

LETTER TO THE EDITOR

How to build experimentally a non-spreading wavepacketJakub Zakrzewski^{†‡} and Dominique Delande[‡][†] Instytut Fizyki Mariana Smoluchowskiego, Uniwersytet Jagielloński, ul. Reymonta 4, 30-059 Kraków, Poland[‡] Laboratoire Kastler–Brossel, Université Pierre et Marie Curie, T12, E1, 4 place Jussieu, 75272 Paris Cedex 05, France

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Abstract. We show, by an exact numerical simulation, how to prepare a non-spreading electronic wavepacket in Rydberg atoms, using the nonlinear interaction of the Rydberg electron with a circularly polarized microwave field. The wavepacket can be prepared by a mixed diabatic/adiabatic switching on of the microwave field, which is studied in detail for realistic conditions.

The nonlinear interaction between the Rydberg electron of a hydrogen atom and an external microwave field leads to an appearance of novel quantum-mechanical objects—the non-spreading electronic wavepackets [1, 2]. Classically, a nonlinear resonance takes place when the Kepler frequency of the Rydberg electron is close to the frequency of an external microwave field. In effect, a *stable* periodic orbit with the period of the external drive exists around which the classical motion is completely *locked* on the microwave field. Hence, a wavepacket localized near the stable periodic orbit will remain localized: the usual spreading is suppressed.

The Hamiltonian of the driven system is time-periodic: thus, any solution of the Schrödinger equation is a linear combination of periodic in time ‘Floquet’ states. Some Floquet states are localized near the stable periodic orbit; they are non-spreading wavepackets as they follow the classical periodic orbit, returning exactly to their initial shape after one period. These wavepackets are intrinsically different from the usual ones, like the coherent states of the harmonic oscillator, as they are not built from linear superpositions of several eigenstates, but rather from a *single* Floquet state, automatically ensuring their non-spreading character.

Such wavepackets have been predicted and numerically observed in model systems [3] and in the hydrogen atom illuminated by a microwave field of either linear [1] or circular [2] polarization. In the latter case, the stable periodic orbits were first found by Klar [4]. In the frame corotating with the microwave field, one obtains an autonomous Hamiltonian (in au):

$$H = \frac{\mathbf{p}^2}{2} - \frac{1}{r} + Fx - \omega\ell_z, \quad (1)$$

where F and ω are the amplitude and frequency of the microwave field polarized in the x - y plane and ℓ_z is the angular momentum operator. The eigenstates of H are nothing but the Floquet states of the time-dependent Hamiltonian in the laboratory frame.

The stable periodic orbit locked on the driving field, a circle in the laboratory frame, appears in the rotating frame as a stable equilibrium point, located at $(x, y = 0, z = 0)$ with x the solution of:

$$\frac{1}{\omega^2 x^2} + \frac{F}{\omega^2} = x. \quad (2)$$

The above-resonance picture is valid only when the microwave field, F , is a small perturbation. When F is increased, chaotic motion appears around the resonance island and invades larger and larger regions of phase space. The island surrounding the equilibrium point exists until $F \approx 0.11556\omega^{4/3}$ where the equilibrium point turns unstable; above this value, the motion is mainly chaotic.

Following the initial discussion of non-spreading wavepackets in this system [2], it has soon been shown [5] by accurate diagonalization of equation (1) that their lifetime against ionization is extremely long (of the order of millions of Kepler periods) making their experimental observation feasible. The initial [5] simplified two-dimensional (2D) hydrogen atom analysis (restricting the motion to the plane of polarization) has been later extended to the fully three-dimensional (3D) system [6]. Furthermore, the wavepackets may be viewed as solitons travelling along the energy axis when analysed in the language of level dynamics [7].

