Multiple Time Scales in the Microwave Ionization of Rydberg Atoms

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and
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We investigate the time dependence of the ionization probability of Rydberg atoms driven by microwave fields, both numerically and experimentally. Our exact quantum results provide evidence for an algebraic decay law on suitably chosen time scales, a phenomenon that is considered to be the signature of nonhyperbolic scattering in unbounded classically chaotic motion.

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The time scales characterizing classical probability transport in Hamiltonian systems with mixed regular chaotic dynamics have been subject to theoretical studies for more than one decade [1–8]. Whereas the probability for a particle to stay in a confined region of phase space is expected to decay *exponentially* for *globally chaotic* (hyperbolic) systems [6], the decay has been shown to *slow down* in the presence of regular regions and/or of remnants of invariant tori (cantori) in classical phase space, i.e., in the case of *nonhyperbolic* scattering [1–3,5,6]. For sufficiently large time intervals the survival probability $P(t)$ of a particle moving in mixed phase space has been observed to decay algebraically in numerical studies of several model systems [2–5]. In particular, algebraic decay of $P(t)$ has been numerically observed in the classical dynamics of a Rydberg electron driven by a microwave field of linear [7] and of circular polarization [8], constrained to the axis or the plane defined by the electromagnetic field vector. In these studies, the decay exponents α in $P(t) \sim t^{-\alpha}$ allowed a reasonable fit of the numerical results on about 1 order of magnitude of the interaction times of the atoms with the microwave field.

From the point of view of quantum mechanics, the structure of classical phase space at a given amplitude and frequency of the driving microwave field is faithfully reflected by the Floquet eigenstates of the Rydberg atom in the field [9,10]. The "initial conditions" in a numerical or laboratory experiment to be compared to the above-mentioned classical predictions are therefore defined by the initial population of the different Floquet eigenstates $|\epsilon_i\rangle$ at $t = 0$, i.e., by their relative weights $|c_i|^2$ at the instant of time when the microwave amplitude experienced by the atoms reaches its maximum value F, which further on is kept constant.

The survival probability (averaged over the initial phase of the microwave field) can then be written as

$$P(t) = \sum_i |c_i|^2 \exp(-\Gamma_i t), \qquad (1)$$

where Γ_i is the field-induced width of the Floquet state that accounts for the coupling to the atomic continuum and gives rise to the ionization of the Rydberg electron. Note that we assume a microwave pulse envelope with a flat top of length t and rising and falling edges of duration $\tau \ll t$, a situation that is met in our setup as well as in other currently used experimental setups [11].

From (1) it is not at all obvious that $P(t)$ will show algebraic decay for long interaction times as in the classical picture. A nonexponential decay is only possible [12] if several Floquet states significantly contribute to the decay, i.e., if the distribution of the $|c_i|^2$ of the initial state in the Floquet basis has a significant width [13,14]. Only for regular classical dynamics one single Floquet state may dominate the decay process for all times, leading to a monoexponential behavior of $P(t)$ [12]. For a quantum system with underlying mixed regular chaotic classical dynamics the width will be generically large, for any envelope of a realistic laboratory microwave pulse, as a consequence of the abundance of avoided crossings of any size in the Floquet spectrum [15]. The actual shape of the pulse envelope seen by the atoms will define the details of the initial distribution of the $|c_i|^2$ and hence of the initial conditions for quantum probability transport, but it will leave unaffected its general characteristics. Hence, Eq. (1) typically leads to a multiscale behavior of $P(t)$, which asymptotically may approximate an algebraic decay law $P(t) \simeq t^{-\alpha}$ [5]. However, the exponent α may depend on the switching of the microwave field [8]. Our main interest in this paper is the algebraic behavior, rather than the numerical value of the exponent together with its dependence on the various experimental parameters.

The purpose of the present contribution is to address the relevance of the classical picture of probability transport

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for *real* atomic systems, on the grounds of an *exact* numerical treatment of the 3D hydrogen atom exposed to a microwave field of linear or circular polarization, and of results from *laboratory* experiments on rubidium Rydberg states.

Let us first discuss our numerical experiments. We skip the details [12,16] of the theoretical approach and only recall that we use a combination of the Floquet picture together with a complex dilation of the Hamiltonian describing the atom in the field, to obtain quasienergies ϵ_i and decay rates Γ_i of the Floquet eigenstates $|\epsilon_i\rangle$. The influence of spontaneous emission, irrelevant for our discussion, is omitted. With these ingredients we calculate the survival probability of the initial bound state population distribution given by the weights $|c_i|^2$.

