

Quantum Dynamics of a Circular Rydberg State in a Microwave Field

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We present the first complete quantum treatment of the ionization of a circular Rydberg state by a linearly polarized microwave field. Experimentally accessible ionization threshold fields as well as the dynamics of the resonance eigenfunctions are investigated, together with a comparison to the behavior of a quasi-one-dimensional state subject to a microwave, and of a circular state in an intense, high frequency laser field.

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The ionization of hydrogen or hydrogenlike atoms by a strong electromagnetic field represents a major topic of current research in atomic physics. Two of the most striking effects in this area are the “dynamical localization” [1–9] of Rydberg atoms in strong, linearly polarized microwave fields and the “adiabatic stabilization” [10–14]—i.e., ionization probability *decreasing* with *increasing* laser intensity—of ground state or weakly excited atoms by intense, high-frequency laser radiation. As the relation between internal (atomic) and external (field) parameters defining the problem at hand are generally quite distinct for these two schemes, also the underlying physical mechanisms dominating the ionization process are different. However, a comprehensive and sufficiently general physical picture of both domains is highly desirable and of broad interest. This paper, relying on a realistic description of the system, presents a first step in this direction and demonstrates an astoundingly simple property of the dynamics of the electronic wave functions extending over the entire parameter space. The special choice of a circular initial Rydberg state not only facilitates the identification of this general feature but also allows for a very clear distinction from pulse-induced stabilization effects [15], due to the strongly reduced density of states accessible to a circular Rydberg electron. Pulse-induced or dynamical stabilization is therefore *not* discussed in this paper.

The inhibition of diffusive ionization of Rydberg states by a linearly polarized microwave is considered as a pure quantum phenomenon, contrasting the highly nonlinear classical dynamics which should lead to ionization at lower field amplitudes than observed for the real quantum system. This quantum coherence effect has been baptized dynamical localization [1] and is hitherto restricted to atoms which are essentially concentrated along one spatial dimension, i.e., to quasi-one-dimensional Stark or low angular momentum states. Under these conditions it has been shown theoretically that, for sufficiently high frequencies, the classical diffusion in the second degree of freedom is much slower than in energy [1] and that the quantum transport leading to ionization is basically one dimensional. Whereas this one-dimensional approach has been validated by a recent *ab initio* quantum calculation [8], a rigorous quantum treatment is yet missing for fully

3D initial states of the atoms [3].

This Letter provides first complete quantum calculations modeling the ionization of a circular Rydberg state by a coherent microwave field of linear polarization. We investigate the frequency dependence of the experimentally accessible ionization thresholds, defined by those values of the microwave field ionizing 10% of the atoms during the predefined interaction time, and discuss the possibility to check our predictions by current experiments. In addition, the dynamics of the wave functions of the quantum problem are considered. Comparison to ionization thresholds and typical wave functions of a quasi-one-dimensional initial state is supplied. Finally, the comparison with the dynamics of circular states exposed to high frequency laser fields, in the domain where adiabatic stabilization is numerically observed, shows the underlying link between dynamical localization and adiabatic stabilization.

For the subsequent calculations we use the same approach as described in Ref. [8]. The Hamiltonian H describing (in the velocity gauge and using atomic units) a 3D hydrogen atom subject to a linearly polarized (along the z axis) microwave field is given by

$$H = \frac{\mathbf{p}^2}{2} - \frac{1}{r} - \frac{Fp_z \sin(\omega t)}{\omega}, \quad (1)$$

where F and ω are the field amplitude and frequency, respectively. The periodicity of H in time allows for its transformation to a time-independent Hamiltonian \mathcal{H} , as a consequence of the Floquet theorem. Exploring the complex dilation technique, we obtain ionization probabilities as well as the temporal dynamics of the resonance eigenfunctions of \mathcal{H} [14]. All ionization probabilities are averaged over one cycle of the microwave field, as they are in current laboratory experiments [3,4,6]. The amplitude is *constant* in (1), which corresponds to switching on and off the microwave radiation experienced by the atoms *adiabatically*. An estimate of the possible influence of dynamical transitions of Landau-Zener type between different eigenstates of \mathcal{H} [5], induced by the envelope of the microwave pulse in real experiments, can be realized by diagonalizing \mathcal{H} for different values of the field amplitude and adiabatically following the eigenstate which originates from the unperturbed initial state of the atom.