The wavepackets being localized in the resonance island, their energies are accurately given by a harmonic analysis around the equilibrium point, as described in [5]. They are characterized by three quantum numbers, n_+ , n_- , n_z , representing the excitation along the normal modes with frequencies ω_+ , ω_- , ω_z (given explicitly in [5, 6]). The minimum wavepacket is $n_+ = n_- = n_z = 0$ and is a coherent superposition of mainly circular states $|n, l = m = n - 1\rangle$, with the mean n , n_0 , determined by the microwave frequency $n_0 \approx \omega^{-1/3}$ [5, 6].

The experimental preparation of such a non-spreading wavepacket is not obvious: as direct optical excitation is impossible [5], an indirect scheme has to be used, which has been first briefly described in [5] and that we now discuss in detail. It consists of two stages: preparation of the circular state of a given principal quantum number n_0 (which is by now a standard procedure [8]) and careful switching on of the microwave field with ω matching the Kepler frequency, n_0^{-3} , of the initial circular state. In analogy with a classical picture where one can expect that a classical distribution of particles, uniformly distributed on the initial ring (the equivalent equilibrium points due to the azimuthal symmetry of (1) at $F = 0$), will slowly reassemble around the stable equilibrium point building a localized ‘wavepacket’ when the microwave field amplitude is slowly increased, we proposed [5] that such a procedure will produce the localized wavepackets also in the quantum world.

Let us now turn to a careful analysis of what a ‘slow’ increase of the microwave field should be. If the turn-on is too fast, the atomic state cannot follow the changes in the equilibrium points and remains frozen. If the switching is too slow, it may either ionize or get transferred to another state if a large avoided crossing is met. A quantitative analysis is needed to know how the various energy levels of the Hamiltonian evolve with F . Because of the mixed regular-chaotic dynamics, the evolution of the energy levels with F is usually very complicated. Typically the wavepacket is non-degenerate with other states, except close to resonances between normal modes.

However, when F tends to zero, ω_- goes to zero [5] since it corresponds to an angular normal mode along which the equilibrium is marginally stable as F goes to zero. Hence, all the n_- states (including the $n_- = 0$ wavepacket) are degenerate for very small F ; the states may mix and, therefore, a better analysis is necessary. Figure 1 shows the exact energy spectrum of the Hamiltonian, equation (1), in this region. For clarity—smaller numbers of

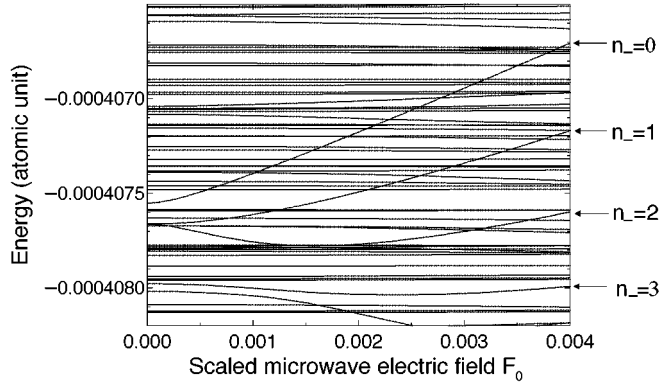


Figure 1. Energy levels of the 2D hydrogen atom exposed to a circularly polarized microwave field with frequency $\omega = 1/(60.5)^3$ (in au) as a function of the scaled microwave field $F_0 = F(60.5)^4$. The important states contributing to the dynamics are indicated by their value of n_- . The non-spreading wavepacket ($n_- = 0$) is prepared from the $F = 0$ situation by a mixed diabatic/adiabatic increase of the microwave field, which goes diabatically through the tiny avoided crossings but adiabatically follows the energy curve of the wavepacket.

energy curves—this plot is done for the 2D system; similar conclusions are obtained for the 3D system, the only difference being that the effective principal quantum number is a half-integer, $n_0 + \frac{1}{2}$, in 2D and an integer, n_0 , in 3D. We here choose $n_0 = 60$ and, accordingly, the microwave frequency is $\omega = 1/(60.5)^3$. In the plot, the non-spreading wavepacket is indicated by the arrow $n_- = 0$, while the excited states along the ω_- mode are indicated by their n_- values. From the Hellman–Feynman theorem, the slope of the energy curve is the average dipole of the state. Thus the states localized near the equilibrium point, with large average dipoles, have the largest slopes in the figure. When F is decreased, the various n_- states are closer in energy and undergo strong level repulsion: finally, they evolve into the circular states as $F = 0$, with energies (in the rotating frame):

$$E(n) = -\frac{1}{2n^2} - \omega(n-1). \quad (3)$$

These ‘important’ states are only weakly coupled to other ‘unimportant’ states which hence can be considered separately.

