Hundred photon microwave ionization of Rydberg atoms in a static electric field

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We present analytical and numerical results for the microwave excitation of nonhydrogenic atoms in a static electric field when up to 1000 photons are required to ionize an atom. For small microwave fields, dynamical localization in photon number leads to exponentially small ionization while above quantum delocalization border ionization goes in a diffusive way. For alkali atoms in a static field the ionization border is much lower than in hydrogen due to internal chaos.

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After two decades of investigations initiated by the pioneer experiment of Bayfield and Koch in 1974, the main features of the microwave ionization of highly excited hydrogen atoms are well understood. Generally, this ionization takes place due to emergence of chaos, in the corresponding classical problem, above a certain microwave intensity threshold. In the chaotic regime, quantum excitation proceeds via a diffusive absorption and reemission of field photons, which eventually leads to ionization. For a real atom the quantum process can be either close to the classical diffusion, or strongly suppressed when the field $\epsilon$ is below a quantum delocalization border $\ell_q$. This suppression is due to dynamical localization of classical chaos which is produced by quantum interference. In the localized case the distribution over the levels drops exponentially with the photon number. This dynamical localization is analogous to the Anderson localization in quasi one-dimensional disordered solids, with the photon number playing the role of lattice index. The quantum delocalization takes place if the localization length $\ell_\phi > N_l$, where, in atomic units, $N_l = 1/(2n_0^2\omega)$ is the number of photons required for ionization, with $n_0$ being the initial principal quantum number and $\omega$ the field frequency.

An absolutely different scenario for the microwave ionization of alkali Rydberg atoms had been proposed by Gallagher et al. based on experimental results in the regime with small rescaled frequency $\omega_0 = \omega n_0^3 \ll 1$. According to this scenario, ionization appears due to a chain of consecutive Landau-Zener transitions between nearly levels, which eventually brings the electron into the continuum. In the presence of quantum defects the Stark manifolds exhibit a structure of avoided crossings only for sufficiently strong fields with $\epsilon > 1/n_0^3$. Indeed, the experiments were in agreement with the $\epsilon \sim 1/n_0^3$ dependence for the microwave ionization threshold and not with the static field border $\epsilon_s = 1/(9n_0^3)$. This was considered as an experimental confirmation of the above scenario. However, this picture doesn’t explain in fact how the propagation actually occurs via the chain of these transitions and why the overlapping of two nearby levels guarantees that the electron will pass through the whole chain. The comparison between the thresholds for hydrogen and for alkali atoms clearly shows that the border is lower in the latter case and therefore it is related to the quantum defects $\delta_l$ of alkali atoms. Since these defects are different from zero only for orbital moment $l < 3$, the situation is purely quantum and cannot be treated by the quasi-classical approach used for hydrogen atom. This is the reason why so long after experiments have been made no detailed theory has been developed. To demonstrate a theoretical difficulty we note that hundreds of photons are required to ionize atoms in this regime ($N_l = n_0/2\omega_0 = 300$ for $n_0 = 60$ and $\omega_0 = 0.1$).

In this Letter we propose another mechanism for the microwave ionization of Rydberg atoms in a static electric field which is qualitatively different from the Gallagher et al. scenario. We argue that ionization in this case is not due to Landau-Zener transitions but to a quantum diffusive excitation in energy $E$ (or in the photon number $N = E/\omega$). Such quantum diffusion in the low frequency regime $1/n_0 < \omega_0 \ll 1$ becomes possible due to appearance of quantum chaos for Rydberg atoms in a static electric field. Indeed, a recent theoretical study showed that the level spacing statistics $P(s)$ in such atoms, for a sufficiently strong static field, is described by the Random Matrix Theory. These results indicate also a chaotic structure of eigenstates. This internal quantum chaos can lead to a diffusive excitation in energy even for a quite weak microwave field. In this respect, the situation is different from the hydrogen atom where chaos could appear only above some classical field threshold. Another consequence of internal chaos is an increase of density of effectively coupled states $\rho_c$. This gives a larger localization length $\ell_\phi (\ell_\phi \propto \rho_c)$ and therefore significantly decreases the delocalization border as compared to the hydrogenic case. A similar effect of chaotic
enhancement of localization length due to internal chaos has been studied recently for hydrogen atoms in magnetic and microwave fields [1].

