Microwave Ionization of Highly Excited Hydrogen Atoms: A Test of the Correspondence Principle

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For the parameters accessible in current experiments, the quantum suppression of classical chaos arises from a reduction in the effective number of coupled states. A simple theory using conventional quantum methods provides a good description of the dynamics near the threshold fields for classical chaos and recovers the classical, Fokker-Planck equation for large fields where many states are coupled.

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Recent experimental[1,2] and theoretical[2-7] studies of the ionization of highly excited hydrogen atoms by strong microwave fields have generated great interest because the classical description of the nonlinear electron dynamics exhibits a transition from regular behavior to chaos when the microwave field exceeds a sharp threshold. Since the regular orbits remain bound while the chaotic orbits ionize, the theory predicts a clear experimental signature for the manifestation of classical chaos in this simple quantum system. In fact, comparisons of the measurements by Koch *et al.*[1] of the threshold fields for the onset of ionization for states with principal quantum numbers ranging from $n_0=32$ to 90 in a 9.92-GHz field show excellent agreement with the classical calculations for the onset of chaotic ionization (see Fig. 1). Although quantum mechanics appears to be incapable of exhibiting the dynamical instability which defines classical chaos, the linear quantum dynamics can successfully mimic the onset of classical chaos for these highly excited states, which provides a strong confirmation of Bohr's correspondence principle. However, these comparisons have been restricted to low scaled frequencies, $\Omega_0=n_0^3\times\Omega<1$ (a.u.), where one or more photons must be absorbed to reach the next level and 45 or more photons are required to ionize. But Casati, Chirikov, Shepelyansky, and Guarneri (CCSG)[5] have suggested that if the experiments were to increase the scaled frequency to $\Omega_0>1$, then quantum interference effects (related to Anderson localization in solid-state physics[8]) would suppress the chaotic ionization. Since $\Omega_0>1$ can be achieved by either increasing the frequency or n_0, their results appear to predict a dramatic failure of the correspondence principle with *increasing quantum number.*

In this Letter we present a novel analytical description of the quantum dynamics for $\Omega_0>1$, using conventional methods of quantum theory, which extends and clarifies the localization theory of CCSG[5,9] and shows that their theory is only applicable to much larger quantum states than those accessible in current experiments. In particular, our analytical and numerical calculations for a 1D model of the experiment (which has been very successful for $\Omega_0\lesssim 1$) indicate that, for microwave fields near the threshold for classical chaos in the high-frequency experiments of Koch at 36 GHz, the quantum dynamics is dominated by a sequence of single-photon transitions which only couple relatively few, near-resonant states. As a consequence, the correspondence between the classical and quantum descriptions breaks down, despite the large quantum numbers, because of the low density of coupled states.

Our classical calculations[4,10] for the onset of diffusive

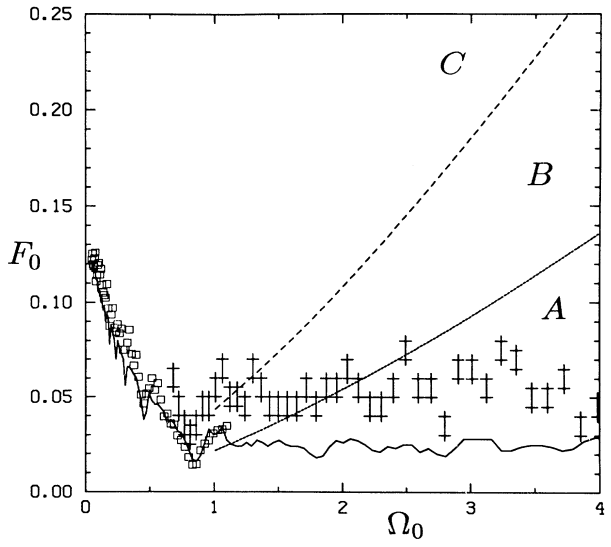

FIG. 1. Comparison of the scaled threshold fields for ionization as functions of scaled frequency (1) measured in the 9.92-GHz experiments of Koch *et al.* for $n_0=32$–90 (squares), (2) determined by the classical theory for the onset of chaos (heavy solid line), and (3) calculated from the solution of the time-dependent Schrödinger equation for $n_0=50$–90 at 36 GHz with $n_c=95$ (crosses). The boundaries corresponding to $a=1$ (heavy dotted line) and $a=2$ (heavy dashed line) are also displayed which roughly divide the parameter space into three regions described by (A) the sequential two-level Rabi theory, (B) the truncated basis of near-resonant states, and (C) the Fokker-Planck equation.

ionization predict threshold fields (shown in Fig. 1) which remain relatively constant between $F_0=n_0^4F=0.02$ and 0.03 from $\Omega_0=1.0$ to 4.0. In contrast, our quantum calculations[10] with 100 to 200 bound-state basis functions and 200 discretized continuum states show that the perturbed wave packet rapidly evolves to a steady-state distribution over the bound states with negligible ($\ll 1\%$) excitation to the continuum even at fields a factor of 2 or 3 higher than the classical thresholds.

