

Exponential photonic localization for the hydrogen atom in a monochromatic field

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We present theoretical and numerical results showing that the probability distribution on energy levels of highly excited hydrogen atoms produced by a monochromatic field should be exponentially localized in the number of absorbed photons. These results allow for a new interpretation of underthreshold ionization and also for an estimate of the ionization rate.

The strong underthreshold ionization observed in highly excited hydrogen atoms in a microwave field¹⁻⁶ has been related to the onset of chaotic motion in the classical atom and, indeed, good agreement has been found between experimental data and numerical results of computer simulations of classical dynamics.³ At the same time, however, quantum interference effects may place strong limitations on chaotic motion. This fact was numerically demonstrated and theoretically justified on simple models⁷⁻¹⁰ for which, in contrast to the unlimited diffusion taking place in classical action space, the quantum distribution f_n over unperturbed levels stays localized, with a localization length l that, under semiclassical conditions, is roughly equal to the classical diffusion coefficient. If the latter is constant, then one has exponential localization, i.e., the time-averaged steady-state distribution is approximately $\bar{f}_n \propto \exp(-2|n - n_0|/l)$, n_0 being the initially excited level. This phenomenon of "quantum limitation of classical chaos" was then numerically detected even in the one-dimensional hydrogen atom in a monochromatic electric field;^{11,12} the important difference from previous models being that classical diffusion coefficient increases here with n . Because of this peculiarity, under appropriate conditions delocalization and unlimited diffusion close to the classical one may occur, thus explaining the observed agreement between experiments and classical computations. However, the nonconstancy of the localization length in n did not allow for a theoretical prediction for the form of the steady-state distribution in the localized case. As a matter of fact, this distribution displays, on high- n values, a characteristic peak structure, produced by multiphoton transitions, the explanation of which seemed out of the reach of localization theory. This latter theory seemed therefore unable to achieve a complete description of the excitation process, and to determine ionization rates.

In this paper, we show that localization theory can actually be modified, so as to include these essential details of the excitation process. The basic result will be that an approximate exponential distribution is again obtained by plotting the distribution against the number of absorbed photons (and not, as previously, against the state number n). We shall see that numerical data from computer

simulations of the quantum H atom nicely confirm our theoretical predictions.

We begin by describing a general method for determining the localization length. By applying this method to the hydrogen atom, we shall show that the localization length in the number of absorbed photons is constant, so that the usual picture of exponential localization applies.

Let us consider a quantum system with a density of unperturbed energy levels ρ which is acted upon by an external monochromatic perturbation with frequency ω . Then the system initially concentrated on some unperturbed level may start diffusing in energy. This may happen due to an irregular distribution of levels (such a model was used to describe the excitation of molecules in a monochromatic field by Akulin and Dikhne¹³) or also when the perturbation is strong enough to give rise to a chaotic diffusion in the classical limit.

However, quantum effects will lead this diffusion to a halt after a time $t_D \approx 4\pi^2/(\omega\Delta\sigma)$,^{10,14} where $\Delta\sigma$ is the average spacing of quasienergy eigenvalues significantly contributing in the evolution ($\hbar=1$ here and in the following). In estimating $\Delta\sigma$ we shall distinguish between two opposite situations, according to whether all unperturbed levels take part in the diffusion process, or not (a quantitative condition discriminating these two cases will be given below).

In the former case, $\Delta\sigma \sim 2\pi/\Delta n$,^{8,10,14} where Δn is the spread over the unperturbed levels at time t_D : $\Delta n = \rho(Dt_D)^{1/2}$, with $D \approx \langle \Delta E^2 \rangle / t_D$ the diffusion coefficient in energy. It then follows that $t_D \sim (4\pi^2/\omega^2)\rho^2 D$ and the localization length in energy, $l \approx \Delta n/\rho \approx a2\pi\rho D/\omega$.

According to numerical experiments, the numerical factor a can be given the value 1.^{10,15} The localization length in the number of absorbed photons is then $l_\phi \approx 2\pi\rho D/\omega^2$.

Now, the change in energy due to a single one-photon transition is ω , so that $D = 2\omega^2 W$, where W is the one-photon transition rate. According to Fermi's "golden rule" $W = (\pi/2) |\mu_{E,E+\omega}|^2 \varepsilon^2 \rho$, where $\mu_{E,E+\omega}$ is the dipole matrix element, and ε is the field strength. Thus we finally get¹⁶