![Diagram](image)

**Fig. 1.** Diffusion rate ratio $D_q/D_0$ as a function of microwave frequency $\omega_0$ for Rb (circles), Na (triangles) and Li (squares) at $n_0 = 60$, $\epsilon_{s0} = 0.02$. The full line is drawn to guide the eye. The upper insert gives $< (\Delta N_s)^2 >$ vs. $\tau$ at $\epsilon_0 = 0.03$, $\omega_0 = 0.1$: diffusive excitation for Rb (full curve $D_q/D_0 = 0.005$) and small oscillations for hydrogen (dotted curve with 100 times magnification). The lower insert illustrates the weak dependence of $D_q/D_0$ on $\epsilon_{s0}$ for Rb at $\omega_0 = 0.1$, $\epsilon_0 \geq 0.015$ (crosses).

To check the above picture of quantum photonic diffusion, we numerically studied the excitation of alkali Rydberg atoms (Rb, Na, Li) in a static electric field $\epsilon_s$ and parallel, linearly polarized, microwave field $\epsilon \sin \omega t$, for magnetic quantum number $m = 0$. We chose the rescaled value $\epsilon_{s0} = \epsilon_s n_0^3 \approx 0.02$ so that the statistics $P(s)$ for levels with $55 \leq n_0 \leq 72$ in Rb and Na was close to the RMT results. At the same time $P(s)$ for Li was closer to Poisson statistics due to a smaller value of quantum defects [1]. The investigation of time evolution, in the eigenbasis of the unperturbed problem ($\epsilon = 0$), showed that an initial eigenstate with energy $E_0$ spreads diffusively over the unperturbed energies $E_\lambda$, namely the square variance of the photon number $\sigma = < (\Delta N_s)^2 > = < (E_\lambda - E_0)^2 > / \omega^2$ initially grows linearly with the number of microwave periods $\tau$. Our quantum simulation allows to determine the value of quantum diffusion rate in energy per unit time $D_q = < (\Delta E)^2 > / \Delta t$. This rate $D_q$ can be compared with the diffusion rate in hydrogen at $\omega_0 = 1$, given by $D_0 = c^2 n_0 / 2$ [2].

![Diagram](image)

**Fig. 2.** Probability distribution over the eigenstates at $\epsilon = 0$ (full line) and in one-photon intervals (circles) for Rb: $n_0 = 60$, $\epsilon_{s0} = 0.02$, $\omega_0 = 0.1$, $\epsilon_0 = 0.005$, $180 \leq \tau \leq 200$, $D_q/D_0 = 0.0051$, $\ell_q = 9.1$. The straight line shows the fit for exponential localization with $\ell_q N = 13.3$. The one-photon probabilities are also shown, in the same conditions, for Na (triangles, shifted down by $10^3$, $\ell_q = 4.8$, $\ell_q N = 6.4$) and Li (squares, shifted down by $10^4$, $\ell_q = 3.1$, $\ell_q N = 5.4$).

The above diffusive excitation in energy induced by internal chaos may eventually be localized at long times due to quantum interference effects in a way similar to photonic localization in a complex molecular spectrum [3]. The localization length $\ell_q$, expressed in the number of photons, is proportional to the one-photon transition rate $\Gamma$ and to the density of coupled states $\rho_c$: $\ell_q \sim \Gamma \rho_c$ [2]. Since $\Gamma \sim D_q/\omega^2$ we obtain

\[ \ell_q = \ell_\phi D_q / D_0 \omega_0^2 n_0, \]  

(1)

