Metastable states in microwave ionization

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The 38-GHz microwave field required to effect 50% ionization of Li atoms decreases from 60 V/cm at $n = 37$, where the microwave frequency is one-third the Kepler frequency, to 45 V/cm at $n = 56$, where the microwave frequency is equal to the Kepler frequency. For $n > 56$ the 50% ionization field is constant at $E \approx 2.4\omega^{5/3}$ until ~80 GHz below the ionization limit, where it begins to decline. In spite of the fact that the pulse is 8000 cycles long, we detect approximately 5% of the initial population in very-high-lying ($n > 215$) states subsequent to the microwave pulse. We suggest that these atoms are trapped in metastable atom-field states during the microwave pulse and relax to the high-lying states when the field is turned off. The existence of such states is predicted by the simple man's model of above-threshold ionization.

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I. INTRODUCTION

In photoionization there is a frequency requirement for the onset of ionization, in field ionization there is a field requirement, and midway between these two processes lies multiphoton ionization, which has both requirements. Microwave ionization appears to be a way to systematically connect these apparently different phenomena. Specifically, it is possible to create high-lying Rydberg states, which may be ionized by the absorption of a single microwave photon, as well as low-lying states in which the variation in the microwave field is slow enough compared to the motion of the Rydberg electron to be quasistatic. Thus, by fixing the microwave frequency and changing the principal quantum number n of the atom it is possible to pass from photoionization to field ionization. The first effort in this direction was the work of Bayfield and Koch [1], who studied the ionization of H atoms of $n \sim 65$ by fields of frequencies 9.9 GHz, 1.5 GHz, and 30 MHz. Although presented in different terms, their results show that at the lower two frequencies 50% ionization occurs at the field $E = 1/9n^4$, the same value as the static field at which the extreme red Stark state ionizes. We use atomic units unless specified otherwise. A somewhat lower field is required for ionization by a 9.9-GHz field, a frequency approaching the Kepler or $\Delta n = 1$ frequency of 24 GHz. Later work confirmed that if the microwave frequency ω is well below the Kepler frequency $\omega_K = 1/n^3$, ionization occurs from the initially populated n state, at the field required for field ionization [2]. However, as the ratio of the microwave frequency ω to ω_K approaches one, multiphoton ionization, by transitions through either real or virtual higher-lying levels, becomes possible, lowering the required field for ionization. While quantum calculations of the ionization fields in this regime have only recently become possible [3,4], some time ago classical calculations, based on the onset of chaotic motion, provided an accurate description of ionization in the regime $\omega \cong \omega_K$ [5]. In classical calculations the scaled frequency $\Omega = \omega/\omega_K$ is often used, and it is convenient to use it here. For a fixed microwave frequency, at low n, $\Omega \ll 1$, and at high n, $\Omega \to \infty$. Although one might expect the classical model to work better for higher-n states, where $\Omega > 1$, it predicts that at a fixed frequency the microwave field required for ionization decreases with n [6]. However, experiments have shown that for $\Omega > 1$ the ionization field only depends on the microwave frequency until the initial binding energy is within a few microwave photons of the ionization limit [7,8]. When the initial Rydberg state is within one or two microwave photons of the ionization limit, we would expect perturbation theory to give an accurate prediction of the ionization rate. However, in the experimentally accessible regime the ionization rates are at least five times lower than expected [8], although perturbation theory is undoubtedly applicable in an inaccessibly low microwave power regime.

Here we report the investigation of microwave ionization of Li by 38.3-GHz microwave fields, from the photoionization limit $n > 420$, where $\omega > 1/2n^2$ and $\Omega > 210$, to $n = 37$, where $\omega \simeq 0.32\omega_K$ and $\Omega = 0.32$. We find a smooth variation in the fields at which 10% and 50% ionization occur, in agreement with previous experiments, in which pieces of the above range were explored. More surprising, we find that for all initial states a substantial fraction, ~5%, of the atoms is found in high-lying Rydberg states subsequent to a 200-ns-long microwave pulse. Atoms have been detected in high-lying states subsequent to exposure to short microwave pulses containing only a few field cycles [9] and trains of up to 40 unipolar pulses with a bias field [10]. However, they have only been observed with long pulses when starting from very-high-lying states [8]. They have not been observed in microwave ionization of low-lying states. In the sections that follow we describe our experimental approach, present our observations, and discuss their implications.

II. EXPERIMENTAL APPROACH

This section begins with a general description of our experimental technique followed by a more detailed explanation of the experimental apparatus. A thermal beam of Li atoms in its ground state passes through the antinode at the center of a 38.3-GHz Fabry-Pérot cavity where the atoms are excited to a Rydberg state by three 20-ns dye laser pulses via $2s \to 2p \to 3s \to np$ transitions. The excited atoms are then exposed to a microwave field pulse, as shown in the timing diagram in Fig. 1. The microwave pulse starts about 50 ns after the laser excitation and lasts for 200 ns. The rise and fall times of the pulse are approximately 20 ns each. After the end of the microwave pulse, typically 300 ns later, atoms are field

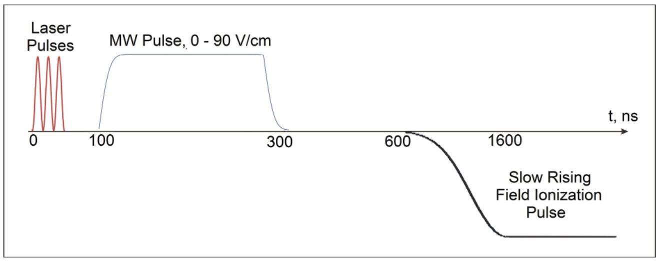

FIG. 1. (Color online) Experimental timing diagram for the investigation of the microwave ionization of Li by a 38.3-GHz microwave (MW) field.

ionized by a slowly rising field pulse. The electrons ejected by the ionization field are detected by a dual microchannel plate (MCP) detector. The time-resolved MCP signal tells us how many atoms were not ionized by the microwave pulse as well as their final-state distribution. The MCP signal is captured by a gated integrator or oscilloscope and recorded for later analysis.

