

Dynamical Localization in More Than One Dimension

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The ionization of hydrogen atoms by a microwave field is numerically studied, for the first time in a realistic picture, using a fully three-dimensional quantum approach with complete inclusion of the continuum. This provides first evidence for the persistence of dynamical localization in higher-dimensional systems. Comparison to the one-dimensional dynamics as well as to experimental results is given.

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The quantum mechanical inhibition of classical, diffusive energy transport in time-periodic Hamiltonian systems, labeled by the title "dynamical localization" because of its formal analogy to Anderson localization in disordered solids, has been a subject of intense research for more than a decade [1-9]. Careful examination of Rydberg atoms exposed to intense, monochromatic microwave fields provided experimental evidence for the appearance of this striking quantum phenomenon in higher-order multiphoton processes [3-5]. Furthermore, experiments revealed a much more detailed structure than predicted by first theoretical attempts and led therefore to more refined and sometimes controversial theoretical approaches [6-9]. Most of these models, however, either rely on three-dimensional but purely classical descriptions or, in a quantum picture, are restricted to one-dimensional representations of the hydrogen atom, incorporating the effect of the continuum partially at most. The purpose of this Letter is to provide the first numerical quantum simulations of the fully three-dimensional hydrogen atom in a linearly polarized microwave field, taking full account of the continuum. The numerical results are compared to some representative experimental data. Additionally, we supply a comparison to one-dimensional quantum calculations, as a contribution to clarifying the question of how far "dynamical localization" depends on the dimension of the classically accessible phase space. This issue is closely related to the inhibition of charge transport in disordered solids, since the strict localization of electron wave functions is known to be peculiar for the one-dimensional case, whereas in higher-dimensional systems extended states can be found [10]. For the 3D hydrogen atom in a linearly polarized microwave field (extended) phase space has dimension five. Axial symmetry preserves the angular momentum projection on the direction of the field polarization, m , and hence leaves only 2 degrees of freedom for the effective dynamics. The fifth dimension is due to the explicit time dependence of the perturbation.

As has been demonstrated by several authors [6, 8, 11-13], the adequate picture to describe quantum systems subject to time-periodic perturbations is given by Floquet theory. In order to solve the time-dependent

Schrödinger equation describing the hydrogen atom in a monochromatic, linearly polarized (along the z axis) microwave field, we make use of this formalism and transform the time-dependent Hamiltonian H into a time-independent Hamiltonian \mathcal{H} . This can be performed in a way analogous to the one described in Refs. [11] and [12], finally leading to (in the velocity gauge)

$$\mathcal{H} = \frac{\mathbf{p}^2}{2} - \frac{1}{r} + K\omega + \frac{F^2}{4\omega^2} + \frac{Fp_z}{2i\omega}(a^\dagger - a), \quad (1)$$

where F and ω are the microwave amplitude and frequency, respectively. Terms in $a^{\dagger 2}$ and in a^2 (a and a^\dagger being rising and lowering operators of the field) have been omitted, which can be formally justified by a gauge transformation resulting in the multiplication of the electron wave function by an irrelevant phase factor. K counts the number of photons of frequency ω exchanged between the atom and the field. \mathcal{H} is commonly known as the Floquet Hamiltonian corresponding to H .

As can be immediately seen from (1), the matrix representation of \mathcal{H} has block structure in the photon number K . The eigenstates $|\phi_j\rangle$ of \mathcal{H} form an orthonormal basis of the Hilbert space associated with \mathcal{H} . Their K components are in fact the Fourier amplitudes of the $2\pi/\omega$ -periodic Floquet states solving the time-dependent Schrödinger equation $i\partial_t\Psi = H\Psi$ subject to periodic boundary conditions.