Here the length $\ell_q$ is expressed via the localization length in hydrogen $\ell_\phi = 3.3 c n_0^3 / 2$ at $\omega_0 = 1$. Notice that in the non-hydrogenic case the density of states is $\rho_c = n_0^3$ due to internal chaos while in hydrogen the effective density is smaller, that is $\rho = n_0^3$ due to existence of an addi-
tional integral of motion \( \mathcal{I} \). The above expression for \( \ell_q \) is valid for \( \ell_q > 1 \) and \( \omega \rho_c = \omega_0 n_0 > 1 \). The localization leads to an exponential decay of probability distribution \( |\psi_N|^2 \sim \exp(-2|N_\phi|/\ell_q) \) in the photon number \( N_\phi = (E_\lambda - E_0)/\omega \).

\[ |\psi_N|^2 \sim \exp(-2|N_\phi|/\ell_q) \]

In order to check the theoretical prediction (1) we numerically computed the quantum evolution following it up to 200 microwave periods. The probability distribution \( f_N \) over the eigenstates of the static field problem (\( \epsilon = 0 \)) with energies \( E_\lambda \), was averaged over 10 - 20 periods to suppress the fluctuations. An initial state at \( \epsilon_0 = 0.02 \) was chosen as an eigenstate with energy \( E_{\lambda_0} \approx E_0 = 1/2n_0^2/\hbar \) and \( n_0 = 60 \). The system parameters were varied in the intervals: \( 0.02 \leq \omega_0 \leq 0.5 \), \( 0.003 \leq \epsilon_0 \leq 0.03 \) and \( 60 \leq N_I \leq 1500 \). The total basis included up to 1150 states.

A typical example of stationary distribution \( f \) is shown in Fig. 2. It clearly demonstrates exponential localization of diffusive excitation. One can note that among the three cases shown (Rb, Na, Li) the most localized is the case of Li, for which the quantum defect is minimal and therefore the internal chaos is the most weak. Notice also that in the case of Na and Li, for \( f_N < 10^{-7} \), the probability \( f_N \) starts to decay in a much slower way with \( \ell_q \approx 25 \) (Fig. 2). We attribute this effect to a significant modification of hydrogenic basis on highly excited levels where a static field becomes quite strong and tunneling effects for probability decay should be taken into account.

In order to find the localization length \( \ell_q \) we first compute the total probabilities \( f_N = |\psi_N|^2 \) in one-photon intervals \( |N_\phi - 1/2, N_\phi + 1/2| \) around integer values of \( N_\phi = E_\lambda/\omega \) and then extract the numerical \( \ell_{qN} \) value from the least square fit for \( \ln f_N \). The analysis of the numerical data \( \ell_{qN} \) shown in Fig.3 confirms the theoretical prediction (1) for \( \ell_q \) with \( D_q \) rates taken from Fig.1.

Equation (1) allows to determine the quantum delocalization border above which localization effects become unimportant and ionization goes in a diffusive way. This happens for \( \ell_q > N_I \), which gives the quantum delocalization border for the rescaled field \( \epsilon_q = \ell_q n_0^2 \):

\[ \epsilon_q = 0.4 \omega^{1/3} \omega_0^{1/6} \sqrt{D_0/D_q}. \]

Our numerical data indeed show that above this border complete delocalization takes place, contrarily to the case of hydrogen atom at the same field parameters (Fig.4). The strong fluctuations in the hydrogenic distribution \( f_N \) indicate that it is quite inhomogeneous inside the atomic shells.

\[ \epsilon_q = 0.4 \omega^{1/3} \omega_0^{1/6} \sqrt{D_0/D_q}. \]

