser pulses longer than this (the usual case) are effectively infinite as far as localization is concerned, for almost all the atoms are either ionized (at higher intensities) or localized (at lower intensities) by the end of the pulse, which allows a clear test of the localization hypothesis. This is, of course, not a difficulty in the case of CO<sub>2</sub> laser pulses. However, microwave experiments have much longer ionization and localization times, and short pulse length has been a difficulty in testing the localization theory by means of microwave experiments. There are two advantages to using the very-



high-frequency regime considered here as opposed to the microwave regime to ensure the pulse duration is long enough to test localization theory. First, the cycle time of the external field is much shorter in the very-high-frequency case, which immediately gives a faster time scale to the process. The second advantage is more subtle, but very important. In the very-high-frequency case, there is a very large gap between the classical chaos threshold and the threshold of delocalization, as shown in Fig. 1. In contrast, when the external and atomic frequencies are of the same order of magnitude, as they are in current microwave experiments, delocalization occurs only slightly above the classical chaos threshold. Naturally, the ionization due to chaos will be much faster at the delocalization threshold if it is many orders of magnitude higher than the classical chaos threshold than if it is barely greater than the classical chaos threshold. But the large difference between the classical chaos and delocalization thresholds in the very-high-frequency case is also useful for a more basic reason. This gap between the classical chaos and delocalization thresholds is, in other words, a large region of localized chaos. Since we are interested in studying the quantum localization of classical chaos, a large region where this occurs is desirable. An example: If one measures the final-state distributions of the atoms after exposure to a strong ac field pulse, as in the microwave experiments,<sup>16</sup> one can study the localization length at intensities below the delocalization threshold. Here the relevant time scale is the localization time  $t_B$ , and as long as the pulse duration is longer than  $t_B$ , one can be sure the effect is really localization as opposed to just a finite pulse length effect. Since  $t_B$  is typically shorter than the chaotic ionization time, this is a less stringent condition, and the pulses can be shorter in this type of experiment. In the recent microwave experiment, the experimental pulse time, which was 7.5 ns, was not significantly longer than even the localization time  $t_B$ .<sup>8</sup> In this situation, the classical and quantum distribution lengths ( $\Delta n$ ) are comparable, and it was not possible to conclude on the basis of experiment alone that the localization actually occurs.<sup>16</sup> Here is a further advantage of the very-high-frequency regime, for a final-state distribution type of experiment: Recall that  $t_B$  decreases as the ac field strength (or intensity) decreases. In the very-high-frequency regime, with its very large region of localized chaos, if the pulse duration is not significantly longer than  $t_B$ , we can simply turn down the intensity and reduce  $t_B$ . The large region of localized chaos ensures that we can do this without leaving the region of classical chaos. In contrast, if we want to do this in the regime where atomic and external frequencies are of the same or-

der of magnitude, the region of localized chaos is so small that any reduction in the intensity means the system is no longer chaotic and so there is no localization anyway. This is the difficulty that was experienced in the attempt to verify localization theory using microwave experiments.

Even though the chaotic ionization rates are higher than the one-photon rates, it may not be enough experimentally, if the one-photon ionization is already saturated by the time the chaos appears, and so no more ions can be created by chaos effects. There are two ways around this problem. First, one could, instead of measuring the number of ions produced by a laser pulse, measure the spectral structure of the atom in the presence of the ac field in the vicinity of  $n = 18$ , and use the width of the lines observed to obtain the ionization rates. This would also allow one to test the proposed threshold for the appearance of spectral structure, and would avoid the effects of turning on and off the pulse, which were not included in these calculations. Second, if one wants to measure the rate by counting the number of ions and wants to include the finite pulse effects, one could use short pulses. Figures 9–11 indicate that pulses shorter than 20–1000 ps, depending on which lines on which figure are followed, would be necessary to investigate this effect by measuring the number of ions without saturating the one-photon ionization.

In conclusion, we have applied the localization theory to the case of a hydrogen atom in an external dc field, which is important for comparison of experiment with one-dimensional calculations. The dc field has a drastic effect on the delocalization threshold, sometimes more than an order of magnitude. We have presented an extension of the localization theory to the high-frequency regime, taking into account the changes in the nature of the frequency spectrum which accompany the high-frequency case, and compared the resulting delocalization thresholds with those computed from a straightforward extrapolation of the delocalization equations which have been successful in low-to-medium frequency cases. We have introduced a threshold criterion for the appearance of large spectral effects due to localized chaos, and computed its value. These calculations show that an interesting, experimentally accessible regime is achievable for the investigation of the relevance of classical chaos to quantum systems with very-high-frequency driving forces. These predictions can be tested, and experimental confirmation of one of the extensions to the delocalization theory presented here would be an impressive demonstration of the breadth of applicability of that theory.

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