Considering the time-independent part of the Floquet states, we can make use of the oscillator representation of the dynamical group $SO(4,2)$ of the hydrogen atom [14]. Expanding therefore the Fourier amplitudes of the Floquet states on a Sturmian basis, which is precisely the eigenbasis of the above-mentioned oscillator representation, we are finally provided with analytic expressions for the matrix elements and with selection rules which induce a block-banded structure of \mathcal{H} with few nonzero elements. The Sturmian basis is here particularly well suited, as it allows for a complete representation of both the discrete states and the continuum, and consequently of the ionization process, in a discrete basis. Space-time-inversion symmetry leaves the generalized parity $\Pi = (-1)^{l+K}$ (l designating the angular momentum) as the second conserved quantity of our problem (besides m).

According to Eq. (1), for vanishing field amplitude F , the spectrum of \mathcal{H} will be composed of Coulomb spectra which are shifted with respect to each other by $K\omega$, K being an integer of arbitrary sign. Thus, all the discrete eigenvalues corresponding to Rydberg series of photon order K will be imbedded into the continua of lower photon orders. As a result of the coupling with these continua, all the discrete eigenvalues turn into resonances with finite autoionizing widths [15]. A powerful tool to analyze such a situation and to obtain the real as well as the imaginary parts of the resonances is the technique of complex dilatation [16]. Studies of the hydrogen atom in a static magnetic field [14] as well as in dc and ac fields [12] have revealed the outstanding efficiency of this method. It amounts to making the replacements $\mathbf{r} \rightarrow \mathbf{r}e^{i\theta}$, $\mathbf{p} \rightarrow \mathbf{p}e^{-i\theta}$ in the Hamiltonian (1), yielding the non-Hermitian operator

$$\mathcal{H}(\theta) = \frac{\mathbf{p}^2}{2} e^{-2i\theta} - \frac{e^{-i\theta}}{r} + K\omega + \frac{F^2}{4\omega^2} + \frac{Fp_z e^{-i\theta}}{2i\omega} (a^\dagger - a), \quad (2)$$

and leaving the banded structure of \mathcal{H} unaffected. For our purposes, the key property of the rotated Hamiltonian $\mathcal{H}(\theta)$ ($0 \leq \theta \leq \pi/4$) is that the resonances of \mathcal{H} coincide with the complex eigenvalues of $\mathcal{H}(\theta)$ and that their real and imaginary parts are θ independent, provided they have been uncovered by the rotation of the continua [15]. Furthermore, we implicitly use that the resolvent operator of \mathcal{H} can be expanded onto eigenstates of the rotated Hamiltonian in a finite region near the nucleus [14]. Although this seems like a straightforward application of analyticity properties, the nonunitary character of the complex dilatation causes $\mathcal{H}(\theta)$ to be non-Hermitian, giving the expansion an unusual character. In a basis of real functions, the matrix of $\mathcal{H}(\theta)$ is complex symmetrical, and the eigenvectors $|\phi_j(\theta)\rangle$ are nonorthogonal. The left eigenvectors $|\phi_j(\theta)\rangle^\tau$ are the transposed values of $|\phi_j(\theta)\rangle$, not the Hermitian conjugates, and they are normalized so that $|\phi_j(\theta)\rangle^\tau \cdot |\phi_i(\theta)\rangle = \delta_{ji}$. This leads to the biorthogonal spectral decomposition:

$$\mathcal{H}(\theta) = \sum_j E_j(\theta) |\phi_j(\theta)\rangle \cdot |\phi_j(\theta)\rangle^\tau, \quad (3)$$

where the $E_j(\theta)$ are the complex eigenvalues of $\mathcal{H}(\theta)$.