The microwave cavity consists of two brass mirrors 40.5 mm in diameter with 75.9-mm radii of curvature. The on-axis spacing between mirrors is 44.5 mm. The cavity is operated on the TEM_{012} mode at a frequency of 38.34 GHz with $Q = 5100$. A Hewlett Packard (HP) 83550B sweep oscillator with a 83550A rf plug-in is modulated to a pulsed mode by a 8112 HP pulse generator. Its output, frequency doubled by a Phase One SP40 frequency doubler, is sent through a 0- to 50-db variable HP R382A attenuator into the vacuum chamber using a WR-28 waveguide and into the cavity through an iris in one of the cavity mirrors. The microwave system generates a 38.34-GHz pulse with a 0- to 90-V/cm amplitude and a variable width. The microwave field amplitude is calibrated using the method described by Cheng *et al.* [11] and we are able to determine the amplitude of the pulse with an uncertainty of 15%.

The microwave cavity is surrounded by four aluminum plates, which, along with the cavity mirrors, can be grounded or separately biased to null the stray field in the interaction region [8]. The minimum stray field we can obtain is 8 mV/cm and additional bias fields can be added if desired. A negative voltage pulse with a 1-μs rise time is applied to the bottom plate 300 ns after the end of the microwave pulse, providing the field ionization pulse (FIP). This time delay provides enough time for the electrons produced by photoionization or microwave ionization to leave the interaction region before the FIP starts. Thus only atoms that are left in bound states after the microwave pulse are detected. To record the photoabsorption spectrum, the delay between the excitation time and the FIP is decreased to 50 ns to collect all of the electrons. The electrons resulting from field or photoionization are pushed by the FIP towards the MCP through a 1-cm-diam hole in a plate above the interaction region. The MCP signal is then amplified in a SRS SR445A amplifier and is either captured in a SRS SR250 gated integrator and boxcar averager or recorded as an oscilloscope trace. We can choose when to put the integration gate of the boxcar, recording either the signal from the states with selected binding energies or all bound states of $n > 37$. When the boxcar is used the signal going to the boxcar is averaged over 300 samples and sent to the computer for recording. Since high-lying states are ionized earlier in the rising FIP, the time-resolved oscilloscope traces can be used to infer the final-state distribution of bound states surviving the microwave pulse. To show the final-state distributions vs laser tuning or microwave field amplitude we map each trace to a grayscale to produce an image representing the distribution of bound states subsequent to the microwave field.

The apparatus is triggered at a 500-Hz repetition rate by a frequency-doubled Nd:YLF laser, which is used to pump the dye lasers. The 527-nm pulse of the pump laser is sliced in time by Pockels cells to form two 20-ns pulses. The first 20-ns pulse pumps the $2s$-$2p$ and $2p$-$3s$ Littman-Metcalf [12] dye lasers, while the second one pumps the last $3s$-np transition double-grating Littman laser [13]. The output of that laser is taken from the wedge mirror, used in the cavity, in order to decrease the amount of amplified spontaneous emission in the beam. The linewidth of the laser is about 8 GHz and its frequency can be tuned to populate any Rydberg state of $n > 35$. A relative frequency measurement of this laser is obtained with the help of a 52.42-GHz free spectral range étalon and an optogalvanic signal from the 16 274.0212-cm^{-1} $2p^5(2p_{3/2})3s$-$2p^5(2p_{3/2})2p$ Ne line provides an absolute calibration. The three laser beams are sent to the vacuum chamber and focused to less than 1-mm-diam spots where they cross each other and the atomic beam. The laser field and microwave field are polarized vertically throughout the experiment.

III. OBSERVATIONS

The initial objective of this work was to study the ionization thresholds of the Li Rydberg states in a microwave field of 38 GHz not only near the ionization limit ($\Omega \rightarrow \infty$) [8], but also in the region where the classical orbital Kepler frequency of the Rydberg states is larger than the microwave frequency of 38.3 GHz ($\Omega < 1$). In order to observe the ionization behavior, we continuously change the binding energy of Rydberg states by scanning the frequency of the laser driving the $3s \rightarrow np$ transition while recording the field ionization signal from the MCP. We repeat such scans in zero field, when the microwaves are off, as well as in the presence of microwave fields of different amplitudes. Obtaining these spectra for many microwave fields allows us to extract the threshold fields required to ionize a given fraction of the initial population. Unless stated otherwise, data are taken for 200-ns long, 38.34-GHz microwave pulses in such a way that the signal from all bound states ionized by the FIP is recorded by the gated integrator. Continuously changing the initial binding energy of the excited atoms by scanning the laser frequency, we obtain spectra showing the population found in bound states after the microwave pulse. Figure 2 shows spectra recorded with the microwave field off [Fig. 2(a)] as well as spectra obtained in the presence of fields of 36 V/cm [Fig. 2(b)] and 90 V/cm [Fig. 2(c)]. The zero of energy of the horizontal scale is the ionization limit. In zero field we observe resolve bound Rydberg states at energies lower than −300 GHz and a flat signal at higher energies, where the spacing between adjacent levels is smaller than the linewidth of our laser, which occurs at $n \approx 100$. At an energy of

