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Excitation mechanisms for hydrogen atoms in strong microwave fields

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Abstract. — We analyse the excitation mechanisms which determine the dynamics of highly excited hydrogen atoms traversing a waveguide. Experimental measurements to verify predictions derived from Floquet theory are suggested.

1. Introduction.

Rydberg atoms exposed to short, strong microwave pulses [1-4] provide a particularly attractive possibility to study short-time phenomena in quantum mechanics. In such experiments the amplitude of the microwave field can be adjusted to be of the same order of magnitude as the Coulomb field binding the electron, therefore, a major theoretical task is the development of new, non-perturbative methods. On the experimental side essential data required for a thorough comparison of theoretical predictions and experimental results such as length and shape of the pulse can be determined with great precision. Thus, there is a unique opportunity to unravel the mechanisms governing the quantum dynamics of systems in strong periodic fields; insights gained from an analysis of the microwave problem should also throw light on the similar problem of atoms interacting with strong laser pulses.

In a recent work [5], we have applied the Floquet picture to the description of microwave ionization experiments performed at high scaled frequencies $n^3 \omega \gg 1$, using cavities [2]: an atom experiences, when entering the cavity, a slowly rising field which leads to diabatic-adiabatic motion on coarse-grained quasienergy surfaces such that the state that finally ends up in the cavity is a superposition of Floquet states. The results of the ionization experiments performed in the cavity are determined by the amplitudes of these Floquet states which, in turn, reflect both the structure of the underlying quasienergy spectrum and the shape of the fringe field [5].

The concept of adiabatic motion is, of course, of central importance in many areas of physics. For instance, within the framework of the Born-Oppenheimer theory in molecular physics one encounters adiabatic evolution on energy surfaces; at avoided crossings of energy levels Landau-Zener transitions occur. For periodically time-dependent quantum systems such as atoms in strong laser fields or Rydberg atoms in microwave fields, there is adiabatic evolution of Floquet states on quasienergy surfaces [5, 6]. Now an important new point appears: The quasienergy spectrum is organized in Brillouin zones. This fact leads to avoided
crossings of quasenergies corresponding to « distant » states, Landau-Zener transitions among Floquet states, therefore, open the possibility of exciting high lying states with large probability. Thus, in the case of Rydberg atoms interacting with a pulsed microwave field there is a variety of processes which are induced by the pulse shape, i.e. by the parameter motion and the resulting adiabatic or diabatic evolution of the wave function. Obviously such effects can not be understood by an approach [7] which simply assumes a temporally constant microwave amplitude.

In the present paper we will discuss the question which experimental signatures detectable in present-day experiments can be employed for an unambiguous test of predictions derived from the Floquet theory. We will, therefore, analyse a paradigmatic type of experiment: we assume a beam of fast, excited hydrogen atoms to traverse a rectangular waveguide operated in the TE_{10} mode; thus, the envelope of the microwave pulse « seen » by the atoms is assumed to be half a sine wave, modified by the presence of the entrance holes in the waveguide.

Whereas in cavity experiments the pulse shape has a « flat top », in the situation considered by us the amplitude varies during the whole interaction time and it should be clear immediately that experimental results obtained with both types of experimental set-up can differ significantly even though the maximal amplitude and the frequency employed may be the same.

We will proceed in two steps: in the following chapter we present model calculations which illustrate the excitation mechanisms for the type of experiment under consideration. Seen in a more general context, these calculations serve to establish the fact that adiabatic motion on quasenergy surfaces is of central importance for the dynamics of periodically driven quantum systems. We then turn to the question of experimental consequences and suggest measurements which could serve as a test of the viability of the Floquet picture.

2. Excitation mechanisms for atoms in microwave fields.

For our model calculations we use the (1+1)-dimensional Hamiltonian

\[ H = H_0 + H_{\text{int}} \]

(2.1)

describing a « 1-dimensional hydrogen atom »

\[ H_0 = \frac{1}{2} p^2 + \begin{cases} -\frac{1}{x^2}, & x > 0 \\ +\infty, & x \leq 0 \end{cases} \]

(2.2)

interacting with a linearly polarized microwave field

\[ H_{\text{int}} = \lambda(t) x \sin \omega t, \]

(2.3)

where \( \lambda(t) \) denotes the time-dependent amplitude of the field and \( \omega = 2\pi/T \) its frequency. We denote stationary eigenstates of \( H_0 \) by \( \varphi_n(x) \), \( n \) being the principal quantum number, and the Floquet states of the full Hamiltonian (2.1) by \( u_n(x, t) \) with quasienergy \( \epsilon_n \).

