PHYSICAL REVIEW A VOLUME 50, NUMBER 3 SEPTEMBER 1994

Short-pulse microwave ionization of Na Rydberg atoms

M. Gatzke,[1] B. Broers,[2] L.D. Noordam,[2] R.B. Watkins,[1] and T.F. Gallagher[1]
[1] *Department of Physics, University of Virginia, Charlottesville, Virginia 22901*
[2] *Fundamenteel Onderzoek der Materie, Institute for Atomic and Molecular Physics, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands*

(Received 18 October 1993; revised manuscript received 4 March 1994)

We have measured the microwave ionization threshold electric fields for ns-state Na Rydberg atoms from $n = 24$–33 as a function of the duration of the ionizing microwave pulse. Pulses as short as 2.8 ns were used, containing fewer than 25 cycles of 8.55-GHz radiation. While the fields required to produce the onset of ionization were independent of pulse duration and exhibited the n^{-5} scaling characteristic of ionization by diffusion through higher-energy states, the fields required to produce nearly complete ionization increased dramatically as the pulse was shortened. Additional measurements of the populations of bound states after the shortest microwave pulses showed that up to $\sim 10\%$ of the atoms were excited to states above the initially populated ns state and the adjacent $(n-1)$ manifold of higher angular momentum states, while most were either ionized or remained in these states. From this observation we conclude that the rate-limiting process for ionization is the transition from the initially populated ns state or the adjacent manifold to the next higher manifold, not the diffusion through higher-lying states.

PACS number(s): 32.80.Rm

I. INTRODUCTION

A sequence of experiments has shown that nonhydrogenic atoms can be ionized by low frequency ($\omega \ll 1/n^3$) microwave fields much smaller than the static fields required for ionization. Specifically, the threshold fields required for ionization scale as $E = 1/3n^5$ [1–4], where n is the principal quantum number. This dependence of the threshold field on n is easily understood with the aid of Fig. 1, which shows an energy-level diagram for some of the $m = 0$ states of Na in an electric field. Here m is the azimuthal orbital angular momentum quantum number. As indicated in Fig. 1, $E = 1/3n^5$ is the field at which the extreme n and $n+1$ Stark states with $m = 0$ intersect. These two states are coupled by the Na^+ core and have an avoided crossing, as shown by the inset of Fig. 1.

Microwave ionization of Na can be thought of in terms of multiphoton transitions or in terms of Landau-Zener transitions at avoided crossings of Stark states [3,5]. Since we are particularly interested in short pulses the Landau-Zener description is more convenient. Our present understanding of microwave ionization can be summarized in the following way [1]. Consider a Na atom that is initially prepared in a state of principal quantum number n in zero field and then exposed to a long microwave pulse. As the amplitude of the microwave field rises atoms are quickly spread over all the Stark states of the same n, even in very small microwave fields. These transitions occur because of the core-induced avoided crossings of the Stark states at zero field. If the field amplitude reaches $1/3n^5$, an atom in the highest-energy Stark state of principal quantum number n is brought to the avoided crossing with the lowest-energy $n+1$ Stark state at the peak of the microwave field, and an n to $n+1$ Landau-Zener transition can occur. A field amplitude high enough to drive the n to $n+1$ transition can easily drive the analogous intermanifold transitions between higher n states. Consequently, an atom can make transitions up to a higher-lying state above the classical field-ionization limit, where it ionizes. In long (1 μs) pulses of microwaves a sharp threshold field for microwave ioniza-

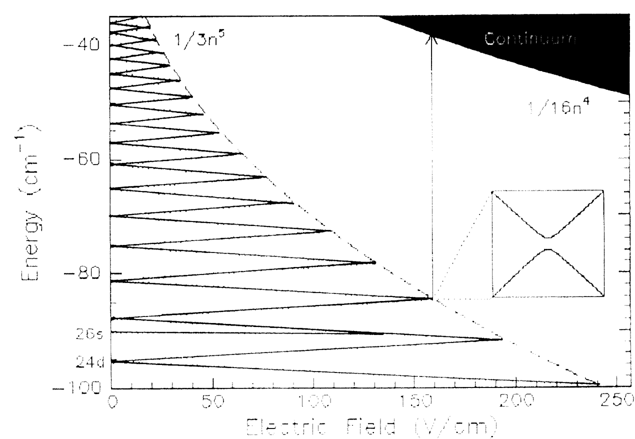

FIG. 1. Energies of the extreme Stark states in Na with $24 \leq n \leq 39$ and $m = 0$ as a function of electric field. In addition, the 26s energy is plotted to illustrate the effect of the nonzero quantum defect in Na. The dashed line indicates the electric field strength where adjacent Stark manifolds start to overlap ($E = 1/3n^5$), which also marks the threshold field for microwave ionization by consecutive Landau-Zener transitions through the avoided crossings (see inset) between levels of different n. The bound-continuum boundary is the classical field-ionization limit ($E = 1/16n^4$). The arrow represents 175 microwave photon absorptions required for the initially bound $n = 25$ state to reach the ionization limit.