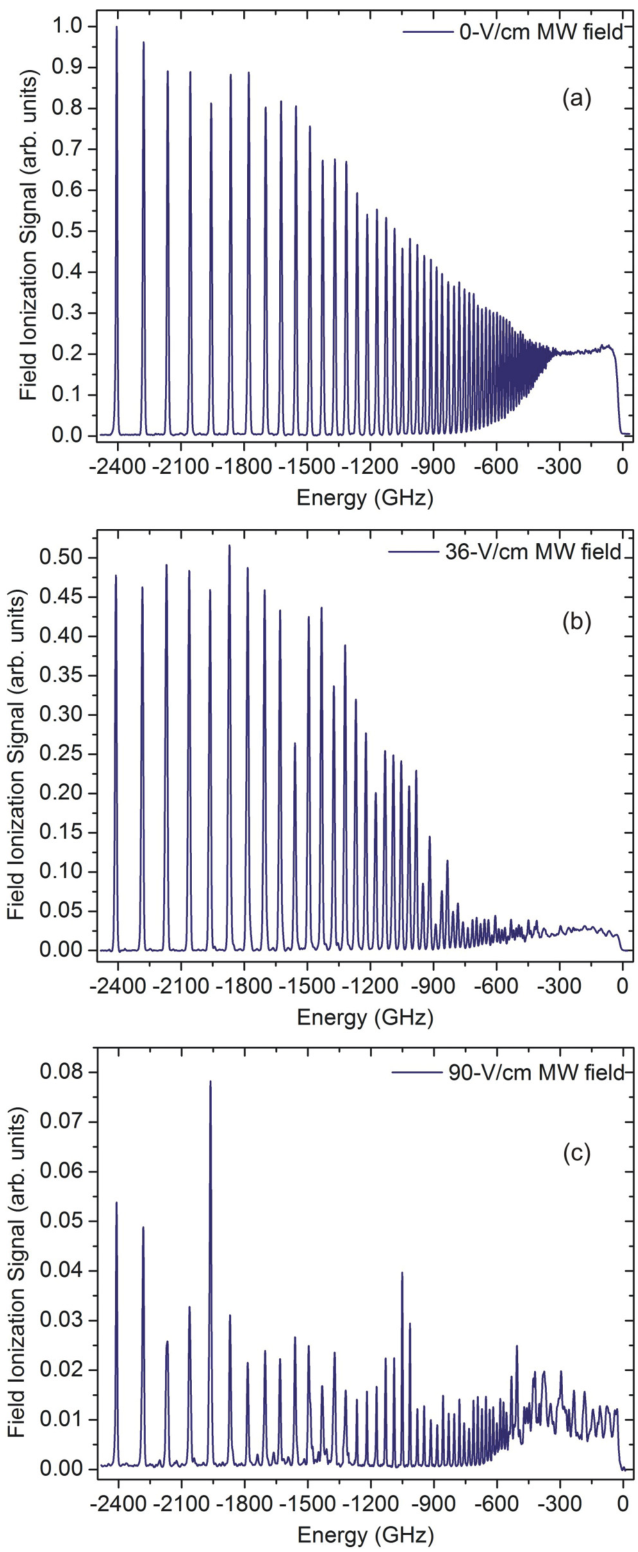

FIG. 2. (Color online) Field ionization signal as a function of the laser frequency tuning, in terms of energy relative to the ionization limit, after exposure to a microwave pulse of amplitude (a) 0 V/cm, (b) 36 V/cm, and (c) 90 V/cm. The horizontal axis is calibrated by the étalon and optogalvanic signals. The first peak on the graphs corresponds to $n = 37$ at -2410 GHz.

-45 GHz, the signal starts falling to zero because photoelectrons are not detected when the laser is tuned over the ionization limit. Energies are measured relative to the zero-field limit. Due to the finite linewidth of the laser, the signal drops to zero with a finite slope and we denote the middle of that slope as the depressed ionization limit. The limit is depressed due to a nonzero static field in the interaction region. We estimate that stray field using $\Delta W = 2\sqrt{E}$. Typically, the data are taken when the depressed ionization limit is at -18 GHz, which corresponds to a stray field of 8 mV/cm. In the 36-V/cm scan of Fig. 2 we see a clear drop in the survival probability at an energy of -900 GHz ($n = 60$), several states above the point where the classical orbital frequency of the atom most closely matches the microwave frequency, i.e., where $\Omega = 1$. The match occurs at $n = 56$, which has an energy of -1050 GHz. The 90-V/cm scan reveals several notable features. First, there is a state at -1960 GHz ($n = 41$) that is much more resistive to ionization than the adjacent states. Second, states at energies near -1050 GHz, where the microwave frequency matches the Kepler frequency, are remarkably stable. Third, a large number of atoms are left in bound states after a 200-ns, 90-V/cm pulse. For comparison, we note that a 90-V/cm static field would ionize states down to an energy of -1300 GHz. Finally, there is very noticeable modulation at the microwave frequency just below the limit, which agrees with previous observations [8]. This modulation is most apparent in Fig. 2(c).

Figure 2 shows only typical spectra obtained for several microwave fields. From the set of all such spectra we can extract the threshold fields for ionization. Taking spectra in the microwave field and normalizing them by the zero-microwave-field signal at the same laser frequency, we are able to determine the fraction of atoms surviving vs microwave field amplitude for each Rydberg state. Here we present such data for the range $37 < n < \infty$, which encompasses $1/3 \leqslant \Omega < \infty$. In Fig. 3(a) we show the microwave fields required to ionize 10% and 50% of the initial population vs binding energy.