Actual experiments done with real, three-dimensional hydrogen atoms are often performed in the presence of a static electric field. The effect of such a field is obvious: the quasienergy Hamiltonian of the three-dimensional hydrogen atom in a microwave field of constant amplitude is invariant with respect to the symmetry operation

\[ S = \begin{cases} x \rightarrow -x \\ t \rightarrow t + \frac{T}{2} \end{cases} \]

(2.4)
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This symmetry is broken by an additional static field. As is well known, symmetry is essential in the theory of level crossing; the static field thus not only leads to a distortion of the quasienergy surfaces but also to the appearance of further avoided crossings of quasienergies.

In the one-dimensional model, the situation is different: one could, of course, include a term simulating a static field of strength $\lambda_s$ in the model (2.1) and write

$$H' = H_0 + \lambda_s x \sin \omega t - \lambda_s x.$$  

However, the $(1+1)$-dimensional quasienergy Hamiltonian is lacking the symmetry $S$ even without the term $-\lambda_s x$. Therefore, this term does now not give rise to new anticrossings in the quasienergy spectrum but only to a Stark-shift of the individual levels. Furthermore, the basic mechanisms we want to illustrate — effectively adiabatic evolution on smooth quasienergy surfaces, Landau-Zener transitions at reactive avoided crossings — remain unaltered. In order to keep the model as simple as possible we therefore drop the term $-\lambda_s x$ and employ the Hamiltonian (2.1) for our numerical analysis.

All calculations are performed in a basis of 101 bound states of $H_0$ ($n_{\text{min}} = 40$, $n_{\text{max}} = 140$). We first compute the quasienergy spectra for three different frequencies $\omega/2\pi = 13.00$ GHz (Fig. 1), 15.51 GHz (Fig. 2) and 18.00 GHz (Fig. 3). When displaying quasienergies graphically, a technical difficulty occurs: a quasienergy $\epsilon_n$ actually is a class

$$\{\epsilon_{nm} = \epsilon_n + m\omega | m \in \mathbb{Z}\}$$

Fig. 1.—Part of the quasienergy spectrum for $\omega/2\pi = 13.00$ GHz. Quasienergies are labelled by principal quantum numbers corresponding to the static Hamiltonian $H_0$. 

\[\text{Fig. 1.} - \text{Part of the quasienergy spectrum for } \omega/2\pi = 13.00 \text{ GHz. Quasienergies are labelled by principal quantum numbers corresponding to the static Hamiltonian } H_0.\]
Fig 2.— Part of the quasienergy spectrum for $\omega/2 \pi = 15.51$ GHz

of physically equivalent representatives $\epsilon_{nm}$, the first Brillouin zone

$$\frac{-1}{2} \leq \frac{\epsilon}{\omega} < \frac{1}{2}$$

hence, contains one representative of every class. An attempt to display all levels contained in one Brillouin zone would obviously result in a black picture, no structures would be recognizable. One therefore has to select from all the levels certain dynamically relevant ones. We do this by plotting only those quasienergies for which the corresponding Floquet states are concentrated in a certain subspace of the full Hilbert space. For figures 1 and 2, this subspace is spanned by eigenfunctions $\{\varphi_{55}, \ldots, \varphi_{85}\}$ of the time-independent system (2.2), for figure 3 by $\{\varphi_{60}, \ldots, \varphi_{80}\}$. Thus, a vanishing line in the quasienergy spectra simply indicates that the Floquet state is mainly contained in a different subspace of the Hilbert space.

The approximation actually implicit in our calculation is that we neglect the imaginary part of the quasienergies which determines the decay properties of the Floquet states [5]. However, since the aim of the present paper is to investigate excitation mechanisms, we will in the following part only consider examples with parameters well below the ionization border.

For the frequencies considered in figures 1, 2 and 3, the levels closest to the main resonance $n^3 \omega = 1$ are $n = 80$, 75 and 72, respectively. In all three cases we observe the same strong
deformation of the group of levels adjacent to the resonant one, the resonant level itself exhibits large avoided crossings only for comparatively high field strengths and thus enables purely adiabatic motion even for relatively large amplitudes. This fact leads to a quantum mechanical explanation [5] of the local stability at $n^2 \omega = 1$ observed in actual experiments [2].