tion is observed at $E \approx 1/3n^5$, the field required to drive the n to $n+1$ transition.

The description presented above implicitly assumes that the microwave pulse is long. Specifically, the model assumes that, when the field strength is such that the first n to $n+1$ transition is possible, the microwave pulse is sufficiently long that none of the steps in the ionization process limit the rate. While relatively sharp threshold fields near $E = 1/3n^5$ are observed for long pulses, it is not *a priori* obvious that such will be the case for short pulses. For example, it may be that in very short microwave pulses the sequence of Δn transitions from n to n_c, the principal quantum number corresponding to the classical field-ionization threshold which scales as $1/n^4$ [6,7], cannot be completed for $E \approx 1/3n^5$.

As a step towards understanding the dynamics of microwave ionization of nonhydrogenic atoms, we have studied the ionization of Na Rydberg atoms by 8.55-GHz microwave pulses as a function of pulse duration, including pulses more than two orders of magnitude shorter than those used in the previous long pulse experiments (typically 500 ns). Specifically, we have started with 500-ns pulses containing 4300 field cycles and reduced the pulse width to 2.8 ns, or about 25 field cycles.

There are two specific questions to be answered by this experiment. First, how does the ionization probability as a function of field depend on the pulse duration? Presumably, it should evolve from a sharp threshold at $E = 1/3n^5$ for a long pulse to a broad threshold extending up to fields as high as the classical field-ionization limit for the shortest possible pulses. Second, is the n to $n+1$ transition still the rate-limiting step for short pulses, or are many atoms left in the states between n and n_c? In the following sections we describe the experimental approach, our observations, and their implications regarding microwave ionization.

II. EXPERIMENTAL APPROACH

In previous measurements of microwave ionization of Rydberg states, high Q resonant cavities were used to produce the largest possible electric fields [1–3]. The long filling or damping time of a cavity precludes its use for generating short microwave pulses. For this reason, the measurements described here were made using a terminated section of rectangular waveguide (either WR 90 or WR 137), operated in the TE_{10} mode at 8.55 GHz. A pair of double balanced mixers whose intermediate frequency ports were driven by a Philips PM 5785B pulse generator with a 1-ns rise time served as a pulse modulator for the microwave signal from a HP 8350B sweep oscillator. The two mixers (Watkins Johnson M14A and M12) were used in series to increase the on-off contrast ratio of the pulses to about 50 dB. The microwave pulses, which were as short as 2.8 ns full width at half maximum, were then amplified by a traveling-wave tube amplifier and attenuated by a variable waveguide attenuator before reaching the section of waveguide defining the interaction region.

Since the general experimental approach is described elsewhere in detail [1–3], only a sketch is presented here. As in the previous experiments, Na atoms in an atomic beam entered a section of microwave waveguide from a small hole in one of the side walls, and two laser beams tuned to the $3s$-$3p$ and $3p$-ns transitions entered from an identical hole on the other side. The excited atoms were created approximately 100 ns before the application of a microwave pulse to the waveguide. A copper plate or septum was fitted into the waveguide perpendicular to the microwave electric field. A high voltage pulse applied to the septum after the microwave pulse produced an ionizing electric field which was used to analyze the atoms which survived the microwave ionization process. Depending on the polarity of this field, either Na^+ ions or electrons resulting from either microwave or pulsed field ionization were accelerated out of the waveguide through a hole in the top and then detected using microchannel plates.