The value of F at which the width of this adiabatic continuation of $|n_0 l_0 m_0\rangle$ takes the value corresponding to an ionization of 10% during the given interaction time defines the threshold for adiabatic switching of the microwave.

Our simulations have been performed for an initial value of the principal quantum number $n_0 = 23$, due to the limitations on central memory of current supercomputers. However, taking advantage of the well known classical scaling properties of the Hamiltonian (1) [16], and introducing scaled field amplitude F_0 , scaled frequency ω_0 , and scaled microwave interaction time t_0 according to

$$F_0 = n_0^4 F, \quad \omega_0 = n_0^3 \omega, \quad t_0 = n_0^{-3} t, \quad (2)$$

we can use values of the scaled variables which are typical for laboratory experiments. It has been shown in [8] that this approach allows one to reproduce experimental results fairly well. Possible sources of discrepancies between laboratory experiments performed at typical values of n_0 close to 60 [3,4,6] and our numerical experiments ($n_0 = 23$) are the effective size of \hbar , together with dynamical transitions between eigenstates of \mathcal{H} . Also the preparation of the atoms in a microcanonical distribution of l_0 and m_0 over the energy shell defined by n_0 [3] cannot *a priori* be excluded from inducing local structures in the ω_0 dependence of $F_0(10\%)$. Whereas the latter point requires a more detailed study which is beyond the scope of this paper, we will give some estimate on how far the switching of the microwave field may alter the results for a circular initial state obtained from our diabatic approximation. Figure 1 shows the

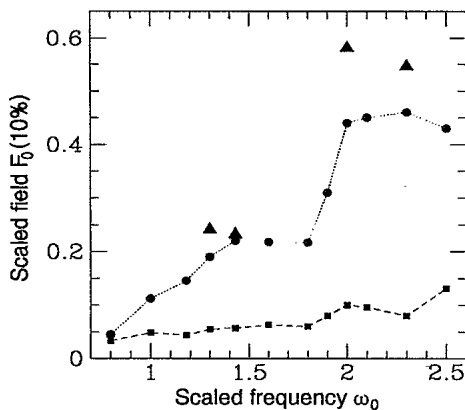


FIG. 1. Ionization of 3D atomic hydrogen by a linearly polarized microwave field. Scaled ionization threshold field $F_0(10\%) = n_0^4 F(10\%)$ vs scaled frequency $\omega_0 = n_0^3 \omega$, at fixed principal quantum number $n_0 = 23$. Microwave interaction time: 4.64×10^{-10} s. Circles, dotted line: Circular initial state $|n_0 = 23, l_0 = 22, m_0 = 22\rangle$. The pyramids indicate the threshold field for the same initial state, but for adiabatic switching of the microwave field. Squares, dashed line: extended, quasi-one-dimensional initial state $|n_0 = 23, l_0 = 1, m_0 = 0\rangle$.

scaled 10% ionization threshold field $F_0(10\%)$ for the circular state $|n_0 = 23, l_0 = 22, m_0 = 22\rangle$, in the diabatic approximation over the whole frequency range and for adiabatic switching of the microwave at four values of ω_0 . For comparison, also the threshold for the quasi-one-dimensional state $|n_0 = 23, l_0 = 1, m_0 = 0\rangle$ is plotted. The dependence of $F_0(10\%)$ on ω_0 displays a much steeper slope and a clearly increased stability towards ionization for the circular state with respect to the quasi-one-dimensional state. There is a local maximum in the vicinity of $\omega_0 = 2.0$, due to a classical resonance slightly power-broadened by the electromagnetic field. For practically all frequencies we find one individual eigenstate of \mathcal{H} having an overlap of ca. 50% or more with the initial state $|23, 22, 22\rangle$ and therefore clearly dominating the dynamics. For the quasi-one-dimensional state, on the contrary, the dynamics is determined by a significantly larger number of eigenstates of \mathcal{H} , each of them having—in turn—a rather small projection on $|23, 1, 0\rangle$ of typically 5% to 10%. As pulse-induced transitions between eigenstates of \mathcal{H} should be favored the larger the number of states involved in the dynamics, adiabatic switching should not alter the global behavior of the circular state depicted in Fig. 1. This is confirmed by the corresponding adiabatic thresholds displayed in the same figure. They systematically lie above the diabatic thresholds but do not change the qualitative dependence of $F_0(10\%)$ on ω_0 . Hence, the switching of the microwave field in a laboratory experiment is not crucial for the enhanced stability of the circular state, nor does it induce a significant change of the typical field strengths necessary to ionize the atoms. Since recently circular Rydberg states of rubidium with $n_0 \geq 65$ have been successfully prepared in the laboratory [17], one should expect that our predictions can be checked by real experiments.