In the following we will use these spectra to explain the behaviour of solutions of the time-dependent Schrödinger equation

$$H \psi(t) = i \partial_t \psi(t).$$

In order to model the pulse shape seen by atoms traversing a waveguide we choose

$$\lambda(t) = \lambda_{\text{max}} \sin \left( \frac{\pi t}{t_p} \right), \quad 0 \leq t \leq t_p,$$

where $t_p$ denotes the interaction time. As initial states at $t = 0$ we take the eigenstates $\varphi_{n_0}$ of $H_0$.

As our calculations show, basically three different mechanisms governing the short-time behaviour have to be distinguished: adiabatic motion (a), Landau-Zener transitions (b) and transitions due to near degeneracies at vanishing field strength (c).
a) To give an example for adiabatic motion we choose $n_0 = 72$, $\omega/2 = 13.00$ GHz, $t_p = 98$ T, $\lambda_{\text{max}} = 3.44$ V/cm (in atomic units, the scaled parameters are: $n_0^4 \lambda_{\text{max}} = 0.018$, $n_0^3 \omega = 0.737$). We decompose the Schrödinger wave function evolving from the initial state $\varphi_{72}$ with respect to the eigenstates $\varphi_i$ of $H_0$

$$\psi(x, t) = \sum c_i(t) \varphi_i(x)$$  \hspace{1cm} (2.8)

at time $t = t_p/2$, when the maximal field amplitude is reached (Fig. 4), and at $t = t_p$, i.e. at the end of the pulse (Fig. 5). Although several states $\varphi_i$ are excited in the middle of the pulse, after the pulse practically all probability is again concentrated on the initial state. The explanation for this non-diffusive behaviour is obvious from the quasienergy spectrum in figure 1: since the quasienergy surface $e_{72}$ is smooth in the relevant interval of field strength, i.e. does not show any reactive avoided crossing [5], we conclude that the time evolution is almost adiabatic. This means that the wave function $\psi$ is described by a single Floquet state $u_{72}$ at every instant of time. Thus, from the point of view of Floquet theory only a single state is excited during the whole process; this state «moves» adiabatically on its quasienergy surface $e_{72}$ in response to the continuously changing amplitude. However, the naive decomposition (2.8) for $t = t_p/2$ obscures this simple behaviour.

b) We now take $\varphi_{72}$ as initial state, $\omega/2 = 15.51$ GHz, $\lambda_{\text{max}} = 4.0$ V/cm [$n_0^4 \lambda_{\text{max}} = 0.021$, $n_0^3 \omega = 0.880$]. Again we decompose the wave function according to (2.8) after the pulse, this time, however, we plot the coefficients $|c_i|^2$ as functions of the pulse

![Fig 4](image1.png)  \hspace{1cm} ![Fig 5](image2.png)

Fig. 4 — Decomposition with respect to static states (2.8) of the wave function which evolves from the initial state $\varphi_{72}$ under the influence of the pulse (2.3), (2.7) for $\omega/2 = 13.00$ GHz, $\lambda_{\text{max}} = 3.44$ V/cm, $t_p = 98$ T, the decomposition has been performed at $t = t_p/2$ when the maximal amplitude was reached. This wave function is an almost pure Floquet state.

Fig. 5 — Decomposition of the wavefunction evolving from $\varphi_{72}$ after the pulse at $t = t_p$, the parameters used are the same as in figure 4.
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Fig. 6 — Occupation probabilities of the bound states \( n = 72 \) (upper full line), 78 (lower full line), 79 (larger dashes) and 84 (short dashes) after the pulse as a function of the pulse length \( t_p \) in units of \( T = 2 \pi / \omega \). The initial state was \( n_0 = 72 \); final states other than the ones displayed are of minor importance. The parameters are \( \omega / 2 \pi = 15.51 \) GHz, \( \lambda_{\text{max}} = 4.00 \) V/cm.

Fig. 7 — Part of the quasienergy spectrum relevant for the dynamics of the initial state \( n_0 = 72 \) at \( \omega / 2 \pi = 15.51 \) GHz (cf. Fig. 6), the straight lines indicate Landau-Zener transitions.