For example, Fig. 2 shows typical results for the 1D quantum calculations for $n_0=80$ exposed to a microwave field with a scaled frequency of $\Omega_0=2.8$ which was turned on slowly over twenty periods, remained at full power for twenty periods, and then was turned off slowly over twenty periods. [If we extend the calculation to 100 periods of the peak field, the location and the approximate heights of the narrow peaks (which are accentuated by the slow turn on and turn off) are unchanged.] Although $F_0=0.03$ is greater than the classical threshold for the onset of chaotic ionization, the quantum distribution in Fig. 2 remains localized near the initial state with an exponentially decreasing series of sharp, evenly spaced peaks which are one-photon energy apart. At $F_0=0.05$ the distribution is still dominated by narrow "photon" peaks, which span only one or two unperturbed states, but the level of excitation has increased. Although our 1D calculations show little true ionization on the time scales of the experiment, the measurement process involves small static fields which counts all excitation above a critical n_c as ionization. Numerical estimates of the threshold fields for 5%–10% excitation above $n_c=95$ (which is the approximate n_c for Koch's "quenching" experiments) using the final distributions for $n_0=50$ to 90 in 36-GHz fields are shown in Fig. 1.

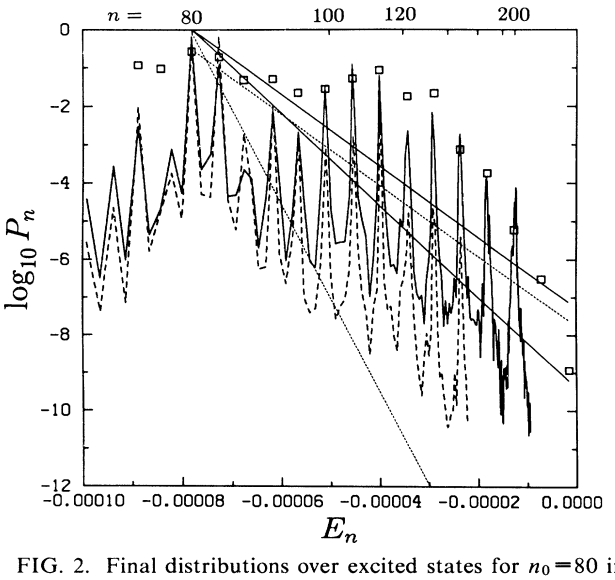

FIG. 2. Final distributions over excited states for $n_0=80$ in a 36-GHz field ($\Omega_0=2.8$) at $F_0=0.03$ (dashed curve) and $F_0=0.05$ (solid curve) corresponding to $a=0.36$ and 0.59, respectively, are compared with the exponential decay rates predicted by the two-level theory (light solid lines) and the localization theory (light dotted lines). The squares show the results of the truncated-basis calculations (Ref. 11).

These results confirm that the classical and quantum dynamics are very different at high scaled frequencies; however, a close examination of the distributions in Fig. 2 reveals that the quantum mechanism for the suppression of the chaotic ionization is very different from that invoked by CCSG.[5,9] In particular, their localization theory[9] predicts that the distribution should consist of a sequence of broad peaks which decay exponentially in energy as $f_E \simeq \exp(-2N_\phi/l_\phi)$, where N_ϕ is the number of photon energies ($E_\phi=\Omega$) from the initial state and $l_\phi \simeq 3.33F^2/\Omega^{10/3}$ is the "photonic locatization length." Then, by requiring that l_ϕ be comparable to the number of photons required to reach the continuum, $N_I=n_0/2\Omega_0$, they derive an estimate for the "quantum delocalization border" for the onset of diffusive ionization given by

$$F_D \simeq \Omega_0^{7/6}/(6.7n_0)^{1/2}, \tag{1}$$

which is much larger than the classical thresholds shown in Fig. 1.