Since experiments on microwave ionization measure ionization probabilities at a given interaction time of the atoms with the radiation field, we have to evaluate the ionization probability $P_{\text{ion}}(t)$ of an atom initially prepared in a state $|\psi_0\rangle$. We do not present here the rather technical details of the calculation, but only recall the important property of the time-evolution operator being nothing but the Laplace transform of the resolvent $[\mathcal{H}(\theta) - z]^{-1}$ (z complex valued) of $\mathcal{H}(\theta)$. Because of (3), this latter can be computed from the eigenvalues and eigenvectors of the rotated Hamiltonian. Thus, P_{ion} may

be readily obtained after the diagonalization of $\mathcal{H}(\theta)$. Since current experiments average over the phase of the microwave field experienced by different atoms, P_{ion} has yet to be averaged over one field cycle. As usual [3–5], we define the ionization threshold field $F(10\%)$ as the microwave amplitude necessary to ionize 10% of the atoms within the given interaction time. Using a finite basis for the resolution of the problem shifts the continuum to lower values in energy, roughly comparable to the experimental limitation on the distinction between very high lying bound states and the real continuum. In experiments this led to the introduction of the cut-off value n_c of the principal quantum number [3], such that excitation to $n > n_c$ is considered as ionization. In our numerical experiments we made sure that the effective continuum induced by the basis size corresponds roughly to the experimental one, taking properly into account the scaling laws (4) mentioned below. In order to display the dependence of the threshold field on the microwave frequency, we finally got to extract $F(10\%)$ from several diagonalizations of $\mathcal{H}(\theta)$, performing sweeps in F at different values of ω , and for fixed initial principal quantum number n_0 .

Our simulations have been performed for an initial value of the principal quantum number $n_0 = 23$. The diagonalization of $\mathcal{H}(\theta)$ has been performed using a stable implementation of the Lanczos algorithm. The convergence of our results has been checked by the dependence of $P_{\text{ion}}(t)$ on the size of the truncated basis (in the number of Sturmian functions as well as in the number of photon blocks), on the angle of rotation θ , and on the scaling parameter of the Sturmian basis. The 1D calculations could be performed on a Sun Sparc2 workstation spending roughly two weeks of computation time. The diagonalization of the rotated 3D Hamiltonian (2) on a CRAY2 for one pair of field parameters ω and F needs of the order of one and a half hours of CPU time. This and the limitations on central memory of current supercomputers restrict our calculations to $n_0 \leq 31$ with the current approach. However, taking advantage of the well known classical scaling properties [2] of the Hamiltonian (1) and therefore introducing scaled field amplitude F_0 , scaled frequency ω_0 , and scaled microwave interaction time t_0 according to

$$F_0 = n_0^4 F, \quad \omega_0 = n_0^3 \omega, \quad t_0 = n_0^{-3} t, \quad (4)$$

we can use the typical values of the experiments [3–5] for ω_0 . Fixing n_0 at 23 and appropriately rescaling the interaction time as prescribed in Eq. (4) we should end up with a physical situation close to the experimental one.

Figure 1 shows the 3D ionization probability of the state $|\psi_0\rangle = |n_0 = 23, l_0 = 1, m_0 = 0\rangle$ [5] as a function of the microwave interaction time t . As, in general and independent of the dimension, several Floquet states with comparable widths may be populated by the