The full transition from circular states at $F = 0$ to n_- states localized in the regular island can be quantitatively understood in terms of a Mathieu equation [9], but such a detailed analysis is not required for our purpose. The crucial point is that the wavepacket we want to prepare has the *largest* energy among the important states. Hence, if we start from the *largest* energy state at $F = 0$ and adiabatically increase the field strength, we prepare the wavepacket. The optimal condition for adiabatic transfer is obtained when the wavepacket is as far (in energy) as possible from the other important states: there, the initial energy gap is maximum and the adiabaticity criterion less stringent. A simple calculation from equation (3) shows that this is obtained for

$$\omega_0 = \omega n_0^3 = 1. \quad (4)$$

A rough estimate is that the switching time has to be longer than the inverse of the minimum energy gap, that is of the order of n_0^4 , i.e. n_0 microwave periods. If $\omega^{-1/3}$ is not exactly an integer value, an efficient excitation of the wavepacket is still possible provided the initial state has the largest energy among the important states, that is roughly provided

$|\omega^{-1/3} - n_0|$ is smaller than 0.5. For the 2D system, the previous results apply, provided all integer values are replaced by half-integers.

Next, the effect of ‘unimportant’ states has to be taken into account. These are non-circular states (in the $F = 0$ limit) or chaotic states not lying in the resonance island at non-zero F . As discussed in [5, 7] and visible in figure 1, they may have energies close to the one of the wavepacket, but are weakly coupled to it: it results in a series of very small avoided crossings which have to be passed *adiabatically*, otherwise some unimportant states are populated.

Hence, the whole excitation process has to be done in a mixed diabatic/adiabatic way: adiabatic at the very beginning when the coupling with important states is crucial for assembling the probability density in the vicinity of the equilibrium point, diabatic at higher field strengths when crossing the energy curves of the unimportant chaotic states. The adiabatic condition can be easily satisfied using a smooth pulse rise of the form, e.g.:

$$F(t) = F_{\max} \sin^2 \left(\frac{\pi t}{2T_{\text{switch}}} \right) \quad (5)$$

for $0 \leq t \leq T_{\text{switch}}$.

To test the previous analysis, a numerical experiment is done with a microwave pulse described by equation (5). We start from a circular state and numerically integrate the time-dependent Schrödinger equation in the rotating frame. To avoid any approximation, we choose an ‘exact’ approach: we expand the wavefunction onto a Sturmian basis and compute the time evolution of the expansion coefficients. The Sturmian basis being complete, no information is lost. The (small) multiphoton ionization to the continuum is fully taken into account by the use of complex coordinates (just as for ‘exact’ calculations of resonances of the time-independent Hamiltonian by numerical diagonalization, see [5, 6]). We use a basis set of size $\simeq 23\,000$ for the 2D system and $\simeq 43\,000$ for the 3D case. The time-propagation is done using a fifth-order, stiffly accurate embedded Rosenbrock scheme [10]. The numerical details of the method will be published elsewhere. Errors due to truncation of the Sturmian basis and time propagation have been carefully controlled. We thus claim that our results do not involve any other approximation than neglecting relativistic, QED and finite nuclear mass effects.