In Fig. 3(a) we show the calculated ionization fields. At low n, where $1/3 < \Omega < 1$, we show the 10% ionization fields calculated by Krug and Buchleitner [3]. When $\Omega < 1$ the finite size of the Li^+ core leads to ionization fields far lower than those observed in H, but for $\Omega > 1$, the presence of a finite-sized core is inconsequential and the calculated ionization fields for Li and H are the same. For example, scaling the experimental hydrogenic 36-GHz ionization fields to 38 GHz at $n = 37$ yields an ionization field of 125 V/cm [14], five times higher than the Li 10% ionization field. In contrast, the scaled H, $n = 56$ field is 19 V/cm, in good agreement with the Li 10% ionization field. The intermediate regime $1/3 < \Omega < 1$, where the effect of the core is changing, poses the greatest theoretical challenge and for this reason the agreement between the calculation of Krug and Buchleitner and our observations is most impressive.

To a first approximation, we observe an n-independent ionization field for $\Omega > 1$ for both the 10% and 50% ionization fields. Jensen *et al.* [15] proposed that in the $\Omega > 1$ regime ionization occurs by a sequence of transitions through excited states. The requirement is that the dipole coupling due to the microwave field equals the average detuning from resonance of the transitions of the sequence. Since both the coupling and the detuning scale as $1/n^3$, this condition is met for all n

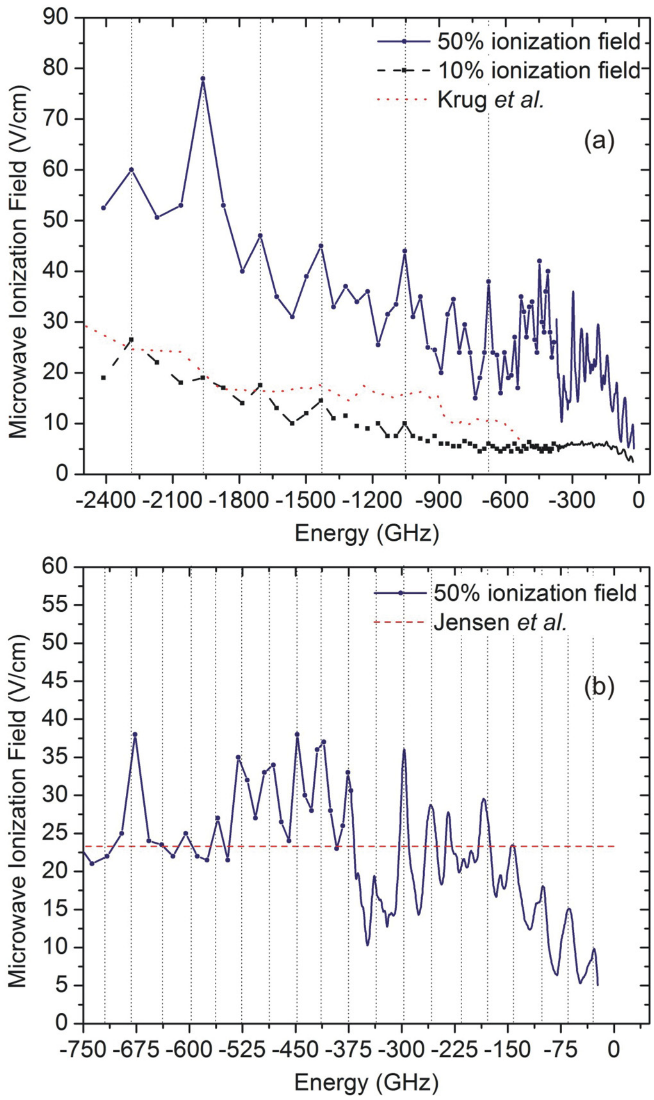

FIG. 3. (Color online) (a) Microwave fields as a function of atomic energy for 50% and 10% ionization. Vertical dotted lines on the graphs correspond to the energies of Rydberg states for which the classical Kepler frequency equals 3ω, 2.5ω, 2ω, 1.5ω, ω, and 0.5ω, respectively, where $\omega/2\pi = 38.34$ GHz is the MW frequency. Our observations for the 10% field are in good agreement with theoretical calculations made by Krug and Buchleitner [3] for the ionization of Li by a 36-GHz MW field (red dotted line) for $1/3 < \Omega < 1$. We are unaware of a calculation for 10% ionization fields at 38.3 GHz in the region from -500 GHz to the IL. (b) Expanded view of the 50% ionization fields in the $\Omega > 1$ regime. Our result is in excellent agreement with a prediction of Jensen *et al.* [15] (red dashed line) that the average ionization field does not depend on n. That prediction, however, is not intended to reproduce the experimental structure that we observe. As shown by the vertical dotted lines spaced by $\omega/2\pi$, the structure in the ionization fields is at the microwave frequency.

when

$$E = 2.4\omega^{5/3}. \quad (1)$$

This prediction, shown by the red dashed line in Fig. 3(b), is in excellent agreement with the 50% ionization fields from $\Omega = 1$ to within four microwave photons of the ionization limit. A feature that we do not yet understand is the peak at $n = 41$ in the 50% ionization field.

