Half-cycle microwave stabilization of rubidium Stark atoms

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(Received 7 May 1999)

We report on the observation of stabilization of laser-excited rubidium Stark atoms above the classical ionization limit with the help of short (9 ns) unipolar electric-field pulses. Dipping the field down from 40 to 0 V/cm and back to 40 V/cm, 30 ns after photoexcitation into the continuum, yields a large bound-state population. The results suggest an additional mechanism of the recombination as seen in the THz experiment by Bensky, Campbell, and Jones, [Phys. Rev. Lett. **81**, 3112 (1998)]. [S1050-2947(99)10111-2]

PACS number(s): 32.80.Rm, 33.80.Rv

The behavior of Rydberg atoms in fast oscillating electric fields has been studied extensively by several groups [1–4]. In particular, the ionization behavior with microwave electric-field pulses has been of interest. For ionization of Rydberg atoms in a static field, the corresponding electric field F is given by $F=1/16n^4$, at microwave frequencies ionization already occurs at fields as low as $F=1/3n^5$ [3] (atomic units are used unless stated otherwise). In a microwave field, successive Landau-Zener crossings between a Stark state from a certain n manifold to a Stark state from one higher-lying n manifold causes the ionization to occur. Once in the continuum, successive Landau-Zener transitions can still occur within the lifetime of the continuum states (route A in Fig. 1) [5].

The question now rises of whether the inverse process can also occur: would it be possible to induce bound states with microwave fields starting out in a (static) field-induced continuum, and make a Landau-Zener crossing to more deeply bound states by half of a cycle of the microwave field (route B in Fig. 1)? And, if this is possible, does a microwave field play an important role when the electron is in the field induced continuum? Bensky, Campbell, and Jones [6] reported on similar ''recombination'' schemes using half-cycle pulses in the subpicosecond (terahertz) regime. In this paper we report on the remarkable finding that unipolar pulses in the nanosecond regime can actually be used to ''recombine'' an electron in the field-induced continuum with its parent ion. For the THz experiments reported in Ref. [6], the half-cycle pulse is much shorter than the Kepler orbit period, and the action of the pulse can be described as a momentum kick: $\Delta p=\int_{-\infty}^{\infty}F dt$. For the nanosecond field pulse reported here, the pulses are relatively slow, and a description in terms of level crossings seems to be more applicable. The study of schemes to recombine free particles is interesting from the point of view that an efficient way to recombine an ion with an electron has still not been found.

Schematically the experimental procedure we use is as follows (see also Fig. 2): first we excite gas-phase rubidium atoms with an 8-ns laser pulse above the classical saddle point; then we apply the unipolar pulse by which we hope to induce bound states, and finally we use pulsed field ionization (explained below) to determine the bound fraction. A thermal atomic rubidium beam (produced by a resistively heated oven) is pointed in between two capacitor plates, separated by 10.0 mm. The plate furthest away from the detector (a set of microsphere plates) is connected to a box that generates the voltage ramp for the pulsed-field ionization detection (-400 V in 500 ns) superimposed on an offset of -40 V. Pulsed-field ionization (PFI) [7] is a method which is used to determine how much bound population is present after a certain event. By increasing the electric field gradually, one can ionize a highly excited atom due to the suppression of the ionization limit by the static electric field. The plate closest to the detector is connected to the HP 8141*a* pulse generator, which produces the 9-ns (full width at half maximum) unipolar pulses, the amplitude of which could be tuned from 1.00 to 50.00 V. Through this plate a hole (diameter 10 mm) is drilled, which is covered by a grid to minimize the distortion on the electric field. The parameters varied accurately in this experiment were the frequency of the dye laser, which photoexcited rubidium atoms, and the delay of the field pulse relative to the laser pulse. In a nonresonant two-photon process, rubidium atoms are excited by means of a narrow-band ($\Delta\lambda\leqslant 0.2\,\mathrm{cm}^{-1}$) dye-laser pulse