We consider the ionization of an extremal parabolic state $|n_1 = 22\, n_2 = 0\, m_0 = 0\rangle$, $n_0 = n_1 + n_2 + |m_0| + 1 = 23$ the principal quantum number, by a linearly polarized microwave field, and of a circular state $|n_0 = 23\, \ell_0 = 22\, m_0 = 22\rangle$, by a field of circular polarization, respectively. Both these atomic initial states correspond to classically mixed regular-chaotic dynamics of the Rydberg electron driven by the oscillating field [10,17]. The $|c_i|^2$ are defined by the projection of the atomic initial states onto the Floquet states at microwave amplitude F. Our choice of n_0 is a compromise between the actual principal quantum numbers initially excited in laboratory experiments [11,13], $n_0 \simeq 60$, and the limitations of memory of currently available supercomputers. To establish a qualitative comparison between numerical and state of the art laboratory experiments, we use scaled variables $\omega_0 = \omega n_0^3$, $F_0 = F n_0^4$, which are familiar from earlier studies [11,12]. We fix $\omega_0 = 1.6$ and vary F_0 and t, in order to explore the different regimes of the temporal evolution of $P(t)$.

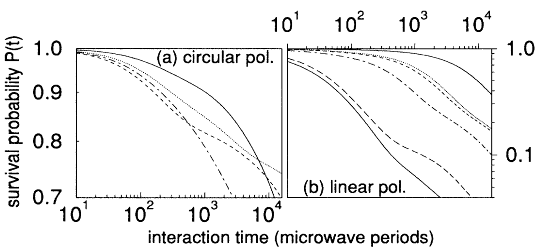

FIG. 1. Survival probability of real 3D atomic hydrogen exposed to a microwave field of variable scaled amplitude F_0 and scaled frequency $\omega_0 = 1.6$, on a double logarithmic scale. Interaction time from 10 to 16 000 microwave periods. $F_0 = 0.05$ (continuous line), 0.063 (dotted line), 0.065 (short dashed line), 0.075 (dash-dotted line), 0.1 (long dashed line), and 0.112 (continuous line with the largest ionization yield). The initial states of the atoms have been chosen to correspond to classically mixed regular chaotic dynamics. (a) Circular state $|n_0 = 23\, \ell_0 = 22\, m_0 = 22\rangle$ in a circularly polarized field; (b) extremal parabolic state $|n_1 = 22\, n_2 = 0\, m_0 = 0\rangle$ (principal quantum number $n_0 = 23$) in a linearly polarized field. Note the variation of the typical decay rate with time, for both initial states and polarizations, but also the difference in the ionization yields at the maximum of the interaction time.

Figure 1 shows the survival probability of an electron initially prepared in a circular (a) or in an extremal parabolic (b) state, for different amplitudes of the driving field, and over 3 orders of magnitude of the interaction time, corresponding to approximately 10 to 10 000 microwave periods T_ω (or Kepler periods T_K of the unperturbed Rydberg electron, at $\omega_0 = T_K/T_\omega = 1.6$). Note that, for both initial states and the respective polarizations of the driving field, the survival probability starts out with a decay that is neither exponential nor algebraic [12] (the latter would result in a straight line on the double logarithmic scale of Fig. 1). For longer interaction times, different Floquet states with different decay rates dominate the ionization process on different time scales, eventually leading to "kinks" in the survival probability, depending on the distribution of the Γ_i. Note that these kinks occur at different values of $P(t)$ in the cases of circular and of linear polarization, respectively, as a consequence of the difference in the relative volumes of regular and chaotic motion in both cases [17]. Since the Floquet states faithfully respect the structure of classical phase space [9,10], we attribute the larger contribution to the ionization yield of the parabolic state to those $|\epsilon_i\rangle$ associated with the chaotic part of phase space and typically shorter lifetimes. States that are associated with invariant tori or cantori should exhibit significantly longer lifetimes [10]. Hence, their contribution to $P(t)$ becomes relevant only at longer interaction times or higher ionization yields. Contrary to that, Floquet states associated with cantori, i.e., with the remnants of Kolmogorov-Arnold-Moser tori [1] almost immediately dominate the ionization of the circular state in the circularly polarized field, due to the more regular structure of classical phase space. It is such states that induce the kinks in the ionization yield, for any polarization of the microwave field, as they typically undergo large avoided crossings with adjacent Floquet states of comparable weight $|c_i|^2$ but distinct lifetime. Note that the separation of time scales in $P(t)$ and hence in the decay rates themselves is in very close analogy to the separation of different domains of classical phase space according to the confinement times of classical trajectories launched in each of these regions [7], and that our quantum results displayed in Fig. 1 reproduce qualitatively the phenomena observed in the classical studies [7,8]: (1) the approximately algebraic decay over ca. 1 order of magnitude of the interaction time, (2) the decrease of the decay exponent as the microwave field strength increases, and (3) the crossover between two decay exponents.