Figure 4 shows the final-state distributions of atoms that survive the microwave fields vs laser tuning. The oscilloscope trace taken for each laser frequency is mapped to a grayscale with black corresponding to large signal amplitude and white to no signal. The vertical scales of Fig. 4 are given both as time and the field of the FIP. Figures 4(a) and 4(b) show the final-state distributions obtained with 36- and 90-V/cm microwave fields. The adiabatic and diabatic ionization fields for the initially excited states are shown by the dashed and dotted lines, respectively. An unexpected discovery is evidence of the extremely-high-lying states that are produced by the 38-GHz microwave field for all initial states. The signal from these states is seen at ionizing fields between 0 and 2 V/cm (times between 10 and 50 ns) in Fig. 4. Both the 36- and 90-V/cm scans in Fig. 4 show the presence of extremely-high-lying states for initial energies down to -2400 GHz for 36-V/cm field and -3300 GHz for the 90-V/cm field. Especially surprising is the 90-V/cm scan, as we see that almost all other population is ionized, but weakly bound states are still present. Other notable features in Fig. 4 are the following. First, we see in the 36-V/cm traces that atoms from the states with initial energies below -900 GHz can be found over a broad range of final energy. Second, final states for the laser tuning in a range where $\Omega < 1$ have their central maximum at the same time, corresponding to states adjacent to $n = 56$ (-1050 GHz) where $\Omega = 1$. In other words, atoms initially in lower-lying states are apparently trapped in the states near $\Omega = 1$. Very little population is found between pulsed ionization fields of 3 and 40 V/cm. However, for $\Omega \geqslant 1$, the pattern changes. Starting from an initial energy of -950 GHz, just two states above where $\Omega = 1$, most atoms, if not ionized, stay in the initial state and several adjacent ones or are found near the limit. In this regime the total bound population in that region drops abruptly, which was also seen in Fig. 2(b). Finally, we note that $n = 36$ atoms, which have the anomalously high ionization field in Fig. 3(a), are found near their initial energy with microwave fields of both 36 and 90 V/cm.

The final-state distributions shown in Fig. 4 imply that bound atoms detected subsequent to the microwave pulse, as shown in Fig. 2, are largely in highly excited states. This suggestion is confirmed in the following way. Simultaneously with the oscilloscope traces of Fig. 4, the field ionization signal was recorded using a 50-ns gate, positioned to detect only the extremely-high-lying states, those within 75 GHz of the depressed ionization limit of -25 GHz. Figure 5 shows the corresponding spectra. First, where the initial states cannot be resolved by the laser there is obvious structure at the microwave frequency, as shown in Fig. 2(c). It is particularly apparent in Fig. 5(b), in which peaks in the number of surviving atoms are observed at integral multiples of the microwave frequency below the depressed ionization limit. Second, we see a peak in the population in the extremely-high-lying states when the laser is tuned near $\Omega = 1$. Figure 5 suggests that this peak is probably due to the fact that atoms are retained longer in the initial states for this laser tuning. Third, the population transferred to high-lying states from the $n = 41$ state at -1960 GHz is much lower that from the adjacent

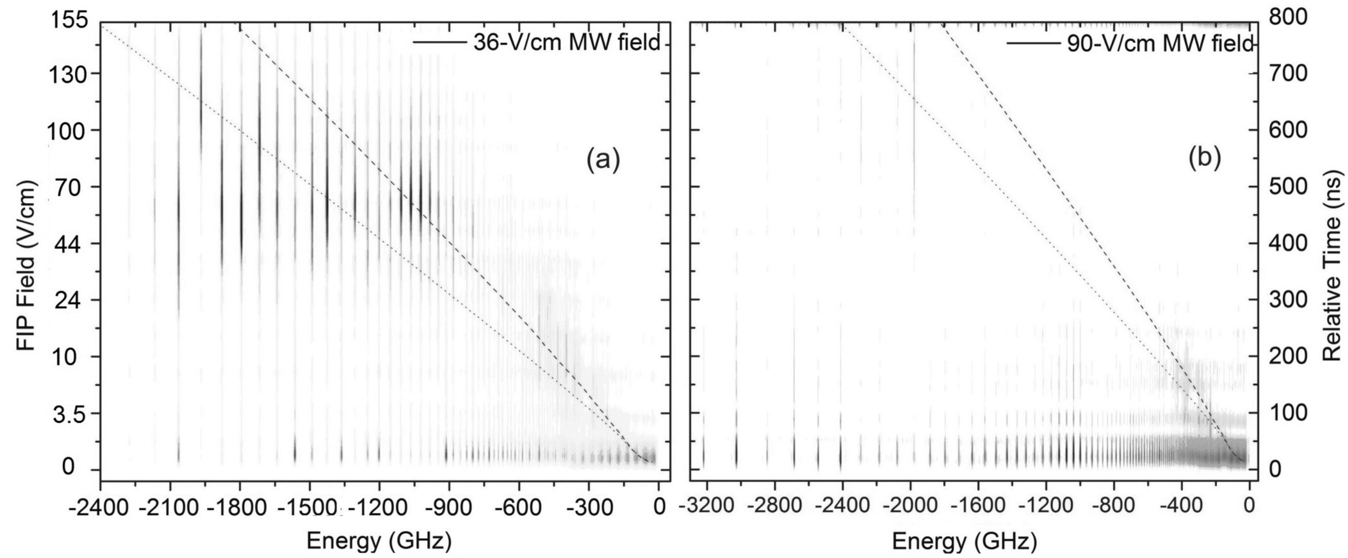

FIG. 4. Final-state distributions as a function of energy for (a) 36-V/cm and (b) 90-V/cm MW fields. The distributions shown are grayscale representations of oscilloscope traces of time-resolved field ionization signals. Black is a large signal and white is no singal. For convenience the vertical scale is shown both as time and the field of the FIP. The dashed lines in (a) and (b) show the diabatic ionization field ($1/9n^4$), while the dotted lines correspond to the adiabatic ionization field ($1/16n^4$). Both the 36- and 90-V/cm plots show the presence of the extremely-high-lying states that are ionized within the first 50 ns of the FIP. The redistribution to states other than the initial or extremely high lying occurs at both higher and lower binding energies, but in no case do we observe a continuous distribution from the initial state to the IL.

states, making this state stand apart from its neighbors. This particular state is also more resistant to ionization, as can be clearly seen in Fig. 3(a). In this case the population remains in low-lying states.