The resulting pattern (Fig. 6) has a clear explanation from the quasienergy spectrum in figure 7: the initial state \( n_0 = 72 \) is shifted into the connected Floquet state \( u_{72} \) and undergoes two Landau-Zener transitions to \( u_{78} \) and \( u_{79} \) during the rise of the field, when the amplitude decreases again interferences occur leading to Stückelberg oscillations visible in the excitation probabilities of the final states. In order to observe these oscillations experimentally one has to vary the interaction time \( t_p \), which up to now has not been done in a systematic manner.

c) As an example for the third mechanism we take \( n_0 = 72, \omega / 2 \pi = 18.00 \) GHz, \( \lambda_{\text{max}} = 6.5 \) V/cm \( [n_0, \lambda_{\text{max}} = 0.034, \ n_0 \omega = 1.021 ] \) and display in figure 8a), .. f) the excitation probabilities \( |c_i|^2 \) of the final bound states as functions of \( t_p \). The intricate oscillation patterns can be understood from the underlying quasienergy spectrum (Fig. 3). Now the initial state is very close to resonance, i.e. several quasienergies are nearly degenerate in the Brillouin-zone at \( \lambda = 0 \). This near degeneracy is rapidly removed for increasing amplitude. Thus, right at the beginning of the pulse the initial state is distributed over several Floquet states which then decouple and evolve on their own quasienergy surface. At the end of the pulse the Floquet states couple again when \( \lambda \approx 0 \) and the resulting interference pattern is determined by the dynamical phases each Floquet state has accumulated.
Fig 8a — Occupation probability of the final bound state $n = 69$ after the pulse, the initial state was $n_0 = 72$. Parameters $\omega/2\pi = 18.00$ GHz, $\lambda_{\text{max}} = 6.5$ V/cm

Fig 8b — As figure 8a for the final state $n = 70$.

Fig. 8c — As figure 8a for the final state $n = 71$.

Fig 8d — As figure 8a for the final state $n = 72$
We finish this section with two remarks concerning the important differences between mechanism b) and c).

The transitions in b) are due to avoided crossings at $\lambda > 0$ which couple states differing by several units in principal quantum number; in our example we had transitions $72 \rightarrow 78, 79$ corresponding to an energy difference of $6.05 \hbar \omega$ and $6.93 \hbar \omega$, respectively. In contrast, the near degeneracy at $\lambda = 0$ in c) perturbatively couples states which are the nearest neighbours of the initial one.

Secondly we note that mechanism c) depends very sensitively on the way the field is turned on and off. We have checked this by smoothing the sinusoidal amplitude (2.7) at the edges such that an exponential function is fitted continuously differentiable at times $t_f$ and $t_p - t_f (t_f \ll t_p)$ of the original pulse. Two examples for the influence of this modification are given in figures 9 and 10 ($t_f = 3 \, \text{T and } t_f = 5 \, \text{T}$). This dependence on details of the pulse shape is not surprising since the pulse is modified exactly when the Floquet states interact, i.e. near $\lambda = 0$. In contrast, as we have checked the same modification is of negligible influence for a) and b).

3. Experimental implications.

We saw in the previous chapter that the Floquet representation offers a very convenient and efficient tool to analyse the dynamics of atoms in pulsed fields. A process that appears to be complicated in the eigenbasis of $H_0$ can be very simple when analysed in the Floquet basis, our mechanism a) being a convincing example for this fact. As a measure of the complexity of a process during the interaction we define the quantity

$$S(t) = - \sum p_i \ln p_i,$$

(3.1)
Fig. 9 — Occupation probabilities of final bound states after a pulse which slightly differs from (2.7). The edges of the pulse have been smoothed such that at \( t = t_r \) and \( t = t_p - t_r \), an exponential function has been fitted continuously differentiable to the original amplitude. For this figure, \( t_r = 3 \text{T} \) and \( t_p = 135 \text{T} \), the other parameters are as in figure 8.

Fig 10 — As figure 9, however \( t_r = 5 \text{T} \)

where

\[
p_i = p_i(t) = \left| \langle u_i^A(t) | \psi \rangle \right|^2 , \tag{3.2}
\]

\( \psi \) is the Schrödinger wave function and the \( u_i^A(t) \) denote the Floquet states corresponding to the instantaneous field strength \( \lambda(t) \). Compared to the «Shannon entropy» recently considered by Casati et al. [8] to characterize localization properties of eigenfunctions, the complexity (3.1) is a measure of the dimension of the space of effective Floquet states involved in the dynamics. For a purely adiabatic process \( S(t) \) stays constant and increases when non-adiabatic phenomena occur, e.g. when the wave function « splits » due to Landau-Zener transitions; if only a single Floquet state is excited in a purely adiabatic process (mechanism a) \( S(t) \) remains identically zero. This fact again nicely illustrates that the « stationary » Floquet states provide the appropriate basis for a description of the dynamics: if in the definition (3.2) of the probabilities \( p_i(t) \) the Floquet states \( u_i \) were replaced by static eigenstates \( \varphi_i \), one would obtain the misleading result that the complexity of an adiabatic process grows during the rise of the pulse and then decreases to its original value. As a striking example for this we show in figures 11, 12 a static state decomposition of the wave function for an adiabatic process. Even though the initial static state is almost completely depopulated in the middle of the pulse this process is adiabatic, i.e. the above defined complexity remains zero for the whole pulse.