Approximate algebraic decay over *more* than 1 order of magnitude of the interaction time can be observed if the decay rates Γ_i and the weights $|c_i|^2$ of the associated Floquet states have a sufficiently broad distribution extending over several orders of magnitude. Under these conditions, several kinks as observed in Fig. 1 will succeed each other on successive time scales, leading, on the average, to an algebraic decay of the survival probability. This phenomenon is illustrated in Fig. 2, which extends Fig. 1, for some of the considered amplitudes F, to longer interaction times. Most of the Floquet states that contribute at this stage of the ionization process seem to be associated with regular regions or remnants of those in classical phase space, according to a preliminary investigation of the Floquet spectrum. This is consistent with classical studies [6,7], where algebraic decay over more than one decade is always associated with trajectories that visit the phase space region that separates regular from chaotic motion. Both the decay exponents observed for circular ($\alpha \simeq 0.25$) and linear polarization ($\alpha \simeq 0.59$) are clearly different from the value $\alpha = 2/3$ predicted in [4], a classical study that, however, did not account for the impact of residual stability islands on the bound state probability decay.

Let us finally provide the results of laboratory experiments that demonstrate the occurrence of qualitatively the same phenomena in the ionization of Rydberg states of ^{85}Rb. For details of the experimental setup we refer to earlier work [13]. In short, we resonantly excite rubidium Rydberg states $n_0 p_{3/2}$ from the atomic ground state, with $n_0 \in [40; 170]$, which then interact with a monochromatic, linearly polarized microwave field. Finally, for well defined values of F, ω, and t, the survival probability is defined as the ratio of the number of atoms that survived the microwave ionization to the number of atoms initially prepared in the Rydberg state $n_0 p_{3/2}$. Changing t with all other experimental parameters fixed allows us to directly monitor $P(t)$. In the present study we chose $n_0 = 84$. The laboratory frequencies were $\omega^{(1)}/2\pi = 12\,059.44$ MHz and $\omega^{(2)}/2\pi = 8867$ MHz, respectively. If we employ the same pragmatic scaling for the nonhydrogenic states of rubidium as in Ref. [13], these laboratory values correspond to scaled frequencies $\omega_0^{(1)} \simeq 2.2$ and $\omega_0^{(2)} \simeq 1.6$, respectively, the latter coinciding with the scaled frequency employed in our numerical experiments. The microwave pulse experienced by the Rydberg states had a flat top of duration $t \simeq 20$ ns–10 μs (corresponding to ca. 10^5 field cycles, comparable to the largest interaction times in our numerical calculations), with smoothly rising and falling wings of a duration of $\tau \simeq 6$ ns (corresponding to 50 to 100 microwave cycles).

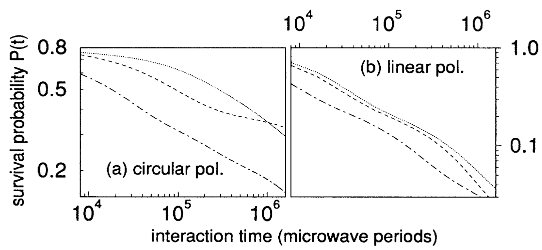

FIG. 2. Same as Fig. 1, but for longer interaction times from 8000 to 1.6×10^6 field cycles. $F_0 = 0.063$ (dotted line), 0.065 (short dashed line), and 0.075 (dash-dotted line). Note the approximately algebraic decay $P(t) \sim t^{-\alpha}$ (corresponding to a straight line in the presented double logarithmic plot) over more than 2 orders of magnitude, both for the circular state in a circularly polarized field [for the two stronger fields (a) ($\alpha \simeq 0.25$) and for the extremal parabolic state exposed to a linearly polarized microwave (b) ($\alpha \simeq 0.59$). Since we are dealing with an open system that classically displays unbounded chaotic dynamics, this algebraic decay can be interpreted as a signature of nonhyperbolic scattering.