Figures 2(c) and 5(c) point to the surprising fact that all the atoms are not ionized by strong microwave fields. Instead, they are found in highly excited states. This point is made explicitly by measuring the fraction of atoms surviving the microwave pulse as a function of microwave field amplitude for several laser frequencies. Figure 6 shows the total survival probability when the integration gate is wide, allowing signals from all bound states to be recorded. Almost all the population loss, due to ionization, occurs at low microwave fields. For fields above 50 V/cm there is minimal population loss with increasing field. The surviving bound population is found entirely in the highly excited states, as shown by Fig. 7, which shows the result of the same measurement taken with the narrow 50-ns gate, to collect only the signal due to the extremely-high-lying states. For initial energies lower than −700 GHz population begins to be observed in the high-lying states when 50% of the atoms are ionized. For states initially bound by less than 700 GHz, population is found in high-lying states at microwave fields less than the 50% ionization field. Finally, the population in these states decays very slowly as the microwave pulse length is increased; we measured the lifetime of the trapped population in the field to be about 50 ns in a 90-V/cm field and 120 ns

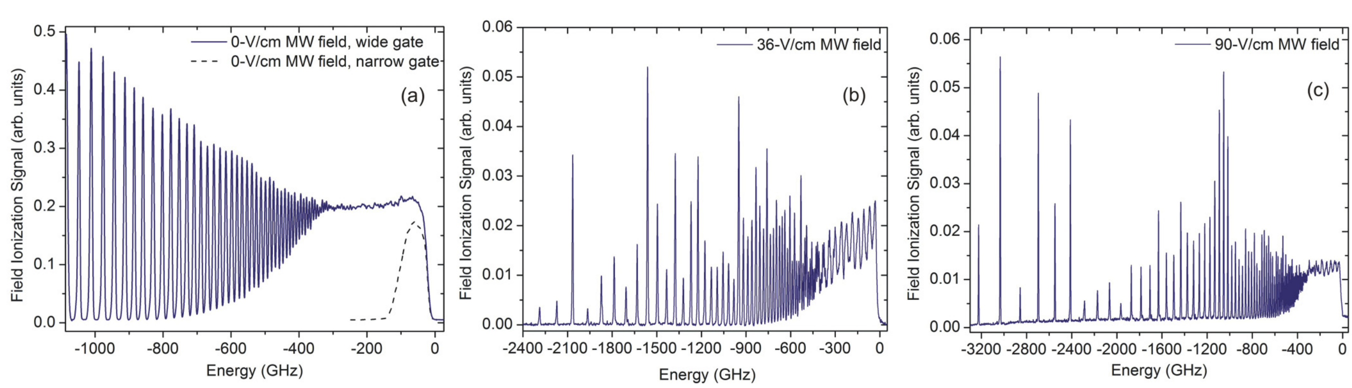

FIG. 5. (Color online) Field ionization signal as a function of energy for three microwave field amplitudes: (a) 0 V/cm, in which case two different gates were used, a wide gate to accept the signal from all Rydberg states of $n \geqslant 37$ (solid line) and a 50-ns gate to detect only atoms within 75 GHz of the IL (dashed line); (b) 36 V/cm, a spectrum recorded with the 50-ns gate; and (c) 90 V/cm, a spectrum recorded with the 50-ns gate. In (b) and (c) we see a nonzero signal within the narrow gate when the laser is tuned to the Rydberg states from which no signal is detected with the same gate when the MW field is off. Not only is the expected microwave structure present near the IL, but also a large fraction of population is observed due to a transfer from the bound Rydberg states to the extremely-high-lying states, even when the laser is tuned to a state with n as low as 30.

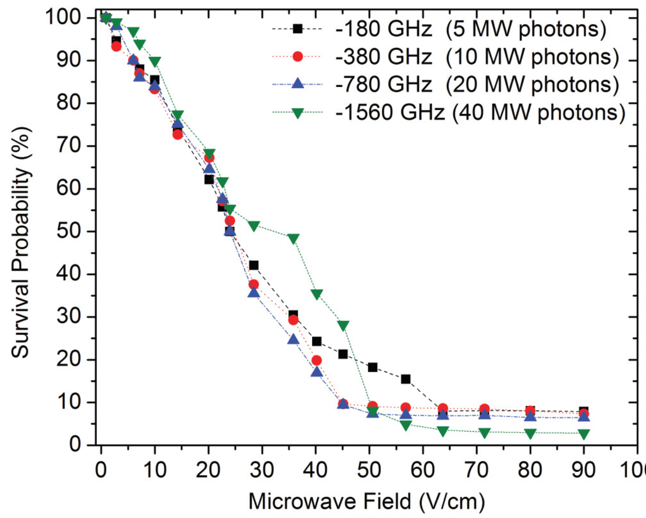

FIG. 6. (Color online) Fraction of atoms surviving the MW field as a function of its strength when the laser frequency is tuned to four different Rydberg states. All bound states are detected (wide gate). For each of these binding energies the 50% ionization field is almost the same, as predicted by Eq. (1). For all of the states minimal further ionization is produced by raising the microwave field above 60 V/cm.

in a 45-V/cm field when the atoms are initially bound by ten microwave photons.