As described in Ref. [1], the ion signals produced from microwave ionization and from ionization by the pulsed field arrive at the detector at different times. Using two gated integrators we were able to observe both the ionized atoms and those surviving the microwave pulse in bound states. When the polarity of the field pulse is reversed to detect electrons, only the surviving atom signal can be observed, because electrons from microwave ionization are dispersed long before the arrival of the field pulse. However, since the electron flight time to the detector is short compared to the rise time of the field-ionization pulse, the time-resolved electron signal indicates the field at which the atoms are ionized. This time-resolved electron signal is a good method of determining the final state distribution of the remaining atoms. Due to the long flight times of the Na^+ ions, it is not possible to temporally discriminate between the field-ionization signals from nearby final bound states using Na^+ detection.

The ionization probability was measured as a function of the peak microwave field amplitude during the pulse by recording the Na^+ signals from both microwave and pulsed field ionization, while scanning the attenuation of the microwave pulses. For simplicity, we shall refer to the peak amplitude obtained during the pulse as the microwave field strength.

III. RESULTS AND DISCUSSION

Typical results for a scan of the Na^+ field ionization signal versus microwave field strength are shown in Fig. 2 for ionization from the initially populated $32s$ state by both 500- and 2.8-ns pulses. Recall that the field-ionization signal is the complement of the ionization probability. In Fig. 2 there is a relatively sharp transition from no ionization to almost 100% ionization for the 500-ns pulse, as observed previously [1–3]. For the 2.8-ns pulse, the onset of ionization occurs at about the same field as it does for the 500-ns pulse, but the field strength required for complete ionization is at least a factor of 3 larger. The data shown in Fig. 2 are representative of all our measurements made for initial Na ns states with principal quantum number ranging from 24 to 33. In

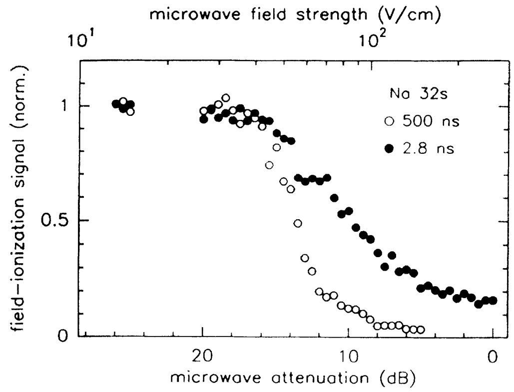

FIG. 2. Field-ionization signal of Na 32s atoms not ionized by the microwave pulse, as a function of the microwave field strength for a 500-ns microwave pulse and a 2.8-ns pulse. The cluster of data points at 25 dB attenuation represents the signal with no microwave pulse.

addition, threshold fields determined by examining the microwave ionization signals were the same as those obtained from the field-ionization signals to within 10%.

For each initial state we examined ionization with microwave pulse durations of 2.8, 10, 20, 50, and 500 ns. In some cases 5-, 100-, and 200-ns pulses were also used. Figure 3 shows a plot of the field strengths at which 10%, 50%, and 80% ionization were observed, as a function of pulse duration τ, for 28s and 31s as the initial state. Since the microwave field strengths attainable in the experiment were sufficient to reach 90% ionization of the 31s state, this threshold is given as well. As can be seen in Fig. 3, the fields required for 10% and 50% ionization exhibit a slowly rising trend with decreasing pulse length, whereas the fields required for 80% and 90% ionization rise much more rapidly. This phenomenon is observed for all initial states, but is more pronounced at high n than at low n, as shown by Fig. 4. In this figure we show the n dependence of the 10%, 50%, and 80% ionization fields for microwave pulse lengths of 2.8 and 500 ns. In addition, the 90% thresholds for the 500-ns pulses are given. For the 2.8-ns pulses, the field strengths required for 90% ionization were generally beyond our maximum. Examination of Fig. 4 reveals that the 10% and 50% ionization fields are close to $E = 1/3n^5$ and nearly independent of pulse length. In contrast, there is a striking dependence of the 80% and 90% ionization fields on pulse length. Furthermore, even for 500-ns pulses these threshold fields rise significantly above the $1/n^5$ dependence at high n. This broadening of the thresholds at high n has been reported previously [3]. In other words, even for long pulses there is no longer a sharp threshold field for ionization. Our findings indicate that 500-ns pulses cannot be considered to be saturating, i.e., lasting long enough to produce complete ionization, for Na ns states with $n > 30$. The most striking point about Fig. 4 is the dramatic increase in the 80% ionization field when the pulse length is shortened to 2.8 ns. As shown, these fields do not exhibit a $1/n^5$ dependence, and they approach the values corresponding to $E = 1/16n^4$ at the highest n studied.

