Half-Cycle Microwave Excitation of Rubidium Rydberg Atoms

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(Received 2 May 1994)

Excitation from *ns*, *np*, and *nd* Rydberg states of rubidium to the adjacent manifolds by electrical pulses of 140 and 9 ns duration up to 200 V/cm is studied. These pulses correspond to half cycles of microwave fields of 2.6 GHz and 40 MHz. The observed population transfer (up to 40%) is not monotonically increasing with the field strength. For the 140 ps pulse local minima are found due to the absence of avoided crossings of appropriate size at the peak of the pulse. For the long pulses Stueckelberg oscillations are observed in the transfer yield.

PACS numbers: 32.80.Rm

Microwave ionization of Rydberg atoms has been studied for more than a decade [1–4]. Over a large range of frequencies, it has been observed [5] that almost complete ionization occurs once the amplitude of the microwave field surpasses the static electric field value at which the adjacent Stark manifolds intersect: $F = 1/3n^5$ (in atomic units). In nonhydrogenic Rydberg states the bluest member of the n Stark manifold couples with the reddest $n + 1$ Stark state, resulting in an avoided crossing of these two states. When the microwave field amplitude is large enough to reach such an avoided crossing Landau-Zener transitions can occur: population is transferred from the initial n state to the next n state. During a half cycle of the pulse the crossing is passed twice. The transfer yield depends critically on the slew rate of the electric field with respect to the coupling of the two states. No population is transferred for very high slew rates: the crossing is passed completely diabatically twice. For very low slew rates the crossing is passed adiabatically twice, and again no population is transferred. Only at intermediate slew rates is population transferred. In this case subsequent cycles of the microwave field can drive population to higher Rydberg states, eventually leading to ionization [3]. In most microwave experiments on Rydberg atoms the radiation lasts for a few thousand cycles (e.g., a 0.5 μs pulse at 8 GHz corresponds to 4000 cycles). Still, the above mentioned interpretation of these experiments is based on a model [3] in which the ionization process is considered as resulting from a sequence of incoherent excitation steps by individual half cycles of the microwave field. A number of questions concerning the excitation dynamics is not answered by these many-cycle experiments. For instance, does the population of the manifold states occur at the first intersection of the initial state with this manifold? How large is the actual population transfer per cycle at these avoided crossings? Also, since the crossings are passed many times, the role of coherence on these excitation processes can be relevant. If the excitation amplitude adds coherently during the pulse, the number of cycles needed to drive the population from the initial state to high lying states is strongly reduced [5]. Recently it has been demonstrated [6] that the climbing (ionization) process is frustrated if the microwave pulse is only 25 cycles long. For complete ionization a higher field strength is required for these short pulses. In another study using short microwave pulses [7] it has been found that population is left in high lying Rydberg states.

To shed light on these questions we report in this Letter on experiments in which the electric field is only on for half a cycle. Transitions from *ns*, *np*, and *nd* states of rubidium to the adjacent manifolds are studied for electrical pulses of 140 and 9 ns duration. The upper halves of these pulses resemble a sine-wave field with frequencies of 2.6 GHz and 40 MHz, respectively. Figure 1 shows the Rb Stark map of the $n = 30$, $n = 31$, $m_j = 1/2$ Stark manifolds and the adjacent *ns*, *np*, and *nd* levels as a function of the static electric field. The size of the avoided crossings around $n = 30$ range from 10^{-3} to 10^{-1} cm^{-1} (30 MHz to 3 GHz). At a field of 18 V/cm the first intersection of the 34*s* with the $n = 31$ manifold is encountered (point B in Fig. 1). However, the coupling of the 34*s* state with these manifold states is small since the manifold states have little p character at these field strengths. Consequently, the size of these avoided crossings is small (0.0014 cm^{-1} at the first intersection). The intersection of the 33*p* with the manifold occurs at 45 V/cm. At this field strength the s state has mixed with the manifold and consequently the size of the avoided crossing is much larger: 0.06 cm^{-1}. The application of 140 and 9 ns pulses allows us to investigate the dynamics of both limiting regimes, i.e., diabatic and adiabatic passages, as well as the intermediate regime. We found that, depending on the pulse amplitude, the population transfer to the manifold can be as high as 40% for a half cycle, but is not monotonically increasing as a function of the field strength. Two passages of the same avoided crossing on the rising and falling edge of the pulse are observed as Stueckelberg oscillations in the transfer yield, demonstrating the importance of coherence in microwave excitation.