To understand more clearly the ionization, redistribution, and trapping of the initial population by the microwaves, we have measured final-state distributions as a function of the microwave field amplitude. Again we recorded the oscilloscope traces as the microwave field was varied with the laser frequency fixed at energies of −192, −386, −770, and −1530 GHz. These traces were mapped to a grayscale as seen in Fig. 8. With an initial binding energy of 192 GHz atoms are

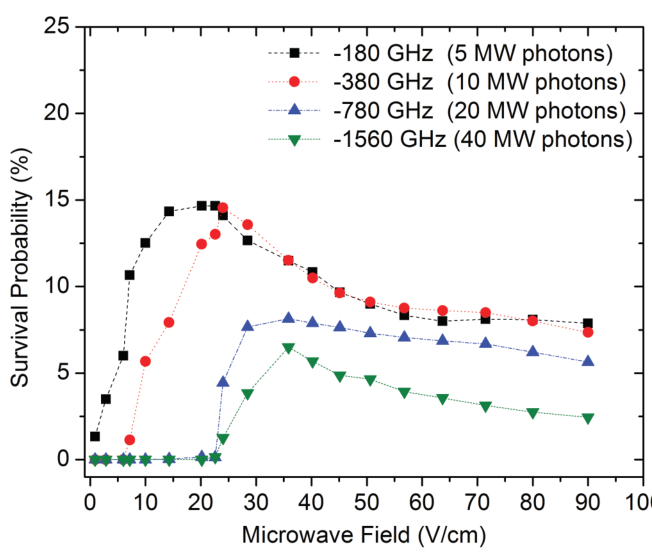

FIG. 7. (Color online) Fraction of atoms surviving the microwave field as a function of its strength when the laser frequency is tuned to four different Rydberg states, the same states as shown in Fig. 6. Only the extremely-high-lying states are detected (narrow 50-ns gate). The field ionization signals at microwave fields above 60 V/cm in Fig. 6 are due to atoms being left in high-lying states.

either transferred to highly excited states, which are ionized at a field of ≈2 V/cm (a time of 40 ns), or ionized. With an initial binding energy of 192 GHz atoms are not completely removed from the initial state until a microwave field in excess of 30 V/cm is reached. However, population begins to appear in the highly excited states at microwave fields below 10 V/cm. An initial binding energy of 386 GHz leads to similar results in that there is a field regime in which atoms are observed at both the initial binding energy, at a time near 80 ns, and in highly excited states, at a time near 40 ns. There is, however, some population observed between the initially excited state and the ionization limit. At binding energies of 386 GHz or less, large numbers of atoms are observed in high-lying states at fields below the 50% ionization fields. When the binding energy is increased to 770 GHz, it is no longer the case that atoms are found in deeply bound states and highly excited states over a range of microwave fields. The atoms are either deeply bound near $\Omega = 1$ or in highly excited states, with few atoms between these energies. At microwave fields between 10 and 30 V/cm it appears that population is transferred to more deeply bound states near $\Omega = 1$, where stable classical orbits exist in the microwave field. Higher-field amplitudes lead to ionization. Finally, when the initial binding energy is 1530 GHz it appears that microwave fields of 20 and 30 V/cm transfer population to states in which the microwave frequency is half and equal to the Kepler frequency, respectively. Above 30 V/cm only highly excited states are observed.

IV. DISCUSSION

With the exception of initial states within 400 GHz of the ionization limit, we observe that highly excited states begin to appear at the field at which 50% ionization occurs. In other words, the production of highly excited states appears to be related to microwave ionization. We suggest that it is in fact a continuation of microwave ionization. While this notion might seem strange, it is suggested by the simple man's model [16–18]. It predicts that electrons ionized at the peak of the microwave field have zero drift velocity, only an oscillation in the field, so the electron remains near the ion core, oscillating in the field. The simple man's model must be modified to take into account the Coulomb potential [19], but the basic idea is unchanged; microwave ionization at some phase of the microwave field produces electrons with zero time-average velocity. These electrons therefore remain in the vicinity of the ion core and are in some sense bound. In classical terms the electrons are trapped in orbits that are synchronous with the microwave field, so the electron does not, on average, gain energy from the field. In quantum mechanical terms they are quasistable atom-field or Floquet states [20]. In either case, when the microwave field is turned off over many field cycles, the atom is left in a highly excited state.

It is useful to compare our results to those of Zhao *et al.*, who examined both experimentally and theoretically the effect of trains of short unipolar pulses with an added bias field. They studied cases in which the scaled frequency Ω was both less than and greater than one. In the experiments, with trains of up to 40 pulses, they found atoms were most likely to be found in high-lying states subsequent to the pulse train if the bias field resulted in a time-average field of zero. This finding is similar