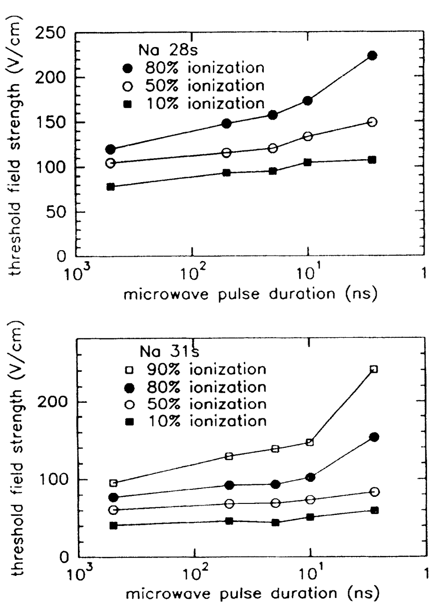

FIG. 3. Microwave field strengths required for 10%, 50%, and 80% ionization as a function of microwave pulse duration for the 28s and 31s states in Na. For the latter state the 90% thresholds are also given.

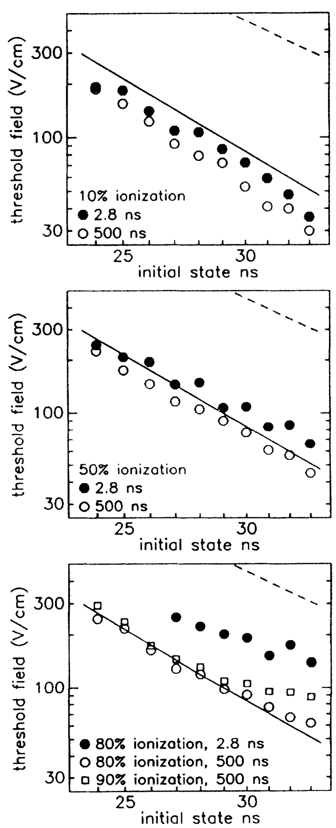

FIG. 4. Threshold microwave field strengths required for 10%, 50%, and 80% ionization by a long (500 ns) and a short (2.8 ns) microwave pulse as a function of the initial ns state. The 90% thresholds for the 500-ns pulses are also given. The thresholds $E = 1/3n^5$ and $E = 1/16n^4$ are indicated by the solid and dashed lines, respectively.

From Fig. 4 it is apparent that for shorter pulses there is a large range of fields beyond $E = 1/3n^5$ for which ionization is not complete. For long pulses, at most field amplitudes, atoms are either ionized or remain in their initial n state. Given the discussion in Sec. I, it seems reasonable to expect that, for short pulses, incomplete ionization may be accompanied by atoms left in other excited states between the initial state and the classical ionization limit. In order to determine the population fractions in other excited states, we examined the field-ionization signals from atoms which survived the ionization process.

As mentioned earlier, the Na^+ field-ionization signal is not well suited for a careful analysis of the final state distribution. For this purpose the time-resolved electron signals are superior. In Fig. 5 we show the electron field-ionization signal of the Na 27s state for several amplitudes of microwave field using a 2.8-ns microwave pulse. The slew rate of the field-ionization pulse in this case was about 0.8 V/cm ns^{-1}. The time of flight of the electron to the detector was about 10 ns. With no microwave pulse, a single peak due to adiabatic ionization of the 27s state is observed [6]. When a microwave field of about 70 V/cm is applied, the initial adiabatic 27s signal is diminished, and the signal peak broadens toward earlier times. The signal which precedes the original 27s peak arises from adiabatic ionization of the $n = 26$, $m = 0$ Stark manifold states. As described in Ref. [1], microwave ionization of $(n + 1)s$ states in Na requires first a transition to the adjacent n Stark manifold and then Landau-Zener (or multiphoton) transitions to states of higher n. Since the s-state quantum defect in Na is 1.35, the $(n + 1)s$ states intersect the n Stark manifolds at $E = 0.7/3n^5$, as shown in Fig. 1 for the 26s state. We observed this mixing of $(n + 1)s$ and the n manifold Stark states to occur for fields exceeding 0.7 times the 50% ionization threshold field, independent of pulse duration.