The observables in actual excitation experiments performed with waveguides are the occupation probabilities of the final static bound states \( \varphi_i \), after the interaction with the
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Decomposition of the wave function which has evolved from the initial state $n_0 = 66$ in the middle of the pulse, $\omega/2 \pi = 18.00 \, \text{GHz}$, $\lambda_{\text{max}} = 60 \, \text{V/cm}$, $t_p = 136 \, \text{T}$. Note that the initial static state seems to be « depopulated »

Fig. 11. — Decomposition of the wave function considered in figure 11 after the pulse

Microwave field and the challenge now is to detect clear evidence for the above described excitation mechanisms. Of course one faces several difficulties: experimental results may be modified by the presence of noise [4] and an accurate final bound state analysis is complicated because of the high degeneracy in real hydrogen atoms. The latter problem could partially be circumvented by working with helium [9] or alkali [3, 4] atoms in order to lift the $\ell$-degeneracy or, as a different possibility, by superimposing a static electric field to achieve quasi one-dimensionality. Despite these complications, we believe that skillfully performed experiments can unambiguously identify the basic mechanisms. To this end we suggest the following measurements:

1) The interaction time $t_p$ can be varied by changing the velocity of the atom beam. Landau-Zener transition probabilities during the pulse do depend on $t_p$ and determine the final bound state distribution. Thus, measuring the $|c_i|^2$ as functions of $t_p$ yields important information about transitions occurring during the pulse. In particular, waveguide experiments are ideally suited for a detection of St"uckelberg oscillations: if the length of the whole pulse is adjusted to be of the order of only 100 cycles of the external field, the dynamical phases acquired by the interfering Floquet states can be prevented from growing too large; in contrast, the flat top of the pulse in cavity experiments [2] lasts longer than 300 cycles and thus induces large phases and a correspondingly sharper interference pattern which is harder to resolve. In order to resolve St"uckelberg oscillations as depicted in figure 6 in such waveguide experiments, the necessary range of velocities translates into a variation of the proton acceleration voltage over a range of some 10 keV.
2) The holes by which the atoms enter and leave the waveguide modify the pulse shape near \( t = 0 \) and \( t = t_p \) such that the actual envelope obtains long tails. It is well known [10] that smoothness conditions at the edges of the pulse are essential for adiabatic behaviour; indeed, as our calculations have shown, for near resonant initial conditions (mechanism c) the \( |c_i|^2 (t_p) \) can be influenced by shape and magnitude of the holes, whereas in case a) and b) properties of the holes have only a negligible effect. Hence, repeating the experiments using waveguides with differently shaped holes will, in general, yield identical results, however, for certain initial states the experimental signal will be characteristic for the microscopic structure of the holes of the particular apparatus employed.

3) In order to investigate pulse-shape dependencies it would be very interesting to operate the waveguide in higher modes, e.g. in the \( \text{TE}_{20} \)-mode which leads to a double pulse; the Floquet states involved thus interfere several times during the interaction which may drastically alter the final bound state distribution.

Of particular importance is an experimental verification of mechanism b), i.e. an excitation of bound states considerably higher than the initial one induced by Landau-Zener transitions at broad avoided crossings. The very same phenomenon occurring for atoms irradiated by short strong laser pulses explains transitions requiring an excitation energy of several \( \hbar \omega \).

We would like to remark that in actual experiments performed in the presence of a static electric field there is an additional possibility of controlling the system: The «scaled frequency» (that is, the ratio of the microwave frequency and the angular velocity of the corresponding classical system for vanishing microwave amplitude) is then no longer given by \( n_0^3 \omega \) but rather becomes a function of the strength of the static field; furthermore, the structure of the Brillouin zone at \( \Lambda = 0 \) will be determined by the energies of the Stark states. Thus, it is an interesting question whether it is possible to observe dynamical behaviour governed by different mechanisms when changing the strength of the static field.

Summarizing, we have theoretically discussed microwave excitation experiments on highly excited hydrogen atoms which are characterized by a continuous change of the amplitude during the interaction time. As we have pointed out, such a time-dependence of the field strength makes the use of adiabatic methods indispensable for a transparent description. We conclude that waveguide experiments could serve as a stringent test for Floquet dynamics when the final bound state distribution is measured as a function of pulse-shape parameters. In this way, microwave experiments could yield important insights into short-time phenomena in strongly interacting quantum systems.

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