However, the key assumptions of their theory, that there are a large number of photon peaks with many states under each peak, are simply not valid for the quantum states which are accessible in current experiments. Since we find that the quantum dynamics is dominated by relatively few resonant states, we have developed an alternative theory which provides new estimates for the width of the quantum distribution as well as a necessary condition for diffusive ionization. Our analysis begins with the important observation[9] that for high scaled frequencies the dipole matrix element for resonant one-photon transitions[4,12] between states n and m with $E_n-E_m \simeq \Omega$,

$$x_{nm} \simeq -0.411(nm)^{-3/2}\Omega^{-5/3} \sim -0.411n^{-3}\Omega^{-5/3}, \tag{2}$$

$$\text{for } n,m \gg 1, \quad |n-m| \ll n,$$

decreases rapidly with increasing quantum number. If we restrict our attention to a pair of consecutive resonant states n_i and $n_{i+1}=[n_i/(1-2\Omega n_i^2)^{1/2}]$ (where $[\cdots]$ denotes nearest integer), then the Rabi theory[13] predicts that starting from state n_i the probability of occupying state n_{i+1} will saturate to a time-averaged value of

$$P_{i+1}=\tfrac{1}{2}W^2/(W^2+\Delta^2), \tag{3}$$

where $W=|x_{n_{i+1}n_i}F|$ is the Rabi width and $\Delta=E_{n_{i+1}}-E_{n_i}-\Omega$ is the detuning from resonance.

For each pair of states the size of the detuning will vary from 0 to the maximum detuning $\Delta_m \approx 1/2n_{i+1}^3$ given by $\frac{1}{2}$ the separation of unperturbed states. If we

introduce a key parameter

$$a = 0.822F/\Omega^{5/3} = 0.822 n_0 F_0/\Omega_0^{5/3}, \quad (4)$$

which is the ratio of the Rabi width to the maximum detuning, then we can calculate a typical value for the time-averaged probability of occupying the excited state by averaging Eq. (3) over the possible detunings giving[14]

$$\bar{P}_{i+1} = \bar{P} = \tfrac{1}{2} a \tan^{-1}(1/a). \quad (5)$$

Since both the Rabi width and the separation of unperturbed levels decrease as $1/n_i^3{}_{+1}$, the parameter a and the mean excitation probability $\bar{P}$ are independent of n_{i+1}. Therefore, when a and $\bar{P}$ are small, we can apply the same two-level analysis to each pair of resonant states, starting from the initial state n_0. This simple theory predicts that the wave function will evolve to a steady-state distribution of narrow peaks decreasing exponentially with a decay length[15] $l_R = -2/\ln(\bar{P})$ which agrees well with both of the distributions displayed in Fig. 2. Moreover, for values of $a < 1$, the Rabi width for each resonance is much less than the local level spacings, so that each peak will be dominated by a *single* state.

Figure 3 shows a comparison of the peak-to-peak decay factors, $\bar{P}$ and $\exp(-2/l_\phi)$, predicted by the two theories, along with the decay factors determined by fitting an exponential to the peaks of saturated distributions, like those in Fig. 2. (The associated error bars were estimated by the range of reasonable fits to the data.) These comparisons show that for small values of a, the CCSG theory greatly underestimates the extent of the excitation to higher levels.

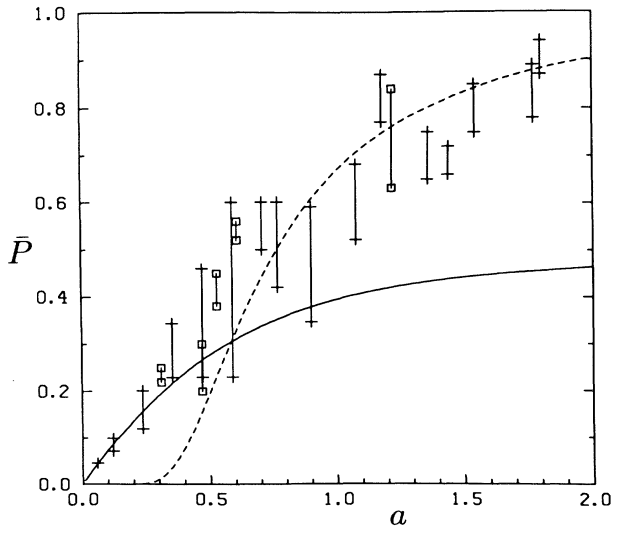

FIG. 3. Comparison of the peak-to-peak decay factors predicted by the two-level theory $\bar{P}$ (solid curve) and the localization theory $\exp(-2/l_\phi)$ (dashed curve) as a function of a with the results from numerical calculations at 36 GHz (crosses) and from some of the published distributions of CCSG (Ref. 9) (squares).

As a increases, the height of each succeeding photon peak increases and the sequential two-level theory breaks down. For values of a between 0.5 and 2 the actual decay factor rises rapidly towards 1.0. (Remarkably, the CCSG theory shows fair agreement with the numerical results despite the fact that the Rabi widths near resonance span only one or two states.) In this parameter regime, where the narrow photon peaks are all strongly coupled, the quantum dynamics can be well described by numerical calculations which keep only the resonant states.[11] For example, Fig. 2 displays a comparison of the results using only 17 resonant states with the full calculation for $F_0 = 0.5$ with 150 basis states.