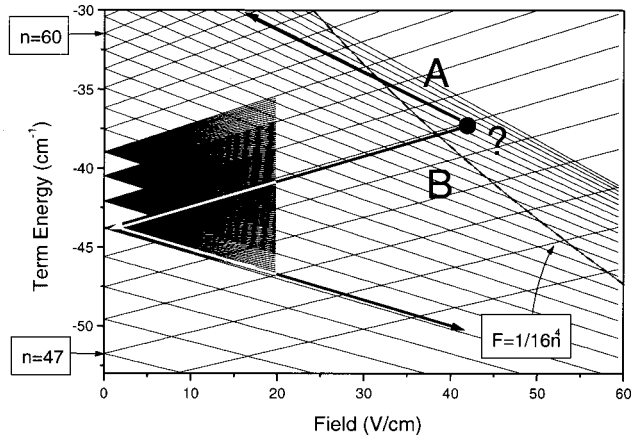

FIG. 1. Stark map of rubidium; only the lowest and highest k states of each n manifold are shown. The dotted line depicts the position of the classical ionization limit. In the experiments the atoms are photoexcited above the field-ionization-induced threshold, and after a fixed delay a unipolar field pulse is applied, which dips the field down and back up again within 9 ns. The question rises in what manner the ionizing states are affected: two trajectories (A and B) are suggested to play a role.

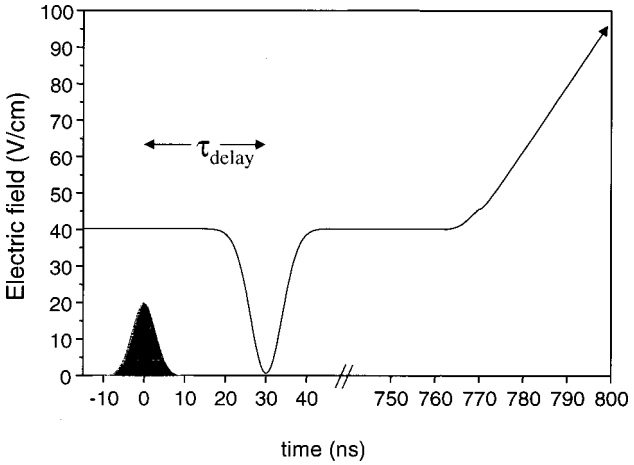

FIG. 2. Timing scheme of the experiment: at $t=0$ ns, a narrow-band laser pulse is used to excite Rydberg states in the vicinity of the saddle point in rubidium in a static field of 40 V/cm. After 30 ns the field is dipped down to zero field and back up again to 40 V/cm within 9 ns. After ~750 ns an electric-field ramp is applied to determine the fraction of the excited state population that is stabilized in a bound state.

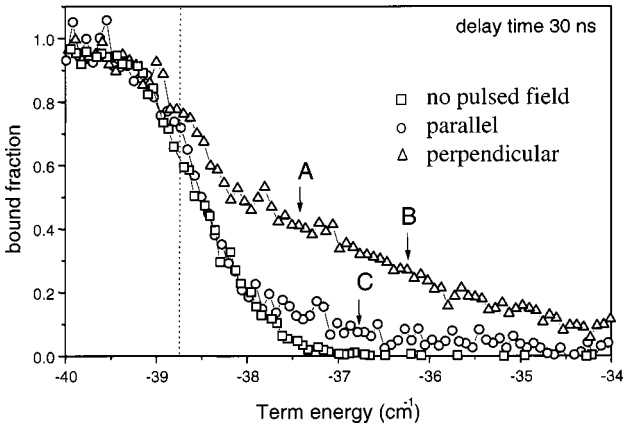

FIG. 3. Bound-state population as function of the frequency of the laser. The unipolar pulse was applied 30 ns after laser excitation. Both polarizations of the laser field with respect to the static electric field are shown. The open squares represents the integrated PFI signal as a function of the wavelength of the exciting laser when no unipolar field pulse is applied. The labels $A(E=-37.4\,\mathrm{cm}^{-1})$, $B(E=-36.2\,\mathrm{cm}^{-1})$, and $C(E=-36.8\,\mathrm{cm}^{-1})$ are markers which will be referred to later on. The vertical dotted line depicts where the classical ionization limit is at 40 V/cm.