The significant reduction of dominant eigenstates in the dynamics of the circular state is due to the reduction of the volume of classical phase space accessible to the electron by the centrifugal potential barrier. The density of states decreases with this volume, and since the electron is additionally prevented from probing the Coulomb singularity, which is at the origin of the nonlinear coupling of different unperturbed states by the microwave, it seems natural that only few eigenstates of \mathcal{H} govern the dynamics under the external perturbation. The reduced spectral density imposes a time scale on the evolution of the quantum system which is *short* compared to the time quasiclassical diffusion would need to induce an appreciable population transfer out of the initial state to other unperturbed atomic states. Consequently, even for short (but finite) interaction times the quantum evolution of the circular state *cannot* mimic any classically diffusive dynamics (living on a continuous frequency spectrum), in sharp contrast to the situation for the extended state $|23, 1, 0\rangle$, where the electron closely approaches the nucleus and the density of states is comparably large. The notion of dynamical localization be-

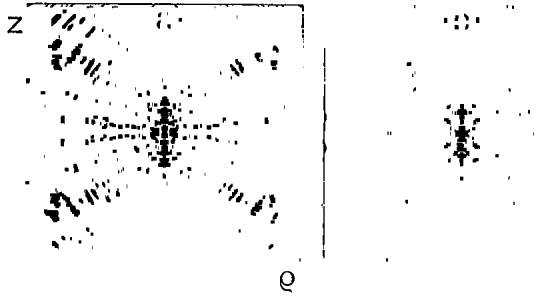


FIG. 2. Contour plots (in cylindrical coordinates z and ρ , field polarization parallel to z) of the one cycle average of the electronic density described by two representative resonance eigenfunctions of the Floquet Hamiltonian \mathcal{H} , for the quasi-one-dimensional state $|23, 1, 0\rangle$. $F_0 = F_0(10\%) = 0.0549$, $\omega_0 = 1.3$. Scales of the plot: $z, \rho = \pm 1000$ a.u. (a.u. is an arbitrary unit). Width Γ and overlap $|\langle \phi | 23, 1, 0 \rangle|^2$ of the eigenfunctions with the initial state: (a) $\Gamma \simeq 9.32 \times 10^{-9}$ a.u., $|\langle \phi | 23, 1, 0 \rangle|^2 \simeq 4.6\%$; (b) $\Gamma \simeq 1.82 \times 10^{-8}$ a.u., $|\langle \phi | 23, 1, 0 \rangle|^2 \simeq 8.7\%$. Each eigenstate overlaps with many unperturbed bound states of the atom.