In a vacuum system (10^{-6} Pa) the rubidium atoms coming out of an oven are excited to a particular Rydberg state by a nanosecond dye laser operating at 10 Hz.

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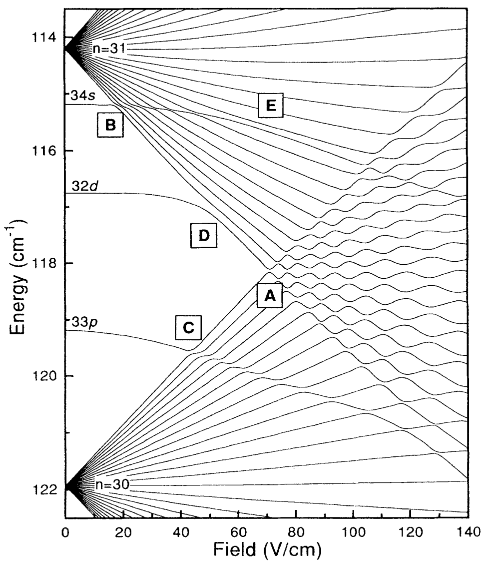

FIG. 1. The rubidium Stark levels as a function of the electric field. Shown are the $n = 30$, $n = 31$ manifolds and the adjacent *ns*, *np*, and *nd* levels. Point *A* depicts the crossing of the bluest member of the *n* Stark manifold with the reddest member of the $n + 1$ Stark manifold scaling as $F = 1/3n^5$. Points *B*, *C*, and *D* depict the first crossing of the *ns*, *np*, and *nd* state with the manifold, respectively.

Starting from the 5*s* ground state an *ns* or *nd* Rydberg state is populated by two-photon excitation. For the one-photon excitation of the *np* states the output of the dye laser is frequency doubled.

The Rydberg atoms are created in between two condensor plates separated by 5 mm. After laser excitation the half-cycle electric pulse is fed to the plates by a rigid line cable. Great care is taken to match the termination impedance of the condensor plates to the rigid line to minimize reflections of the pulse. The actual amplitude and shape of the pulse are measured *in situ* by a high-frequency probe and a sampling oscilloscope. The 140 ps (FWHM), 120 V pulses are produced by a Kentech Instruments APG1 pulse generator. The pulse is attenuated using broadband variable attenuators. The 9 ns (FWHM), 100 V pulses are produced by a HP 8114A programmable pulse generator. By means of state-selective field ionization the population of both the initial Rydberg state and the other Rydberg states, in particular those of the adjacent Stark manifold, are monitored.

The laser polarization is chosen parallel to the electric field, so only $m_j = 1/2$ states are populated during excitation. First we will present the results with the 140 ps pulse. In Fig. 2(a) the observed population tranfer from the 34*s* state to the adjacent manifold as a function of the pulse amplitude is shown. At the first crossing (point *B* in Fig. 1) no net population is transferred to the manifold because the avoided crossings are small (discussed above) and therefore are passed fully diabatically on the rising and the falling edge of the pulse. First at a field amplitude of 70 V/cm a crossing is encountered that can be passed partially adiabatically, thereby making it possible to transfer population to the manifold states. As can be seen in Fig. 2 the transfer yield is not increasing monotonically with the pulse amplitude, but shows a local minimum. At a field strength of 70 V/cm the large avoided crossing (point *E* in Fig. 1) efficiently transfers net population to the manifold states. At higher pulse amplitudes this crossing is traversed more diabatically caused by the higher slew rate at the edge of the pulse. The loss of efficiency cannot be compensated by the avoided crossings that are encountered at 110 V/cm. At even higher pulse amplitudes the number of crossings that is encounterd becomes so large that returning to the 34*s* state becomes less probable. Starting from the 32*d* state, the first avoided crossing (point *D* in Fig. 1) is traversed adiabatically twice, giving no population transfer to the manifold. When the field strength reaches point *A* in Fig. 1, a partly diabatic transition becomes possible, populating the *k* states of the Stark manifolds. The decrease in transfer yield to the manifold at higher fields cannot easily be understood since many crossings are involved. We interpret this as population transfer to the 33*p* state instead of the $n = 30$, 31 manifolds. In these measurements no discrimination between the 32*d* and 33*p* states can be made. Since the amplitude of the pulse exceeds point *A* in Fig. 1, a trajectory is opened leading to population of the 33*p* state on the falling edge of the pulse.