Figure 3(a) shows the survival probability of the atoms on a double logarithmic scale, for $\omega_0^{(1)} \simeq 2.2$, and for two different values of the microwave amplitude. In both cases, for $F = 2.79$ and 3.42 V/cm ($F_0 \simeq 0.024$ and 0.029, scaled with respect to the static field ionization limit), we observe in very good approximation a monoexponential decay. In contrast, Fig. 3(b) shows the transition from a monoexponential to a monoalgebraic decay $P(t) \sim t^{-\alpha}$, $\alpha \simeq 0.44 \pm 0.02$, as the amplitude of the field of frequency $\omega_0^{(2)} \simeq 1.6$ is increased from $F = 1.96$ to 3.92 V/cm, corresponding to $F_0 \simeq 0.017$ and 0.033, respectively. Note that, for both frequencies, the fits extend over more than 2 orders of magnitude of the interaction time. Because of the well-known differences in the ionization behavior between Rydberg states of atomic hydrogen and rubidium, we cannot make a quantitative comparison here between the numerical and experimental results. Note, however, that the experimentally observed decay exponent $\alpha^{\rm exp} = 0.44 \pm 0.02$ is close to the numerical observation of Fig. 2(b), $\alpha^{\rm num} \simeq 0.59$, on comparable time scales.

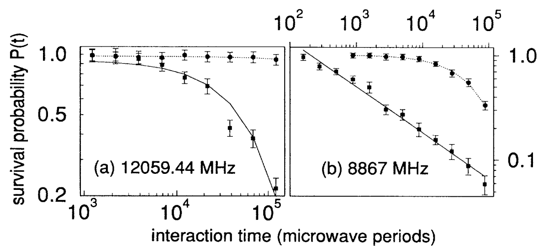

FIG. 3. Survival probability of $84p_{3/2}$ Rydberg states of ^{85}Rb exposed to a microwave field of linear polarization, on a double logarithmic scale. (a) Frequency $\omega^{(1)}/2\pi = 12\,059.44$ MHz ($\omega_0^{(1)} \simeq 2.2$), field amplitudes $F = 2.79$ V/cm ($F_0 \simeq 0.024$; circles), and $F = 3.42$ V/cm ($F_0 \simeq 0.029$; squares). (b) Frequency $\omega^{(2)}/2\pi = 8867$ MHz ($\omega_0^{(2)} \simeq 1.6$), field amplitudes $F = 1.96$ V/cm ($F_0 \simeq 0.017$; circles) and $F = 3.92$ V/cm ($F_0 \simeq 0.033$; squares). The interaction time has been varied from $t \simeq 20$ ns to $t \simeq 10$ μs, corresponding to ca. 10^5 microwave cycles (or Kepler orbits of the Rydberg electron). Clearly, increasing the field amplitude does not change the monoexponential behavior (fitted by the dotted and full line, respectively) observed for $\omega_0^{(1)}$, but does induce a transition from monoexponential (fitted by the dotted line) to monoalgebraic decay (over approximately 3 orders of magnitude, with a decay exponent $\alpha \simeq 0.44 \pm 0.02$, $P(t) \sim t^{-\alpha}$, obtained from the fit represented by the full straight line) for $\omega_0^{(2)}$.

The experimental results may be interpreted following the same arguments as presented before. For the microwave frequency $\omega_0^{(1)}$, the ionization of the initial state $84p_{3/2}$ by the field is essentially dominated by its diabatic continuation in the field, there is no redistribution of the atomic population due to passages through avoided crossings encountered during the rising part of the microwave pulse, for each of the applied field strengths, i.e., also for the larger one yielding almost complete ionization on the experimental time scale. This suggests that, with respect to the time (and energy) scale set by the experimental switching time τ, the frequency $\omega_0^{(1)}$ places us in a regular region of the quantum spectrum, i.e., a regime where, locally, the eigenstates of the atom in the field neighboring the diabatic continuation of the initial state do not undergo avoided crossings of appreciable size (i.e., there are only avoided crossings of width $\Delta\epsilon \ll 1/\tau$) as the driving amplitude F is changed. Hence, all these anticrossings are passed diabatically during the rising part of the microwave pulse, no population is lost out of the diabatic continuation of the initial state. This is completely consistent with an earlier observation [13], which also suggested the excitation of one single Floquet state of the atoms at frequency $\omega_0^{(1)}$, in our experiment.