For fixed \( \omega_0 \) the delocalization border (2) scales as \( \epsilon_q \sim 1/n_0^5 \), namely it is \( \sqrt{n_0} \) times smaller than for hydrogen at \( \omega_0 \sim 1 \). This drop of \( \epsilon_q \) is related to the appearence of internal chaos in nonhydrogenic atoms in a static electric field which effectively enhances the interaction with the microwave radiation. According to (2) the ratio of \( \epsilon_q \) to the ionization border proposed by Gallagher et al. \( \epsilon_G \approx 1/n_0 \) depends only on \( \omega_0 \): \( \epsilon_q/\epsilon_G = 0.4 \omega_0^{1/6} \sqrt{D_0/D_q} \). Our data from Fig.1 indicate that this ratio varies rather weakly with \( \omega_0 \) in the interval \( 0.02 < \omega_0 < 0.5 \) (see Fig.5). In spite of the fact that in this range \( \epsilon_q \sim \epsilon_G \), the physical interpretation is rather different from Gallagher et al. scenario. Indeed, the situation for \( \epsilon_0 < \epsilon_q \sim 1/n_0 \) is strongly nonperturbative since for \( \epsilon_q(2\omega_0/n_0)^{1/2} < \epsilon_0 \) many photons are absorbed with \( \ell_q > 1 \).

The theory developed above allows to understand ionization of atoms in a static electric field. It is possible to
expect that for $\omega_0 \ll 1$ the situation will remain similar even without static field since its role will be played by a slowly varying microwave field. However, one should be careful in extending the theory to a zero static field case. Indeed, our numerical results show that there, the probability distribution in orbital momentum is qualitatively different. For example, at $\epsilon_0 \sim 0.02, \omega_0 \sim 0.2$ only few $l$-states are mixed while with additional $\epsilon_{s0} \sim 0.02$ probability spreads over all accessible $l$. The localization in $l$ space at $\epsilon_{0} = 0$ had been also discussed for Rb atoms in[3]. The physical reason of this difference is related to the fact that the condition $\omega_0 \ll 1$ is not sufficient to treat the microwave field $\epsilon_0$ as quasi-static. For that one should require $\omega_0 \ll \omega_{s0} \approx 3\epsilon_0$ since the precession frequency $\omega_{s0}$ determines oscillations in $l$[1]. However, this condition is not compatible with the requirement $\omega_0 > 1/n_0$ for the considered region of $n_0$. For $\epsilon_{s0} = 0$ chaos is induced by the microwave field, the distribution in $l$ is nonhomogeneous and a detailed theory for this case becomes more complicated as compared to the nonzero static field where internal chaos is already present.

![Diagram](Figure 5. The ratio of the quantum delocalization border $\epsilon_q$ to the border $\epsilon_C = 1/n_0$ vs. $\omega_0$ for Rb (circles) obtained from Eq. (2) and data of Fig.1. The full line is drawn to guide the eye, crosses refer to the cases of Figs. 2,4. Another difficulty for direct comparison with the experiments[3] is that the latter were mainly done in the regime $\omega_0 < 1/n_0$. There is only one case for Na at $n_0 = 28, \omega_0n_0 = 0.76$ and $\epsilon_{s0} \approx 0.024$ which is not far from our regime (Fig. 2d in[3]). Here the experiment gives the ionization border $\epsilon_{ex} \approx 0.002$. This value is about 20 times smaller than the quantum delocalization border given by Eq. (2) with $D_q/D_0 = 0.0027$. We relate this difference to a tunneling ionization in a static field which becomes important for very long interaction times ($3 \times 10^5$ Kepler periods in[3]). Indeed, our preliminary numerical data[1] show that there is a change of slope in the probability decay similar to one in Fig.2 ($\epsilon_0 \approx 4$ for $f_n > 10^{-5}$ and $\epsilon_0 \approx 25$ for $f_n < 10^{-5}$). This means that in this case tunneling ionization plays a dominant role while the microwave only slightly increases its rate. An increase of the principal quantum number up to $n_0 = 60$ strongly suppresses the tunneling in a static field and the diffusive microwave excitation becomes dominant (Figs. 2,4). The effects of tunneling for $n_0 \approx 30$ will become less important for experiments with a shorter interaction time ($\tau \sim 100$) on which the dynamical localization dominates. It is also possible that for such long times as in[3] some noise in a microwave signal can strongly affect ionization. The above discussion shows that more detailed experimental investigations of microwave ionization in a static electric field are highly desirable. They will allow to make a detailed test of dynamical localization theory in the regime when up to 1000 photons are required to ionize an atom.

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