Figure 2 shows snapshots of the electron probability density for the 2D system exposed to a microwave field of the form (5), with frequency $\omega = 1/(60.5)^3$ and maximum scaled field $F_0 = F_{\max}/\omega^{4/3} = 0.03$. The initial state is the circular $|n = m = 60\rangle$ state and the switching time is $T_{\text{switch}} = 400$ microwave periods. At $t = 0$, the electronic density has its initial circular symmetry. As the microwave field is turned on, it progressively concentrates near the stable equilibrium point, as expected. At the end of the pulse rise, $t = 400$, it is almost indistinguishable from the true non-spreading wavepacket (exact eigenstate of H , equation (1)), which proves the outstanding efficiency of the excitation scheme. In order to measure this efficiency, we compute the overlap of the final state at $t = T_{\text{switch}}$ with the exact eigenstate of H . It is almost 0.94 in the case of figure 2. In figure 3(a), this overlap is plotted as a function of T_{switch} for the same conditions as in figure 2. As expected, some efficiency is lost for short switching times (failure of the initial adiabatic stage) and for long switching times (failure of the diabatic crossings with unimportant states). The optimum switching time is found to be around $t = 400$ microwave periods. A more detailed quantitative analysis of the diabatic/adiabatic scheme suggests that efficient excitation is obtained in the range

$$\begin{cases} T_{\text{switch}} > n_0 \text{ microwave periods} \\ T_{\text{switch}} < n_0^2 \text{ microwave periods,} \end{cases} \quad (6)$$

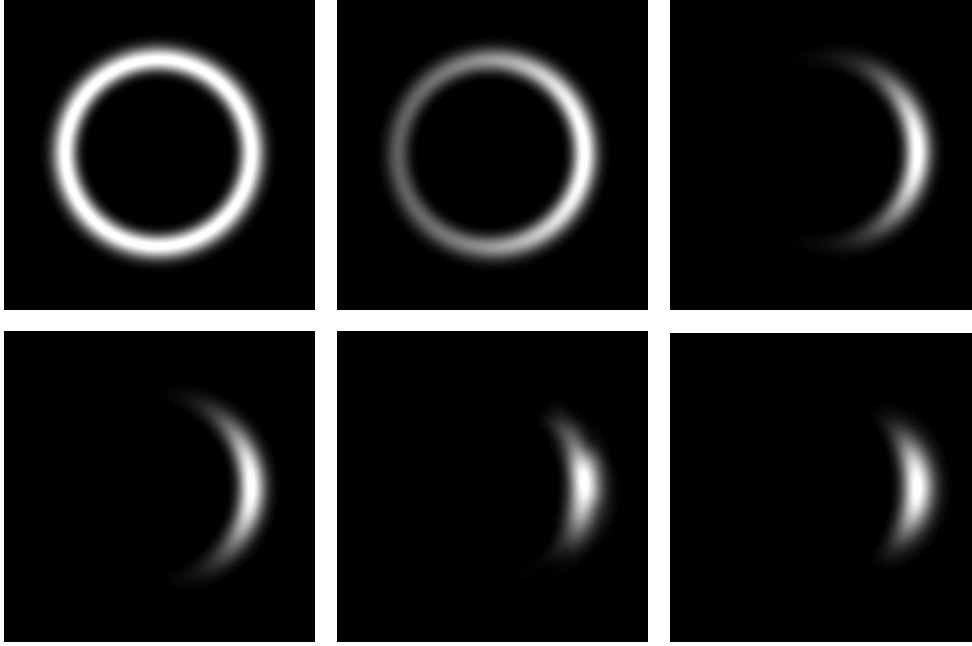


Figure 2. Snapshots of the electronic density during the switch-on of the microwave field using equation (5) with maximum scaled field $F_0 = F_{\max}\omega^{-4/3} = 0.03$ and $T_{\text{switch}} = 400$ cycles of the microwave at frequency $\omega = 1/(60.5)^3$. Top-left, $t = 0$ (initial circular state); top-middle, $t = 20$; top-right, $t = 60$; bottom-left, $t = 100$; bottom-middle, the final state, $t = 400$; bottom-right, the exact eigenstate, almost indistinguishable from the previous one, which proves the high efficiency of the excitation process.

the two conditions being respectively related to the initial adiabatic stage and the diabatic crossing stage. This leaves a large interval of possible switching times.