Finally, for $a \gg 2$ the Rabi width spans many states and both the sequential two-level and truncated-basis theories break down. However, the high density of coupled states suggests another approximation. If we assume that the many states contained in each peak form a quasicontinuum, then the coupling from one peak to the next can be estimated using the Fermi "golden rule."[13] Then, including only the one-photon processes which couple adjacent peaks, we can write down a set of master equations for the transport of probability between the photon resonances,[14]

$$dp_i/dt = -(\alpha_{ii+1} + \beta_{ii-1})p_i + \alpha_{i-1i}p_{i-1} + \beta_{i+1i}p_{i+1}, \quad (6)$$

where p_i is the integrated probability associated with the ith photon peak from the initial state and $\alpha_{ii+1} = \frac{1}{2} \times \pi |x_{ii+1}|^2 F^2 \rho_{i+1}$ and $\beta_{ii-1} = \frac{1}{2}\pi |x_{ii-1}|^2 F^2 \rho_{i-1}$ are the one-photon excitation and de-excitation rates.

For large quantum numbers the density of states, $\rho_{i\pm1} = n_i^3{}_{\pm1}$, cancels the dependence on the final state in the square of the dipole matrix elements, so that only the n_i dependence remains in $\alpha_{ii+1} = \beta_{ii-1} \equiv \gamma_i$. Then, if there are a large number of photon peaks, $N_I \gg 1$, and p_i varies slowly with i, we can approximate the master equation with a diffusion equation,

$$\frac{dp_i}{dt} = \frac{d^2}{di^2}(\gamma_i p_i), \quad (7)$$

by expanding $p_{i\pm1}$ and $\gamma_{i\pm1}$ in Taylor series to second order. Moreover, if we change variables in Eq. (7) from photon number i to quantum number n_i, using the relation $i = (E_i - E_0)/\Omega$, then we recover a Fokker-Planck equation for the distribution, $f(n_i) = p_i di/dn_i = p_i/n_i^3\Omega$,

$$\frac{df(n_i)}{dt} = \frac{d}{dn_i}\left[D(n_i)\frac{d}{dn_i}f(n_i)\right], \quad (8)$$

with the classical diffusion coefficient,[3]

$$D \simeq 0.27 F^2 n^3/\Omega^{4/3}, \quad (9)$$

which is expected from the correspondence principle.[16]

This derivation of the diffusion equation for the quantum probability fills in some missing arguments in

CCSG's theory for the exponential photonic localization[9] which assumed that the evolution of the quantum probabilities would be determined by a random walk between the photon peaks with an effective rate of diffusion[17] given by Eq. (9). In particular, since this diffusive description is only valid when $a \gg 2$, this condition leads to a new field criterion

$$F_0 \gg F_R \simeq 2.4\Omega_0^{5/3}/n_0, \tag{10}$$

which must be satisfied for the quantum dynamics to exhibit diffusive behavior which mimics the classical chaotic dynamics.[18] However, because of the different frequency dependence of F_D and F_R the condition for the application of the CCSG theory, Eq. (10), can be more stringent than the delocalization border predicted by the theory, Eq. (2). In particular, if we consider the ratio $R = F_D/F_R \simeq 0.23 N_I^{1/2}$, then the CCSG theory will only be applicable when $R \gg 1$ or the number of photon peaks between the initial state and the continuum is large, $N_I \gg 19$.

Since the ionization of $n_0 = 72$ to 90 in a 36-GHz field, corresponding to scaled frequencies from $\Omega_0 = 2.0$ to 4.0, requires between $N_I = 18$ to 12 photons, it appears that the CCSG theory will not be relevant to the interpretation of the current experiments.[19] If we assume that $\Omega_0 = 2.0$ is large enough for the high-frequency theory to be valid, then, to achieve a ratio $R = 2$, the experiments must study the ionization of initial states with $n_0 \approx 300$ in 500-MHz fields.

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[14]These approximations are well known in molecular physics. See, for example, M. Quack, Adv. Chem. Phys. **50**, 395 (1982).

[15]We note that this new localization regime corresponds to the "strong disorder" Anderson localization case in solid-state physics, where $\delta = 1/a$ is the disorder parameter, while the CCSG localization theory corresponds to the "weak disorder" limit $\delta \ll 1$.

[16]Of course, for very large values of a neither the classical nor the quantum dynamics will be properly described as a diffusive process.

[17]The same assumptions were used in N. B. Delone, B. A. Zon, and V. P. Krainov, Zh. Eksp. Teor. Fiz. **75**, 445 (1978) [Sov. Phys. JETP **48**, 223 (1978)] to incorrectly derive a Fokker-Planck equation for diffusion in energy rather than action for this problem.

[18]A similar criterion for the validity of the diffusive description of the quantum standard map is mentioned in D. L. Shepelyansky, Physica (Amsterdam) **28D**, 103 (1987).

[19]The same conclusion was reached in Ref. 15 using semiclassical arguments.