above the classic ionization limit in a static field of 40 V/cm. Approximately 35 ns (variable) after excitation, the field is ramped down to 0 V/cm and back up to 40.0 V/cm within 9 ns. The output of this pulse generator was connected to a set of parallel capacitor plates (diameter 50 mm, thickness 1 mm), and care was taken to minimize ringing by matching the impedance of the two capacitor plates to the output of the pulse generator. Approximately 750 ns after excitation, the field is increased to 400 V/cm in 500 ns to field ionize the bound states created by the unipolar pulse.

In Fig. 3 the open triangles and circles show the recombined fraction, for perpendicular and parallel polarizations of the laser excitation with respect to the static electric field of 40 V/cm, when 30 ns after excitation we apply an unipolar pulse which dips the field down to 0 V/cm and back up again to 40 V/cm as a function of the term energy.

In a static electric field of 40.0 V/cm, the photoionization threshold is lowered by -38.7 cm^{-1}; therefore states with a principal quantum number of 54 or higher ionize, since they are above the field-induced saddle point in the modified Coulomb potential. In Fig. 3 we show the recorded bound-state signal as a function of the wavelength of the exciting dye laser in a static electric field of 40.0 V/cm without an electric-field kick (open squares). A sharp step is observed in the PFI signal as a function of the wavelength of the exciting laser. Also, the position of this step agrees with the theoretically predicted value of the threshold $\mathbf{E}=\mathbf{E}_0-1/16n^4$. However, when we ramp the field down and back up again after excitation with a laser pulse with a polarization perpendicular to the electric field, it appears that we are able to recombine free states to bound states. Figure 3 depicts this effect: the step-function shape at the position of the saddle point in the static electric field case totally diminishes when after excitation in a static electric field a unipolar pulse is applied, indicating that we are indeed able to induce bound states from states above the field-induced saddle point. Note that for the perpendicular case even for excitation of states 5 cm^{-1} above the saddle point, a bound fraction of at least 0.1 is still recorded. For amplitudes as low as -25 V/cm of the field kick, this ''recombination'' scheme occurs with roughly the same efficiency.

In terms of Landau-Zener crossing, the observed effects can only be understood if we follow path *B* in Fig. 1. This means that we are doing experiments in the diabatic regime, implying that the slew rate $(\Delta F/\Delta t)$ of our unipolar pulses is greater than the critical slew rate for a diabatic passage from a state i to a state j. This critical slew rate (S_{crit}) can be calculated with the help of the formula

$$S_{\mathrm{crit}}=\frac{\omega_0^2}{\partial(E_i-E_j)/\partial F}\propto\frac{(n^{-4})^2}{n^2}=n^{-10}$$

[8] where ω_0 is the size of the avoided crossing, and $\partial E_i/\partial F$ is the derivative of the term energy of a certain state as a function of the field. The coupling ω_0 can be written as $\omega_0=\delta_m/n^4$, where δ_m is the absolute value of the quantum defect δ_l (only taking into account the contribution from the lowest l state: $l=m$). If we introduce the factor S/S_{crit} (where S is the slew rate of the pulses used in our experiment), we find that in our case (at $n=55$) this factor is $S/S_{\mathrm{crit}}=4/0.1\,(\mathrm{V\,cm^{-1}\,ns^{-1}})=40\gg1$, which indeed places our experiments in the diabatic regime.