These results—obtained for the 2D system—are fully confirmed by the few points (squares in figure 3) computed for the 3D system using the initial circular state $|n = 60, l = m = 59\rangle$, and a microwave frequency $\omega = 1/(60)^3$. As a matter of fact, the 3D results are almost equal to the 2D ones, confirming that the third dimension (perpendicular to the polarization plane) does not play any significant role in this problem. Figure 3(b) shows the efficiency of the excitation scheme as a function of the microwave frequency. It confirms that optimal efficiency is obtained for $\omega_0 = 1$, but that the frequency of the microwave to be experimentally used is not a crucial parameter.

Finally, we now discuss some effects that could perturb the proposed excitation scheme. The first one is the multiphoton ionization of the wavepacket during the microwave turn on. A previous study [6] has shown that the ionization rate of the non-spreading wavepacket has enormous fluctuations, but remains low because it is limited by a tunnelling process. Here, these fluctuations are averaged during the diabatic/adiabatic stage and the total loss by ionization (computed exactly in the numerical experiment) is of the order of 1% for $T_{\text{switch}} = 400$ microwave periods. Hence, it should not be a serious limitation. Once the circular state is created, the electron never goes close to the nucleus: hence, the same excitation scheme can be used for a non-hydrogenic atom, without any perturbation due to the core. For the same reason, spontaneous emission should not be a problem, the lifetime (for $n \simeq 60$) being of the order of many million periods. An imperfect circular polarization

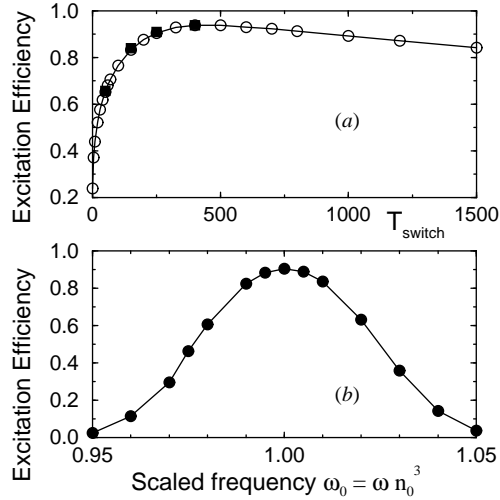


Figure 3. (a) The overlap between the wavefunction obtained at the end of the microwave turn-on and the exact state representing the non-spreading wavepacket, as a function of the switching time, T_{switch} , obtained for the 2D H atom (circles). Filled squares indicate results obtained for a fully 3D atom. F_0 and ω as in figure 2. (b) Same as (a), but for $T_{\text{switch}} = 250$ and changing the scaled frequency $\omega_0 = \omega(60.5)^3$.

will result in a non-static problem in the rotating frame. From the discussion at the beginning of this paper, this should not affect the existence and properties of the non-linear resonance process and the rest of the excitation scheme. The small static field (along z) used to stabilize the initial circular state [8] is also a small perturbation. More generally, the full process relies on the deformation of classical invariant tori of the system, which—from the KAM theorem—are structurally stable against any small perturbation. The only important geometrical factor is that the microwave electric field has to lie in the plane of the initial circular state, which requires that the static field is aligned along the axis of propagation of the microwave. From the general properties of circular states [8], this requires alignment within an angle $\simeq 1/\sqrt{n}$ (few degrees for $n = 60$) and should not be a problem.

In conclusion, we have shown that a non-spreading electronic wavepacket could be experimentally prepared using circular Rydberg atoms exposed to a time-increasing circularly polarized microwave field. Optimal conditions for such an experiment have been derived. For example, for a circular state $n = 60$, a microwave field of frequency 30.46 GHz switched to a maximum field amplitude $F = 11.9$ V/cm in about 15 ns will produce a non-spreading wavepacket with 94% efficiency.

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