comes therefore obsolete for the characterization of the ionization process of the circular state and of the stabilization observed in Fig. 1, as quantal and classical time scales got completely separated and quantum mechanics "inhibit" diffusive ionization *before* any quasiclassical diffusion can prevail. This conclusion is supported by the investigation of typical resonance eigenfunctions of the Floquet Hamiltonian \mathcal{H} , for both initial states compared in Fig. 1, at scaled frequency $\omega_0 = 1.3$ and at the corresponding values of $F_0(10\%)$. "Typical" means here an overlap with the initial state of the atom of about 10% or more. Because of the periodic time dependence of the Hamiltonian, the eigenstates are themselves periodic with the frequency of the driving field. Figure 2 displays, in a contour plot and in cylindrical coordinates, the one-cycle average of the probability distribution of two eigenstates with a projection of about 10% each on the initial quasi-one-dimensional state. The wave functions show an intricate nodal structure in both radial and angular directions, and are complicated linear combinations of the usual (n, l, m) states. A single eigenstate of \mathcal{H} overlaps, hence, with a large number of unperturbed atomic states, an observation which is complementary to the fact that several eigenstates of \mathcal{H} generate the dynamics. Figure 3 displays the electronic density of the resonance eigenfunction which has an overlap of 66% with the initial circular state and a width Γ of 1.01×10^{-9} a.u., for different phases of the external field. The structure of the wave function is very similar to the wave function of the unperturbed state $|23, 22, 22\rangle$, apart from a slight distortion and from the motion along the z axis which follows the oscillation of the driving field. The radial distance of the maximum of the distribution at phase $\pi/2$ of the field [Fig. 3(c)] equals 520 a.u., which differs by less than 2% from the usual estimate for the radial extension $n_0^2 = 529$ a.u. of the unperturbed circular state.

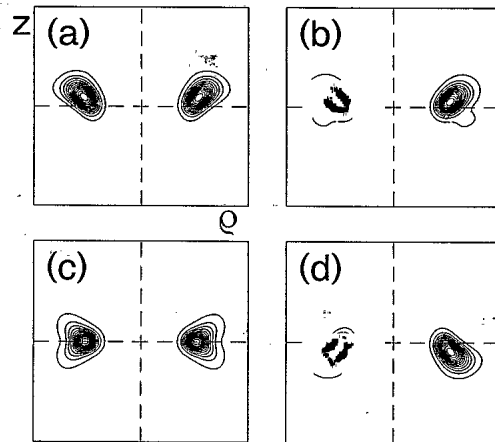


FIG. 3. Contour plots (in cylindrical coordinates) of the time dependent resonance eigenfunction dominating the quantum dynamics of the circular state $|23, 22, 22\rangle$. The crossing of the dashed lines indicates the position of the nucleus. $F_0 = F_0(10\%) = 0.19$, $\omega_0 = 1.3$. Scales of the plot: $z, \rho = \pm 1000$ a.u. Width Γ and overlap $|\langle \phi | 23, 22, 22 \rangle|^2$ with the initial state: $\Gamma \simeq 1.01 \times 10^{-9}$ a.u., $|\langle \phi | 23, 22, 22 \rangle|^2 \simeq 66\%$. Phases of the driving field: (a) 0, (b) $\pi/4$, (c) $\pi/2$, (d) π . Radial distance of the wave function maxima from the nucleus, at phase $\pi/2$: 520 a.u. (The unperturbed state $|23, 22, 22\rangle$ would be represented by two maxima of circular shape at $z = 0.0$ and $\rho \simeq \pm n_0^2 = 529$ a.u.) Maximum excursion in field ($\parallel z$) direction: 100 a.u., quiver length at the same frequency and amplitude: $\alpha = F/\omega^2 \simeq 59$ a.u..

The maximum excursion of the peak of the wave function in the z direction, at phases 0 and π of the field, equals ca. 100 a.u., which is of the same order as the excursion of a free classical electron in an oscillating field, given by the quiver length [11] $\alpha = F/\omega^2 \simeq 59$ a.u. As the effective potential seen by the circular state is locally harmonic, the discrepancy of the two values can be attributed to the vicinity of driving frequency and Kepler frequency of the unperturbed electron at $\omega_0 = 1.3$, which results in a resonant enhancement of the observed oscillation amplitude with respect to α . The almost classical motion of the electronic density in the driving field strongly reminds us of the dynamics of a circular state in a high intense laser field with a single photon energy *larger* than the binding energy of the initial state [12,14]. We therefore display in Fig. 4 contour plots of the state $|n_0 = 7, l_0 = 6, m_0 = 6\rangle$ when exposed to a linearly polarized laser field of intensity 1.0×10^{-2} a.u. and of frequency 0.0428 a.u. [10,13,14]. The lifetime $\tau = \Gamma^{-1}$ of this state equals $\tau = 3.2$ ns and the atom thus experienced adiabatic stabilization with respect to its characteristic minimum lifetime of $\tau = 23.7$ ps at $I = 4.0 \times 10^{-4}$ a.u. (i.e., at *lower* intensity) by 2 orders of magnitude. The maximum excursion of the electronic probability along the field direction equals 54.5 a.u. and matches perfectly the quiver length $\alpha = 54.6$ a.u. The similarity to the wave functions displayed in Fig. 3 is apparent, despite the fact that scaled field amplitude and frequency equal 0.19 and