We would like to stress that the PFI signal clearly shows that we induce bound states from classically free states, and that the effects seen in the experiment are not due to lifetime enhancement of classically free states. However, we did observe that for amplitudes lower than 25 V/cm we were not able to induce bound states. Apparently for those amplitudes the slew rate is below S_{crit}, and no diabatic passage can be made. When we use light which is polarized parallel to the electric field, creating $m=0$, recombination is strongly reduced. Still there is 10% of the initial population which is being recombined at -37 and 2 cm^{-1} above the saddle point. The lifetime of the states photoexcited with parallel polarization is considerably shorter, and thus a smaller fraction sur-

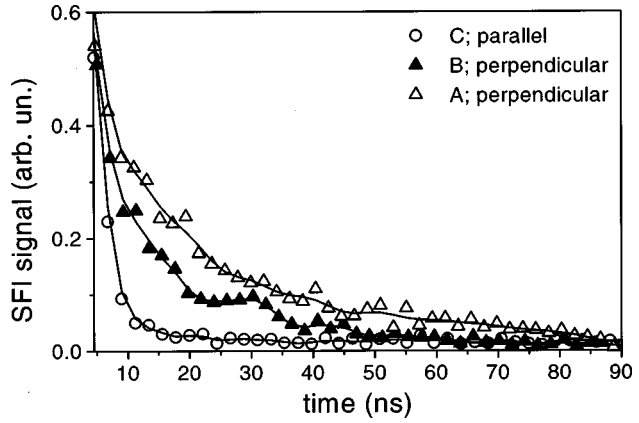

FIG. 4. The three traces show the bound-state population as a function of the time delay of the unipolar field pulse with respect to the ionizing laser pulse. The labels *A*, *B*, and *C* correspond to the markers in Fig. 3. Traces *A* and *B* represent data taken with perpendicular polarization, while trace *C* represents data taken with parallel polarization.

vives the 35-ns waiting time (e.g., Broers *et al.* [9]). In addition, the $m=0$ character induces larger avoided crossings than in the case of perpendicular polarization (creating mainly $m=2$), which also makes it harder to create bound states by Landau-Zener crossings.

We are able to extract information on the lifetime involved in the recombination process when we fix the wavelength to a value which excites the rubidium atoms above the saddle point, and scan the delay time of the unipolar pulse with respect to the time of excitation. In Fig. 4 the bound fraction as a function of the time delay for different values of the wavelength of the ionizing laser is depicted. We again can clearly see that the lifetimes of the parallel excited states are substantially shorter than the lifetimes in the perpendicular case [9]. However, even well above the saddle point there are still states which have surprisingly long lifetimes for both parallel and perpendicular cases. This can be understood with the help of semiclassical arguments: in the case of parallel polarization each scatter wave evolves partially toward the saddle point (along the polarization axis, $m=0$), while for perpendicular polarization not much scatter wave travels toward the saddle point (mainly $m=2$).

We have shown that a fast field kick drives a substantial part of the population, initially above the saddle point and free to ionize, back into bound states. Especially in the case of perpendicular polarization, we achieved efficient recombination probabilities. The lifetimes of states in the field-induced continuum that were excited with light polarized perpendicular to the electric field are of the order of tens of nanoseconds, while the times were shorter for parallel polarization, although still in the nanosecond regime.

Let us now reconsider a microwave ionization experiment in a static electric field: if, after a certain set of cycles of a single microwave pulse, a substantial part of the originally bound population is promoted to the field induced continuum, it will still not ionize due to the relatively long lifetimes involved. Therefore, the Stark states still induce Landau-Zener crossings in the microwave field resulting in above-threshold ionization (ATI) [10] behavior. Above-threshold microwave ionization experiments were already