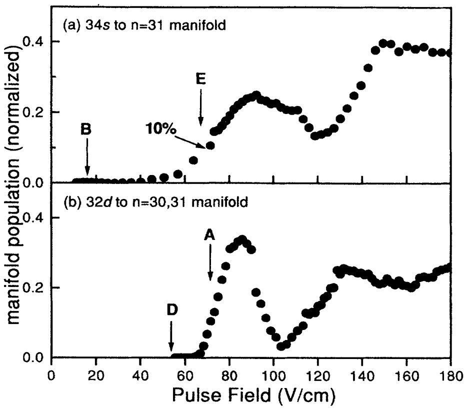

FIG. 2. (a) Measured population transfer of the Rb 34*s* (a) and 32*d* (b) states to the $n = 31$, 30 manifolds after excitation with a half-cycle pulse of 140 ps as a function of the maximum field amplitude.

In Fig. 3 the values of the electric field strength are plotted at which 10% population is transferred to the adjacent manifold for *ns*, *np*, and *nd* as a function of effective quantum number n^*. The local minima always lie above the 10% limit, so the first rising edge determines the threshold field, as marked in Fig. 2(a). n^* is determined according to the quantum defects as $n^* = n - 4$ for the *ns* states, $n^* = n - 3$ for the *np* states,

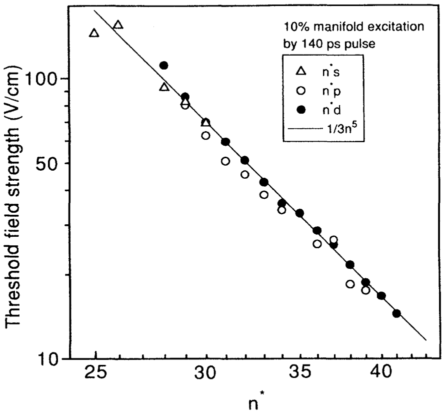

FIG. 3. Measured threshold fields for which 10% of the initial *ns*, *np*, or *nd* population is transferred to the manifold by the 140 ps pulse as a function of n^*.

and $n^* = n - 2$ for the *nd* states. The line is given by $F = 1/3(n^*)^5$ which denotes the n^*, $(n+1)^*$ manifold crossing (point *A* in Fig. 1). The measured threshold fields for the *nd* states lie on this line, confirming that the first intersection of the *nd* state with the manifold is traversed adiabatically. Starting from the *ns* or *np* state the 10% transfer yield also lies at a higher field than the first intersection of these states with the Stark manifold. In contrast to the *nd* states, which are in the adiabatic regime at the first crossings, the first crossings with the manifold are passed diabatically for the *ns* and *np* states.

For the long 9 ns pulse the first crossings of the 36*s* with the manifold states can be passed partially adiabatic in contrast to the short 180 ps pulse, so population can be transferred to the manifold at these crossings. Results are shown in Fig. 4(a). The points I, II, and III denote the crossings of the 36*s* state with the three reddest $n = 33$ manifold states. Although the crossings are passed almost diabatically on the rising edge of the pulse, at the top of the pulse the field changes slowly enough to make a partly adiabatic transition possible resulting in the main peaks in Fig. 4. Following these main peaks oscillations in the transfer yield are observed. We interpret this as follows: When the pulse amplitude exceeds the position of the first crossing there will be amplitude in the adiabatic as well as the diabatic path. The total wave function consists now of two states evolving with different frequency. When the crossing is passed on the falling edge of the pulse these two states have built up a phase difference leading to interference. This phase difference is given by $\Delta\Phi = \int_{t_0}^{t_1} \Delta E(t)\,dt$, where $\Delta E(t)$ is the energy difference between the two levels and t_0, t_1 are the times at which the crossing is traversed. For $\Delta\Phi = N2\pi$ (N integer) the transfer to the manifold is most efficient (constructive interference), while for $\Delta\Phi = (2N+1)\pi$ it is negligible (destructive interference). These so-called Stueckelberg oscillations decrease rapidly with the amplitude of the pulse because the slew rate at the crossing becomes too fast for an adiabatic transition. Note that the origin of these oscillations differs from the recently reported Stueckelberg oscillations in many-cycle pulse microwave excitation [4]. In the present experiment the crossing is passed twice during a half cycle of the microwaves, giving rise to the interference, whereas in the earlier reported experiments the cycle-averaged microwave power shifted a state into resonance on the rising and falling edge of the pulse envelope. The Stueckelberg oscillations demonstrate the relevance of coherence in the excitation of Rydberg states by microwave radiation. The coherent nature of the excitation gives for many-cycle pulses rise to multiphoton resonances [8]. The evolution from Stueckelberg oscillations to multiphoton resonances has been discussed elsewhere [9].