The combined ionization signal from the 27s state and the $n = 26$ Stark manifold states, shown in Fig. 5 for a microwave field of 73 V/cm, has the same time-integrated strength as the signal from the 27s state with no microwaves, indicating no microwave ionization or excitation to higher-lying states. When the field amplitude is increased beyond the microwave ionization threshold (about 117 V/cm for $n = 26$), both signals begin to disappear together. When the gated integrator is set to capture field-ionization signals from both the initial $(n+1)s$ state and the n manifold states (but not $n + 1$ manifold states), a single threshold similar to those shown in Fig. 2 is obtained. For simplicity, we shall refer to both the $(n + 1)s$ state and the adjacent manifold of higher angular momentum states of principal quantum number n as the initial n state. Note that the electron field-ionization signals obtained in this way are produced exclusively by atoms left in the initial n state after the microwave pulse. Given that the results for threshold fields obtained from these electron signals were the same as the thresholds obtained from ion signals to within 10%, we conclude that a large majority of atoms is either ionized by the microwave pulse or remains in the initial n state.

Although we do not see clear evidence for atoms being left in higher-lying states in time-resolved signals such as Fig. 5, if we integrate the signal over times before the arrival of the adiabatic ionization signals of the original $(n + 1)s$ state and the n Stark manifold as a function of microwave field amplitude, we do observe a small signal at microwave fields above the 50% ionization thresholds. In Fig. 6(a) we show temporally integrated signals due to the adiabatic ionization of the initial 30s state and the adjacent $n = 29$ manifold as a function of microwave field strength for both 100-ns and 5-ns pulses. In Fig. 6(b) we show analogous scans obtained with the integrator set earlier to capture signal from adiabatic ionization of the $n = 31$ manifold. Clearly there is an increase in this earlier signal above the background at fields higher than the 50% ionization field for the 5-ns pulse. This is

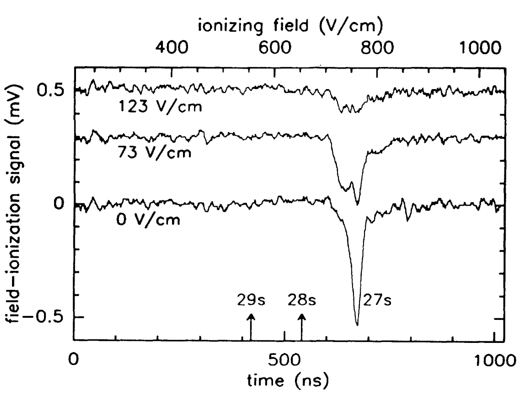

FIG. 5. Time-resolved electron field-ionization signals for an initially populated 27s state after exposure to a 2.8-ns microwave pulse, for three different microwave field amplitudes (0, 73, and 123 V/cm). The signals have been offset for clarity. The arrows indicate the locations of the field-ionization signals from the 28s and 29s states.

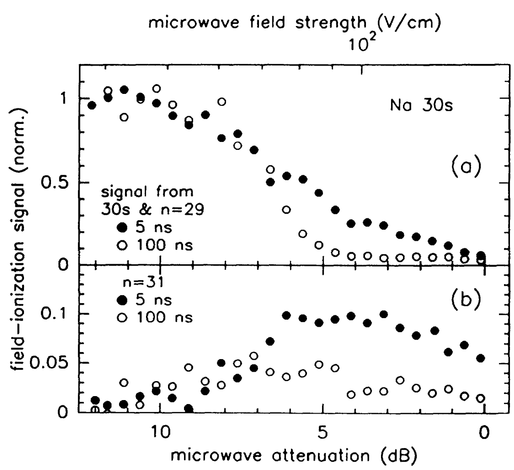

FIG. 6. Electron field-ionization signals from atoms not ionized by the microwave field. The signals are given as a function of microwave field strength for a long (100 ns) and a short (5 ns) microwave pulse. The initial state was 30s. Different gates were set over the time-resolved field-ionization trace to see in which states the atoms are left after the microwave pulse. (a) Signal from the initial 30s state and the $n = 29$ manifold. (b) Signal from the $n = 31$ manifold.

presumably the field-ionization signal from a small fractional population in the higher-lying $n = 31$ manifold states, which ionize in lower fields. Negligible signal is observed from still higher levels. In sum, the 5-ns microwave pulse with a peak field of 100 V/cm ionized about 65% of the atoms, while leaving approximately 25% in the initial $n = 29$ state and exciting approximately 10% to bound states of the $n = 31$ manifold, which were detected by pulsed ionization in fields ranging from 328 to 414 V/cm. The adiabatic ionization field of the $30s$ state is 487 V/cm. The fraction of signal left in these excited states decreased with increasing pulse duration. For pulses lasting 100 ns or more, the fraction remaining in these same excited states was very small, less than a few percent. Similar results were seen for the initial states $24s$, $27s$, and $33s$.