$$l_\phi \approx 2\pi^2 \mu^2 \varepsilon^2 \rho^2. \quad (1)$$

The assumption that all levels are involved in the

diffusion process is satisfied for $\mu\varepsilon > \omega$; indeed, $\mu\varepsilon$ is an estimate for the critical detuning from the resonant Rabi frequency.¹³ In the opposite case, $\mu\varepsilon \ll \omega$, only levels close to resonance will be excited. Then the above argument leading to (1) is not valid, and we shall instead proceed as follows. First we estimate the number of excited levels at time t_D by $\Delta n \sim \bar{N}_\phi \Delta n_\phi$ where \bar{N}_ϕ is the number of absorbed photons at time t_D and Δn_ϕ is the number of excited levels in each resonant zone. N_ϕ grows diffusively in time, and from the above-written transition rate we get $\bar{N}_\phi \sim (\pi\mu^2\varepsilon^2\rho t_D)^{1/2}$, while $\Delta n_\phi \sim \mu\varepsilon\rho$. Since all excited levels lie in zones of width $\sim \mu\varepsilon$ around resonant levels, then the quasienergy eigenvalues can also be assumed to lie in an interval $\sim \mu\varepsilon 2\pi/\omega$ in $(0, 2\pi)$. Therefore, their average spacing is $\Delta\sigma \sim 2\pi\mu\varepsilon/(\omega\Delta n)$. We now put in this expression the above estimate for Δn and substitute the resulting $\Delta\sigma$ in $t_D \sim 4\pi^2/(\omega\Delta\sigma)$; thus we finally get the same previous estimate (1) for $l_\phi \sim \bar{N}_\phi$. However, the structure of the steady-state distribution will now exhibit a chain of peaks with gaps between them. If l_ϕ is but weakly dependent on energy, the population of these peaks, numbered according to the number of photons N_ϕ will decay exponentially $\sim \exp(-2|N_\phi|/l_\phi)$.

The above results are of a general nature, and in principle they may be applied to widely different quantum systems. Let us turn now to the particular case of a one-dimensional hydrogen atom in a microwave field. We write the classical Hamiltonian for this system in action-angle variables (n, λ) and in atomic units,^{4,14,17}

$$H = -\frac{1}{2n^2} + \varepsilon n^2 \cos\omega t \left(\frac{3}{2} - 2 \sum_{s=1}^{\infty} s^{-1} J'_s(s) \cos s\lambda \right), \quad (2)$$

where J_s are Bessel functions. By using the asymptotic expansions for these functions with $s \approx \omega n^3$, $s \rightarrow \infty$ we get the semiclassical expression for the dipole matrix element¹⁸ for a one-photon transition from the initial level n :

$$\mu \approx 0.411/(\omega^{5/3} n^3). \quad (3)$$

On the other hand, since $\rho = n^3$, from (1) and (3) we obtain

$$l_\phi \approx 3.33\varepsilon^2\omega^{-10/3}. \quad (4)$$

Therefore, the localization length in the number of absorbed photons turns out to be independent of energy.

Notice that on multiplying (4) by ωn^3 , i.e., the number of unperturbed levels in a one-photon interval of energy, we obtain the previously derived^{12,14} value of the localization length [Eq. (11) of Ref. 14], with a slight difference in the numerical factor. As explained in Ref. 14 this difference is due to the particular choice of a numerical factor in the classical diffusion coefficient made in Ref. 12. We wish to emphasize, however, that the previous theory was able to justify the form of the steady-state distribution only in a restricted neighborhood of the initially excited level; instead, we have now an approximate description for the overall distribution, including its peak structure.

If the photonic localization length is large enough, then this peak structure will produce a plateau in the original distribution over unperturbed levels.^{12,14} If, moreover, l_ϕ is comparable to the number of photons required for ion-

ization $N_I = (2n_0^2\omega)^{-1}$ then strong ionization will occur. This is the delocalization phenomenon described in Refs. 12 and 14, which leads to diffusive ionization, as in the classical atom. As a matter of fact, the condition $l_\phi \approx N_I$ yields the same expression for the delocalization border as in Refs. 12 and 14 [Eq. (15) of Ref. 14].

We checked the above theoretical prediction on numerical data from extensive computer simulations of the quantum H atom that are fully reported elsewhere.¹⁴ In Fig. 1, a time-averaged distribution is plotted versus the number of photons $N_\phi = N_I - 1/(2n^2\omega)$. An approximate exponential behavior is here evident. We determined the value of l_ϕ by dividing the explored range of values of N_ϕ (to the right of zero) in one-photon intervals, and by taking the maximum of the distribution \bar{f}_n in each interval. A least-squares fit of these values, in semilogarithmic scale, with a straight line, yielded the localization length. The ratio of the l_ϕ thus obtained to the theoretical value (1) was here 1.6.

The results of several such determinations of l_ϕ for different parameter values ($1 \leq \omega_0 \leq 3$; $0.02 \leq \varepsilon_0 \leq 0.16$; $n_0 = 30, 45, 66, 100$) are shown in Fig. 2. Here $\ln \bar{f}_n$ is plotted versus the number of photons $x = 2N_\phi/l_\phi$. With this rescaling, pure exponential localization in all cases would yield the solid line.