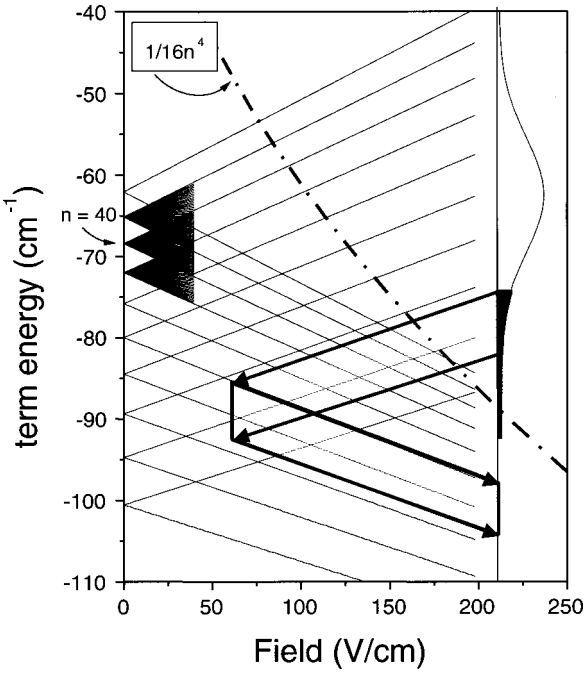

FIG. 5. Schematic representation of the experiment performed in Ref. [6]. A broad-band optical pulse is used to excite states above the saddle point in a static electric field of 212 V/cm. After applying a half-cycle pulse of about 1.5 kV, they were able to observe bound states induced by the THz radiation. In this paper we argue that the inevitable second half-cycle can also play a significant role.

performed by Gallagher and co-workers [11] in the nonperturbative regime, where the intensities from optical ATI experiments were scaled down to the microwave regime. Here we suggest that even much lower intensities of microwave fields in a static electric field already suffice as an alternative way to induce ATI.

Another point worth noting is that we have seen an effect of unipolar pulses as long as 9 ns. General unipolar pulse experiments are performed in the subpicosecond regime, where THz half-cycle pulses with an amplitude of the order of 1 kV are generated by illuminating biased semiconductor wavers with a powerful femtosecond laser pulse [12]. Bearing in mind that this half-cycle pulse propagates through vacuum, it can not be a real half cycle pulse by Maxwell's laws. It should at least have another half-cycle of opposite polarity, so that the pulse has zero impulse. Generally, one supposes that this second unipolar feature has a width at least an order of magnitude larger than the first, and therefore a much lower amplitude, thus contributing to minor effects in the actual experiment. However, we have shown that unipolar pulses with relatively long widths and small amplitudes can effect excited states to a great extent.

Below we argue that such a second unipolar feature is responsible for at least part of the recombination effects observed in the experiments performed in Ref. [6] in the THz regime. In those experiments a broad-band laser pulse ($\Delta E = 10\,\mathrm{cm}^{-1}$) was used to excite states above the saddle point in a static field of about 200 V/cm. They were able to recombine 5% of the generated excited states with the help of a THz 5-kV half-cycle pulse delayed on the order of picoseconds relative to the photoionizing laser pulse. Graphically the excitation scheme is depicted in Fig. 5. If one only takes

into account the reddest 5% of the spectrum of the laser pulse, states very close to the saddle point are excited ($n=35$). In terms of scaled energy these states are in the vicinity of $\epsilon=2E/E_c=-1.68$, which corresponds in our experiment to a term energy of $-32.7\ \mathrm{cm}^{-1}$. We estimate the duration (30 ps) and amplitude (150 V/cm) of the second unipolar feature in the experiments of Ref. [6] by requiring that the total impulse should be zero, and that the duration of the ''slowest'' second unipolar feature is restricted by the plate separation over which the static electric field is applied. The fraction S/S_{crit} (at $n=35$) is now $\frac{5000}{1}\ (\mathrm{V\,cm^{-1}\,ns^{-1}})=5000$, and therefore their second unipolar feature mainly induces diabatic behavior, just as in our experiments. One can scale the duration of the second feature in the pulse of the experiments of Ref. [6] to our n states as follows: $\Delta F/\Delta t$ scales as n^{-10}, while ΔF scales as n^{-4}; therefore, Δt scales as n^6. This means that a pulse of 30-ps duration at $n=35$ would correspond to a pulse of 0.5 ns at $n=55$, which is in fact not far off from our conditions.

In summary, we have observed recombination of Stark atoms by unipolar field pulses. Lifetimes on the order of nanoseconds have been measured, implying that Landau-Zener transitions in the field-induced continuum are possible in a microwave field, giving rise to ATI. A suggestion is made on the contributions of nonunipolar features on half-cycle THz recombination experiments.

We are grateful to T. Hijmans for his comments on this paper. C.W. and L.D.N. were supported by the Stichting Fundamenteel Onderzoek van de Materie (FOM) and the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO). F.R. was supported by the NSF.

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