A different picture emerges for the second frequency $\omega_0^{(2)}$. Here, the increase of the microwave amplitude does induce a redistribution of the atomic population over several Floquet states, which anticross with the adiabatic continuation of the initial state $84p_{3/2}$, during the rising part of the microwave pulse. Hence, the $|c_i|^2$ have a distribution of finite width at $t = 0$. For this frequency, the initial state $84p_{3/2}$ is therefore connected to the irregular part of the quantum spectrum, i.e., the adiabatic continuation of the initial atomic state in the field encounters avoided crossings of a width $\Delta\epsilon$ comparable to or much larger than τ^{-1}, finally leading to a result qualitatively comparable to Fig. 2(b). Note that our experimental distinction between "regularity" and "irregularity" relies on the relative size of the avoided crossings encountered during the rising part of the microwave pulse as compared to the energy resolution set by the inverse of the switching time τ.

In conclusion, algebraic decay—a signature of nonhyperbolic scattering—of the survival probability of Rydberg states in a microwave field was observed over several orders of magnitude of the interaction time. For interaction times of the order of 10^3 or 10^6 microwave periods a crossover from faster to slower and eventually algebraic decay could be observed (for properly chosen values of the field amplitude). This reflects the separation of time scales in the classical probability transport and, finally, the underlying mixed regular chaotic phase space structure itself. In a laboratory experiment, comparable interaction times extending over several orders of magnitude led to monoexponential or monoalgebraic decay of the survival probability of Rydberg states of rubidium, depending on the frequency and the amplitude of the driving field. All other experimental parameters fixed by the measurement of $P(t)$ may therefore be conceived as a probe of regular and irregular domains of the quantum spectrum.

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[1] R. S. MacKay, J. D. Meiss, and I. C. Percival, Phys. Rev. Lett. **52**, 697 (1984).
[2] B. V. Chirikov and D. L. Shepelyansky, Physica (Amsterdam) **13D**, 395 (1985).
[3] J. D. Meiss and E. Ott, Phys. Rev. Lett. **55**, 2741 (1985).
[4] F. Borgonovi, I. Guarneri, and P. Sempio, Nuovo Cimento **102B**, 151 (1988).
[5] G. Zumofen and J. Klafter, Europhys. Lett. **25**, 565 (1994).
[6] Y.-C. Lai, R. Blümel, E. Ott, and C. Grebogi, Phys. Rev. Lett. **68**, 3491 (1992).
[7] Y.-C. Lai, C. Grebogi, R. Blümel, and M. Ding, Phys. Rev. A **45**, 8284 (1992).
[8] R. Gębarowski and J. Zakrzewski, Phys. Rev. A **50**, 4408 (1994).
[9] J. Henkel and M. Holthaus, Phys. Rev. A **45**, 1978 (1992).
[10] A. Buchleitner, thèse de doctorat, Université Pierre et Marie Curie, Paris, 1993 (unpublished).
[11] B. E. Sauer, M. R. W. Bellermann, and P. M. Koch, Phys. Rev. Lett. **68**, 1633 (1992).
[12] A. Buchleitner, D. Delande, and J.-C. Gay, J. Opt. Soc. Am. B **12**, 505 (1995).
[13] R. Blümel, A. Buchleitner, R. Graham, L. Sirko, U. Smilansky, and H. Walther, Phys. Rev. A **44**, 4521 (1991).
[14] R. Blümel, J. Goldberg, and U. Smilansky, Z. Phys. D **9**, 95 (1989).
[15] J. Zakrzewski, D. Delande, and M. Kuś, Phys. Rev. E **47**, 1665 (1993).
[16] J. Zakrzewski and D. Delande (to be published).
[17] J. E. Howard, Phys. Rev. A **46**, 364 (1992); R. Gębarowski and J. Zakrzewski, Phys. Rev. A **51**, 1508 (1995).