Apart from our measurements, two other experiments have been done in which the final states were analyzed after exposure to short microwave pulses, and it is interesting to compare the results of these measurements to ours. In both cases the frequencies were approximately equal to the Δn spacing of the initial state, $\omega \approx 1/n^3$, in contrast to our experiments in which $\omega < 1/20n^3$. Blümel *et al.* [8] examined the effect of 12-GHz pulses on the Rb $84p$ state. With 1.37 V/cm pulses of 20–1000 ns duration their observations were consistent with spreading of the population over the adjacent $n = 81$ manifold of states and $\approx 10\%$ population in the neighboring manifolds, while much less population was left in states removed by more than $\Delta n = 2$. For reference, at $n = 81$ the fields $1/3n^5$ and $1/16n^4$ are 0.49 and 7.5 V/cm, respectively. Bayfield and Sokol [9] exposed H atoms in the extreme $n = 72$ Stark manifold state to 13–18-GHz microwave fields for 7.5 ns. The fields had amplitudes from $0.018/n^4$ to $0.042/n^4$ and were strong enough to produce 5–70 % ionization. In contrast to the Rb results and our own, they observed a spreading of the population over several states. The differences observed in H may be due to the fact that the microwave field was stronger or the fact that H is fundamentally different, having no avoided level crossings between the Stark states. Probably both of these effects play a role.

It is clear from our measurements that more than 90% of the atoms are either ionized by the pulse or remain in the original n manifold, even for the shortest pulses we used. This observation implies that transitions between higher-lying states are not rate limiting for the ionization process, at least not for states in the range $24 \leq n \leq 33$ for 8.55-GHz pulses as short as 2.8 ns, i.e., for pulses as short as 25 cycles. For $n = 30$ atoms the minimum number of field cycles required to make the sequence of Δn transitions from n to n_c is 5. A path requiring the minimum number of cycles tends to be through the extreme Stark states, for then the energy gain per cycle is maximized. This process is shown for $n = 20$ in Fig. 1 of Ref. [1]. For ionization to occur with a five-cycle pulse, a 100% transition probability for the Δn transitions in each field half cycle is needed, an unlikely event. With this thought in mind it is remarkable that a 25-cycle pulse leaves so few atoms between n and n_c. Furthermore, it suggests that the sequence of Δn transitions is probably along the most efficient route, via the extreme Stark states. If transitions between extreme Stark states are relatively rapid, the rate-limiting step in this process is apparently the transition from the initially populated $(n+1)s$ state to the extreme n Stark states. This conclusion is consistent with the measurements of ionization rates by Hettema *et al.* [10] and multilevel Landau-Zener calculations by Harmin [11].

IV. CONCLUSIONS

We have carried out measurements of the microwave ionization of Na Rydberg atoms as a function of the length of 8.55-GHz pulses from 2.8 to 500 ns. While the onset of ionization (10% ionization) occurs at nearly the same field, $1/3n^5$, irrespective of the pulse length, the field required to ionize nearly all the atoms increases dramatically as the pulse length is shortened from 500 ns to 2.8 ns. For long pulses the atoms were either left in the initially populated ns state or the adjacent n manifold, or were ionized. For the shortest pulses we used, we found that as much as 10% of the initial population could be found in the next higher manifold. In no case did we observe significant excitation to states between the initial manifold and the classical ionization limit.

These experiments suggest several interesting avenues to explore. On the experimental front, we suspect that for pulses shorter than 25 cycles a significant number of atoms will be stranded between the initially populated state and the ionization threshold. In the limit of short pulses, the original two-level model proposed to explain microwave ionization is not likely to be at all useful. With pulses of less than 25 field cycles, numerical calculations of the ionization probability become attractive. Theoretically, it is not clear whether quantum or classical methods will prove to be more useful [11–13], but it does seem clear that it should be possible to go beyond the present model.

ACKNOWLEDGMENTS

This research was conducted at the University of Virginia with the support of the Air Force Office of Scientific Research. Two of us (B.B. and L.D.N.) are supported by the Foundation for Fundamental Research on Matter, financially supported by the Netherlands Organization for Advancement of Research. This collaboration is financially supported by NATO (collaboration research Grant No. CRG 920503).

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