In all but three cases considered, the theoretical value of l_ϕ was larger than one, and ε_0 was larger than the classical chaotic border $\varepsilon_c = 1/(50\omega_0^{1/3})$. The dependence of l_ϕ on ε predicted by (4) can be checked in Fig. 3; the theoretical formula correctly works in a range of 5 orders of magnitude of ε^2 .

Quite remarkably, still another independent theoretical justification for photonic exponential localization can be given. To this end, we first reduce the classical dynamics described by (2) to a two-dimensional mapping and then we quantize this mapping; in this way, we obtain a new

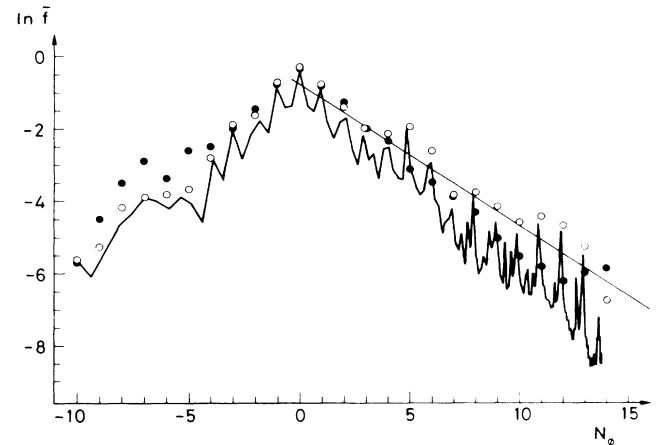


FIG. 1. The probability distribution, averaged from 80 to 120 periods vs the number of photons $N_\phi = N_I - 1/(2n^2\omega)$. Here $n_0 = 100$, $\varepsilon_0 = \varepsilon n_0^3 = 0.04$, $\omega_0 = \omega n_0^3 = 3$. For each integer value of N_ϕ , open circles indicate the probability in the interval $N_\phi - \frac{1}{2}$, $N_\phi + \frac{1}{2}$. The straight line is the result of a least-squares fitting of the peak's value. Filled circles were obtained by iterating the quantum map (6).

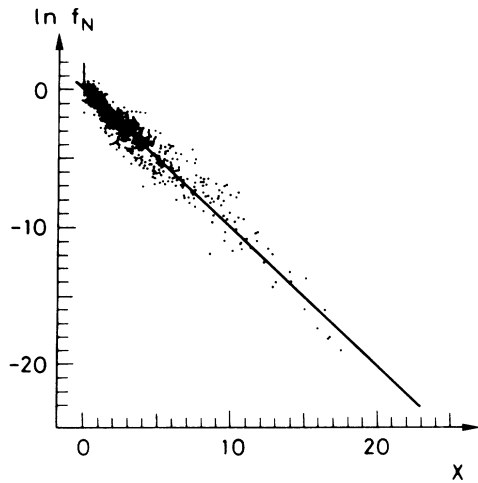


FIG. 2. $\ln f_N$ vs $x = 2N_\phi/l_\phi$. Here l_ϕ is the experimental value, obtained by least-squares fits on 41 distributions as described in Fig. 1, for different values of ϵ_0 , ω_0 , and n_0 . The constant parts in $\ln f_N$ have been subtracted, so that perfect exponential localization would correspond to $\ln f_N = -x$, which is also drawn in the figure (full line).

approximate formulation of the quantum dynamics of the H atom. In order to derive the classical map, we integrate the Hamilton equations of motion over an orbital period of the electron, substituting the unperturbed motion in the field-dependent terms and keeping just the resonant term $s \approx \omega n$.³

We then find that the variables N (energy divided by ω) and $\phi = \omega t - s\lambda$ during an orbital period change ac-

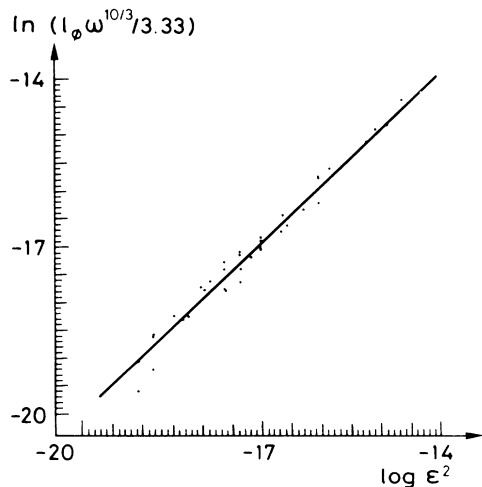


FIG. 3. Plot of the logarithm of the rescaled experimental localization length $\ln(l_\phi \omega^{10}/3.33)$ vs $\ln \epsilon^2$. The solid line gives the theoretical dependence (4). The points are obtained from numerical data of 38 different distributions, with $l_\phi > 1$.