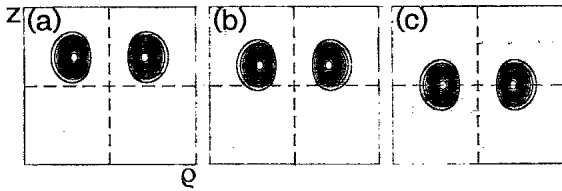


FIG. 4. Temporal evolution of the Floquet state adiabatically populated from the circular state $|n_0 = 7, l_0 = 6, m_0 = 6\rangle$, in cylindrical coordinates, over one quarter-cycle of the laser field. Scales of the plot: $z, \rho = \pm 150$ a.u. Intensity: $I = 1.0 \times 10^{-2}$ a.u.; frequency: $\omega = 0.0428$ a.u. [13,10,14]. $F_0 \simeq 240, \omega_0 \simeq 14.7$. Phase of the field equal to (a) 0.0; (b) $\frac{\pi}{4}$; (c) $\frac{\pi}{2}$. Maximum excursion of the wave function in z direction: 54.5 a.u.; quiver length at same amplitude and frequency: $\alpha = F/\omega^2 = 54.6$ a.u.. Width of the circular state at these field parameters: $\Gamma \simeq 7.48 \times 10^{-9}$ a.u., lifetime $\tau = \Gamma^{-1} \simeq 3.2$ ns. The atom thus experiences adiabatic stabilization [10,13,14] by 2 orders of magnitude with respect to its minimum lifetime of $\tau_{\min} = 23.7$ ps at $I_{\min} = 4.0 \times 10^{-4}$ a.u. Note the similarity to Fig. 3, despite the important difference in the field parameters characterizing the two physical situations.

1.3 in Fig. 3 and roughly 240 and 14.7 in Fig. 4. Again the eigenstate of \mathcal{H} is essentially the unperturbed state $|7, 6, 6\rangle$, oscillating with the frequency of the driving field, and no mixing with other states occurs.

Hence, for the whole parameter range delimited by the examples of Figs. 3 and 4 ionization cannot occur but via a *purely quantal* step-by-step multiphoton ionization process since, as mentioned above, quasidiffusive energy transport over adjacent unperturbed states necessitates many instead of one single eigenstate of \mathcal{H} participating in the dynamics. Adiabatic stabilization finally arises from the *kinematical* inhibition [11] of this *quantal* ionization process, when field frequency and amplitude have been continuously increased to sufficiently high values [13]. Decreasing, instead, m_0 allows for *quasiclassical*, diffusive excitation by the microwave, which will be inhibited by dynamical localization, due to *quantum coherence* effects. Although the mechanisms of adiabatic stabilization and dynamical localization are of *different* dynamical origin, the respective dynamical features do thus coexist and eventually compete. Each of them can be selected by the proper "tuning" of the internal (atomic) or external (field) parameters.

In conclusion, we presented first numerical quantum calculations simulating the microwave ionization of a circular Rydberg state, which is found to be significantly more stable than a quasi-one-dimensional state. The in-

creased stability, which should be observable in new experiments on circular Rydberg states of Rb [17], is accompanied by a remarkably simple structure of the corresponding wave function, providing us with a nice example of quasiclassical motion of an individual quantum state. Comparison to a quasi-one-dimensional Rydberg state in a microwave field and to a weakly excited circular state in a high intense, high frequency laser field suggests that a circular state subject to microwave radiation represents the missing link between dynamical localization and adiabatic stabilization.

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