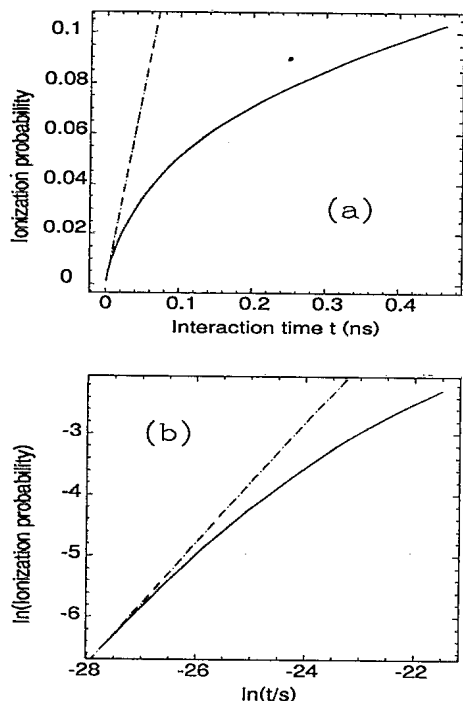


FIG. 1. Ionization probability of a 3D hydrogen atom exposed to a linearly polarized microwave field, as a function of the interaction time. The atoms were initially prepared in the state $|n_0 = 23, l_0 = 1, m_0 = 0\rangle$. The interaction time has been chosen equal to 4.64×10^{-10} s, as a consequence of Eq. (4). Scaled microwave frequency $\omega_0 = n_0^3 \omega = 1.9$, scaled microwave field $F_0 = n_0^4 F = 0.062$. (a) Linear scale. (b) Double-logarithmic plot. Comparison to the purely exponential decay to the continuum (dash-dotted lines in both figures) shows the clearly nonexponential but rather multi-algebraic time dependence, on this scale.

microwave field, the decay to the continuum is clearly seen to be nonexponential. A purely exponential decay should manifest as an approximately straight line of slope one in Fig. 1(b) (dash-dotted line), since, for $\Gamma t \ll 1$, $P_{\text{ion}}(t) = 1 - \exp(-\Gamma t) \simeq \Gamma t$ and $\ln P_{\text{ion}}(t) \simeq \ln \Gamma + \ln t$ hold. The observed slope clearly tends to lower values, as well as it does on the linear plot in Fig. 1(a), and thus rather suggests a (multi)algebraic time dependence, on these time scales. This observation should clearly affect the time dependence of $F(10\%)$ which has been investigated in Ref. [5].

Figure 2 displays the 1D ionization probability of the state $|\psi_0(t=0)\rangle = |n_0 = 23\rangle$ as a function of the scaled field, showing the typical threshold behavior as also observed in experiments. Each point of this curve was obtained from a plot like in Fig. 1(a). For the 1D simulations, we produced complete plots of this kind, at several values of ω_0 between 0.8 and 2.0, whereas in the 3D case we were restricted to the computation of P_{ion} in the vicinity of $F(10\%)$.

Summing up the 1D and 3D results in Fig. 3, we also display some typical experimental results from Ref. [3]. The most prominent feature of our results is the neat in-

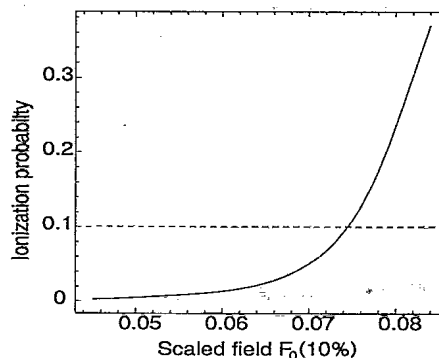


FIG. 2. Ionization probability of a one-dimensional hydrogen atom in a linearly polarized microwave field as a function of the scaled microwave amplitude $F_0 = n_0^4 F$. Initial state of the atom: $|n_0 = 23\rangle$. Microwave interaction time: 4.64×10^{-10} s. Scaled microwave frequency $\omega_0 = 2.0$. The typical threshold behavior known from experiments is clearly displayed.

crease of the ionization threshold with ω_0 , for $\omega_0 > 0.8$. This is the typical "fingerprint" of "dynamical localization" since classically $F_0(10\%)$ should monotonically decrease with ω_0 (for sufficiently long interaction times). It does obviously not depend on the dimension of the accessible phase space, for our special choice of the initial state in the 3D calculations. Comparison to the first theoretical model predicting "dynamical localization" shows that the "quantum delocalization border" derived from this simplified model [9] gives a globally astoundingly