cording to

$$\bar{N} = N + k \sin \phi, \quad (5)$$

$$\bar{\phi} = \phi + 2\pi\omega(-2\omega\bar{N})^{-3/2},$$

where $k = 0.822\pi\epsilon/\omega^{5/3}$. This “Kepler map” yields an approximate description of the motion of the classical electron. It is defined for all bound states ($N < 0$) but carries some of them into the positive energy region, where it is not defined. The real trajectory then goes to infinity, and its energy is determined by the last “kick” [term $k \sin \phi$ in (5)].

It is important to remark that (5) can be locally approximated by a standard map¹⁹ with parameters k , $T \approx 6\pi\omega^2 n_0^5$ and stochasticity parameter $K = k_B T = \epsilon_0/\epsilon_c$, as follows from linearization. Thus (5) shows again that global diffusion is to be expected for $K > 1$, i.e., $\epsilon_0 > \epsilon_c$.

Let us now quantize the map (5). Since this map describes an unbounded motion in ϕ under a periodic perturbation, a new integral of motion will appear (“quasi-impulse”), besides quasienergy; for a given unperturbed level n_0 , it will be just the fractional part of $N_0 = -n_0/(2\omega_0) = -N_I$. Then, putting $N_\phi = N - N_0$, we can represent N_ϕ by the operator $\hat{N}_\phi = -i\partial/\partial\phi$, with periodic boundary conditions in ϕ . The quantized version of (5) will be the following quantum map for the wave function $\psi(\phi)$:

$$\bar{\psi} = e^{-i\hat{H}_0} \hat{P} e^{-ik \cos \phi} \psi, \quad (6)$$

where

$$\hat{H}_0 = 2\pi[-2\omega(N_0 + \hat{N}_\phi)]^{-1/2},$$

and \hat{P} is the projection operator on bound states ($N_\phi < N_I$). This quantum map clearly establishes a link between the H-atom problem and the rotator problem. By using this approximate formulation of the H-atom dynamics, we obtain the following results.

(1) By iterating (6) we find the evolution of the distribution on the number of photons. Then the usual picture of exponential localization with a length $l \approx k^2/2$ (Refs. 10 and 15) should apply, were it not for the dissipation introduced by projector \hat{P} . However, if the localization length thus predicted is less than the number of photons for ionization, a quasistationary exponential distribution is reached even in the presence of \hat{P} . Then from $l \approx k^2/2$ we get the same value (4) previously derived by a quite different method.

The results of a numerical simulation of (6) are reported in Fig. 1, where the steady-state populations of \hat{N}_ϕ eigenstates (filled circles) numerically obtained from (6) are compared with the probabilities in one-photon intervals of energy gotten from the numerical simulation of the Schrödinger equation (open circles). The two sets of data are not very close, but there is an average agreement, especially in a neighborhood of the initial peak.

We recall that, according to numerical data,¹⁰ the steady-state distribution can be satisfactorily described by

the formula

$$\bar{f}_{N_\phi} \approx (1 + 2|N_\phi|/l_\phi) \exp(-2|N_\phi|/l_\phi)/(2l_\phi). \quad (7)$$

(2) The map (6) also allows for the determination of the ionization rate. This is particularly simple for one-photon ionization with $k \ll 1$. Indeed, by estimating the probability for a transition with $\Delta N_\phi = 1$ under one kick, we get the ionization rate in number of map periods $\gamma_\phi = (k/2)^2$. In real physical time, the rate is $\Gamma_\phi = \gamma_\phi (2\pi n_0^3)^{-1}$, which is the same as the standard result^{6,14} $\Gamma_\phi = 0.265 \epsilon^2 / (\omega^{10/3} n_0^3)$.

In general, in the localized regime the ionization rate should be

$$\gamma_\phi \sim \sum_{N=N_I-k}^{N_I} \bar{f}_N \sim k \bar{f}_{N_I},$$

where $N_I > l_\phi > k > 1$. In physical time, this gives

$$\Gamma_\phi \sim k f_{N_I}(k\omega)^{3/2} \sim \omega^3 \epsilon^{-3/2} n_0^{-2} \times \exp(-0.3\omega^{7/3} \epsilon^{-2} n_0^{-2}). \quad (8)$$

(3) From (6) we can also extract a prediction about the distribution in the continuous part of the spectrum. If, as in the classical case, this distribution is essentially determined by the effect of a single kick on states close to the ionization border, then from (6) we get that for $k \gg 1$ this distribution should be $\propto |J_{N_\phi}(k)|^2$, in agreement with results obtained in Ref. 20.

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