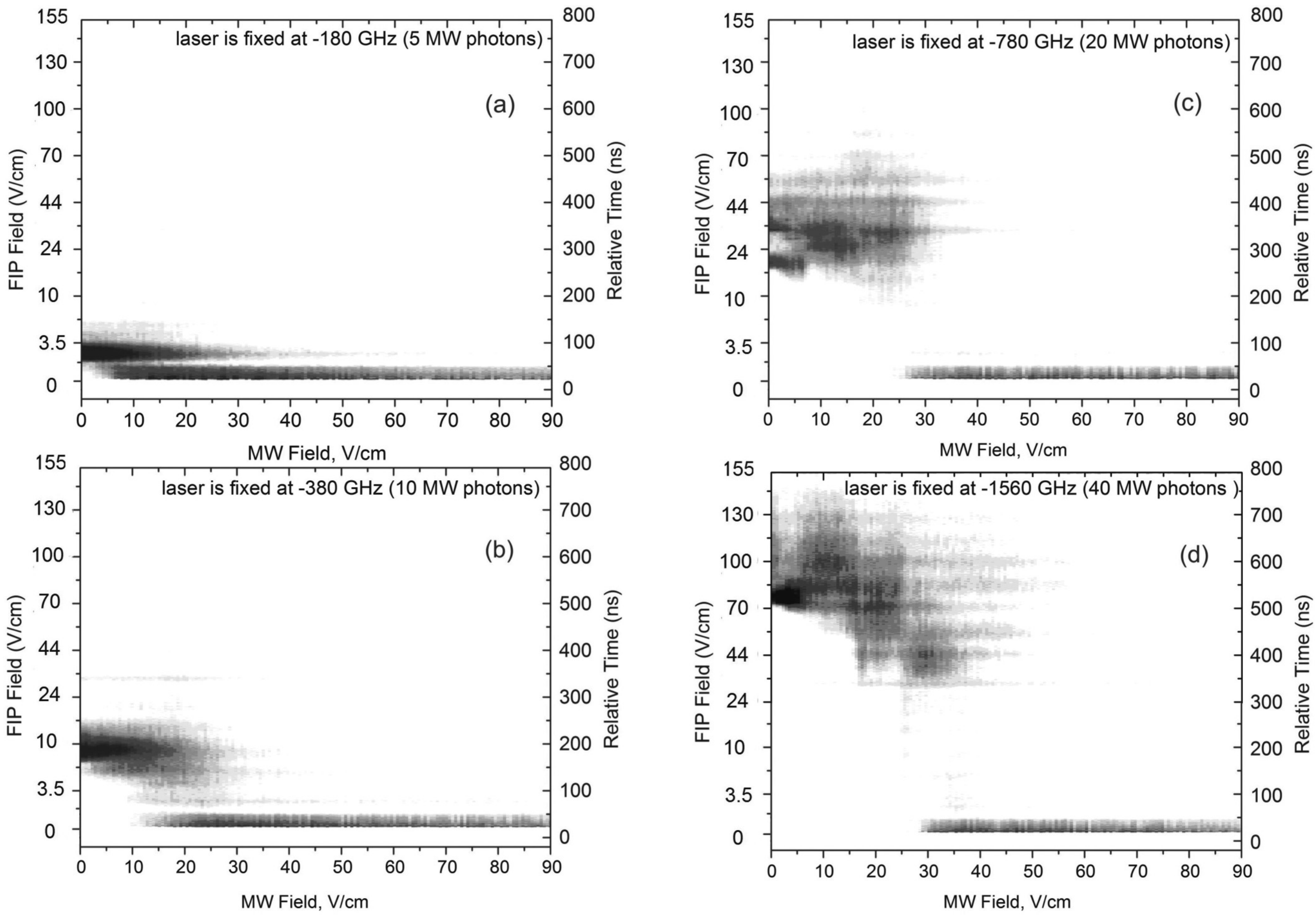

FIG. 8. Final-state distribution as a function of the MW field amplitude when the laser frequency is tuned to four different Rydberg states. As in Fig. 4, the distributions are grayscale representations of time-resolved field ionization signals. For convenience the vertical scale is shown both as time and field in the FIP. The FIP starts at 20 ns. Population is always found in extremely-high-lying states at fields when a large fraction of atoms is ionized. Redistribution of the population at lower microwave fields occurs, but atoms are in no case continuously distributed between the initial state and the IL. When the laser is tuned below −386 GHz, where individual Rydberg states can be resolved, the onset of the signal from the high-lying states coincides with the corresponding 50% ionization threshold.

to our observation that the high-lying states are only observed when the stray fields are less than 20 mV/cm.

The classical calculations of Zhao *et al.*, which included trains of up to 200 pulses, also show that the high-lying states survive longest when the bias field results in a vanishing time average field. They attribute the stability to the fact that the time-average field is zero, leading to a vanishing average force. With a train of 200 pulses the calculations show a 78% survival probability if the atoms are initially in a state for which $\Omega = 3.3$, but only a 10% survival if the atoms are in a relatively-low-lying state, one for which $\Omega = 0.3$. Assuming an exponential decay of the high-lying states, extrapolation of these results to our microwave pulses, which contain 7900 cycles, yields a survival probability of 5×10^{-5} for $\Omega = 3.3$ and 10^{-39} for $\Omega = 0.3$. These survival probabilities are much lower than our observed survival probabilities of 5% in both cases.

That the classical description does not reproduce our results is no doubt related to the fact that classical predictions of ionization fields in the $\Omega > 1$ regime are far too low [6,7]. Possibly the classical calculations miss the fact that it is possible to form stable atom-field or Floquet states in which energy flows back and forth between the electron and the field during each field cycle [20].

If the production of highly excited states is an inherent part of microwave ionization, why were they not observed long ago? Experiments with H were reported in 1974 and the first alkali-metal experiments in 1983 [21]. The most probable reason is that the early microwave ionization experiments were done under conditions in which there were static fields, which destroy the highly excited states. The initial alkali-metal experiments were done in waveguide cavities in which the atoms were 1 mm from a metal septum, with stray fields of at least 100 mV/cm. In later H experiments static fields depressed the ionization limit to $n = 114$, corresponding to a field of 3.4 V/cm [22]. In these experiments the amplitudes of the microwave fields were often in excess of 100 V/cm and stray fields of 100 mV/cm were not thought to be important. Finally, most microwave ionization experiments were done at lower frequencies, which allow stray fields a longer time to destabilize a synchronized orbit during each field cycle.

V. CONCLUSION

Here we report an investigation of microwave ionization, using the same apparatus, from low n, where $\Omega \approx 1/3$, to high enough n that single-photon ionization by the microwave field is possible. The fields required to effect 10% and 50% ionization decrease slowly with n in the $\Omega < 1$ regime and are almost constant in the $\Omega > 1$ regime, except very close to the ionization limit, where pronounced structure is observed in the ionization fields, due to the atoms' being left in highly excited states. These observations are consistent with previous measurements that covered pieces of the range of n explored here [7,8]. What is surprising is that the highly excited states are not only observed when laser excitation is to states very near the ionization limit. Rather, they are a general feature of microwave ionization, irrespective of the initial state. Although their existence may be surprising, the simple man's model suggests that states in which the electron oscillates in the field with no average velocity are formed. Apparently when the microwave field is turned off these states evolve into high-lying Rydberg states.

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