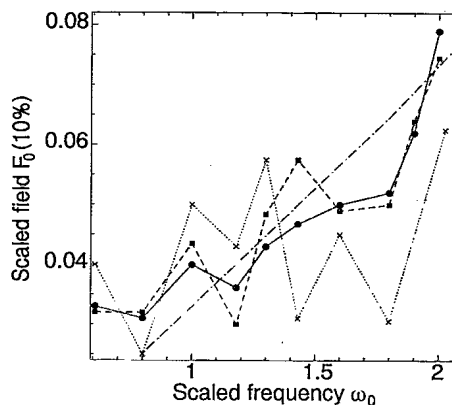


FIG. 3. Ionization of a hydrogen atom by a linearly polarized microwave field. Scaled ionization threshold field $F_0(10\%) = n_0^4 F(10\%)$ vs scaled frequency $\omega_0 = n_0^3 \omega$ for $n_0 = 23$. Crosses, dotted line: experimental values from Fig. 2(a) in Ref. [3]. The values have been chosen to display the typical local structure of $F_0(10\%)$ observed in these experiments. Squares, dashed line: 1D quantum simulations of the ionization of $|n_0 = 23\rangle$. Microwave interaction time: 4.64×10^{-10} s. Circles, solid line: 3D quantum simulations of the ionization of $|n_0 = 23, l_0 = 1, m_0 = 0\rangle$. Interaction time as for 1D. Dash-dotted line: quantum delocalization border derived in Ref. [9] [equation (A.4) with $n_0 = 62$ and cutoff $n_c = 90$].

good description of the physical process. We chose here to plot the estimations of [9] for the experimental values [3] of n_0 and n_c , since the "quantum delocalization border" lacks classical scaling. This would shift the dash-dotted line in Fig. 3 to higher values of $F_0(10\%)$, leaving its functional dependence on ω_0 essentially unchanged. It is this functional dependence which matters, as far as the phenomenon of dynamical localization is concerned.

Comparison to the experimental data shows that our rescaled results reproduce fairly well the experimental ones obtained at principal quantum numbers between 48 and 72. This gives a nice example for classical scaling in quantum mechanics and should be contrasted by the fact that the effective Planck's constant \hbar/n_0 varies roughly by a factor 3 from $n_0 = 23$ (used in the calculations) to $n_0 = 62$ (typical experimental value). Preliminary results for $n_0 = 31$ support the reliability of this (classical) scaling argument. Also the local stabilities attributed to classical resonance islands at $\omega_0 = 1.0$ and $\omega_0 = 2.0$ are clearly visible. Most local structures of the laboratory experiments, however, are not reproduced by our data, for presumably two reasons: first, the selective excitation of individual Floquet states, as a potential consequence of the switching-on and -off of the electromagnetic field in real experiments, cannot be quantitatively reproduced by the diagonalization of \mathcal{H} , since Eq. (1) implies a time-independent microwave amplitude experienced by the atoms. Second, and contrary to our choice of the initial state of the atoms being prepared in a particular, quasi-one-dimensional state with $l_0 = 1$ and $m_0 = 0$ [5], the initial n_0 -substate distribution of Ref. [3] was shown to be close to microcanonical. To model the experiments in detail therefore necessitates an ensemble average over different initial states with variable values of l_0 and m_0 . Such an investigation of the m_0 and l_0 dependence of the ionization behavior will further elucidate the competition between quantal transport in n and l . This is a subject of considerable general interest, as well as with respect to another, on a first glance rather surprising, feature of our results: 1D and 3D calculations exhibit quite good global agreement, thus justifying the one-dimensional model as a fairly good approximation for 3D atoms initially prepared in a quasi-one-dimensional state. The slightly smoother dependence of the 3D threshold on ω_0 suggests that $F_0(10\%)$ exhibits no systematic trend to higher or lower values when the dimensionality of the accessible phase space is increased.

In conclusion, we numerically simulated the time-dependent ionization process of the real 3D hydrogen atom in a microwave field, providing first theoretical ev-

idence for the persistence of dynamical localization in a realistic quantum system of two and a half effective degrees of freedom. For the quasi-one-dimensional initial states under consideration, the 1D dynamics has been shown to mimic the 3D behavior reasonably well, as far as ionization probabilities and threshold fields are concerned.

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