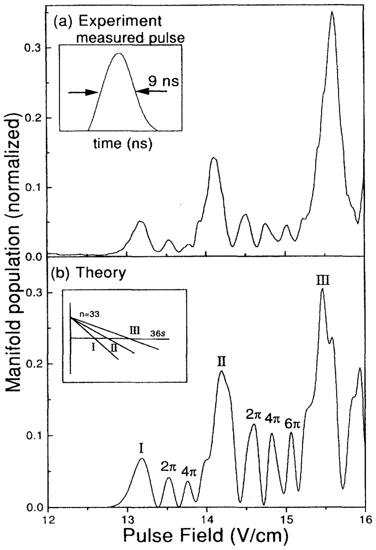

FIG. 4. (a) Measured population transfer of the Rb 36*s* state to the $n = 33$ manifold after excitation with a half-cycle pulse of 9 ns as a function of the maximum field amplitude. (b) Results of numerical integration of the time-dependent Schrödinger equation for the rubidium 36*s* state and the three reddest members of the $n = 33$ Stark manifold. The labels I, II, and III denote the crossings of the 34*s* with the *k* states of the $n = 33$ manifold. The oscillations following the large peaks are Stueckelberg oscillations. The inset schematically shows the relevant Stark levels.

In general the population transfer in a microwave excitation process cannot be calculated by solving the Schrödinger equation since a tremendous number of states are involved. Therefore, a quantitative comparison of theory and experiment is not possible. However, the population transfer from the 36*s* at the intersection with the reddest members of the $n = 33$ manifold has been calculated by means of numerical integration of the time-dependent Schrödinger equation since only seven states are involved. Since for rubidium only m_j is a good quantum number, the manifold is a mixture of states with $m_l = m_j \pm 1/2$. The $m_l = 0$ and $m_l = 1$ states of the same k have a small energy spacing: 10^{-2} cm^{-1}. The diabatic curves and couplings needed for this calculation are extracted from a program that calculates Stark states along the lines of Zimmerman *et al.* [10], using the rubidium quantum defects and fine structure parameters given by Ref. [11]. For this calculation seven states and six coupling constants are taken into account. The actual shape of the 9 ns pulse was digitized on a 500 MHz oscilloscope. Using this shape and the size of the avoided crossings, the net population transfer to the manifold states starting from the 36s state has been calculated. The results of the calculation are shown in Fig. 4(b). Since no free parameters are used for this calculation, the agreement with the experiment is very good. Both the net population transfer (vertical scale) and the position of the main peaks and following Stueckelberg oscillations are predicted well.

The half-cycle experiment indicates that for the calculation of many-cycle microwave ionization coherence has to be taken into account [12]. In the many-cycle pulse experiments the local decrease in transfer yield as shown in Fig. 2 probably cannot be observed since the yield is still high enough to transfer all the population after many microwave cycles. Note that for this half cycle of the microwave field, already 40% of the population is transferred to the adjacent manifold, indicating that the complete depletion of the initial state occurs in a few cycles. Jones, You, and Bucksbaum have recently studied the excitation of Rydberg atoms by a half-cycle far infrared pulse (1 THz) [13]. Whether the amplitude description suitable at the microwave frequencies reported here still holds in the far infrared is an open question.

In summary, we have investigated the population transfer from the Rb *ns*, *np*, and *nd* Rydberg states to the adjacent manifolds after exposure to a half-cycle pulse of 140 and 9 ns. For the 140 ps pulse the observed population transfer was not increasing monotonically, but shows a local minimum due to the absence of appropriate avoided crossings. The transfer yield can be as high as 40%, so only a few cycles are needed to reach ionization of the Rydberg state. For the 140 ps pulse the transfer yield is described using a Landau-Zener model. For the long pulse, however, coherence effects in the population transfer, observed as Stueckelberg oscillations, play a role. Population transfer to the manifold does not necessarily occur at the first intersection of the initial state with the manifold. Both fully diabatic (*ns*, *np*) and adiabatic (*nd*) passage leads to no net population transfer. Only if an avoided crossing of appropriate size with respect to the slew rate of the field is encountered population of the manifold states occurs.

It is a pleasure to acknowledge R. B. Vrijen for helping with the numerical integration code, T. F. Gallagher for fruitful discussion, and B. Broers, A. Lagendijk, and H. G. Muller for carefully reading the manuscript. This work is part of the research program of the "Stichting voor Fundamenteel Onderzoek der Materie (FOM)" which is financially supported by the "Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO)."

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