Production of extremely-high-lying states in 17-GHz microwave ionization of Li and Na

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In the ionization of Li and Na by a 16.91-GHz microwave field, roughly 20% of the atoms are not actually ionized but trapped in highly excited states within one or, at most, two microwave photons of the ionization limit. The highly excited atoms appear at the same field as the onset of microwave ionization, irrespective of whether ionization occurs at the field $E = 1/3n^5$ or $E = 1/16n^4$. The highly excited atoms are only detected in static fields less than 35 mV/cm, a field which roughly corresponds to field ionization of an atom bound by twice the microwave frequency. We also show that Na |m| = 1 atoms require a microwave field of $E = 1/9n^4$ for ionization in zero static field, but the required field for ionization is reduced to $E = 1/3n^5$ if a static field of 1 V/cm is present.

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I. INTRODUCTION

Ionization of ground-state atoms by intense laser pulses in both the tunneling and multiphoton regimes can result in the production of highly excited, or Rydberg, atoms as a by-product. An example of the former is the ionization of He by 1 GV/cm 800-nm pulses 30 fs long. It results in 10% of the atoms being left in states of principal quantum number n between 7 and 15, a rescattering process dubbed "frustrated tunnel ionization" [1,2]. In the multiphoton regime, with somewhat longer pulses, population is also observed to be left in the Rydberg states, but in this case it is attributed to multiphoton excitation brought into resonance by the ac Stark shift during the laser pulse [3,4]. Microwave ionization of Li by 38-GHz fields has shown a similar phenomenon: roughly 10% of the atoms are left in highly excited states within one or two microwave photons of the ionization limit when microwave ionization occurs [5]. Recent measurements of laser excitation in the presence of the microwave field suggest that the high-lying states are produced by resonant excitation, as in laser ionization in the multiphoton regime [6]. However, in earlier experiments the production of highly excited states in the recombination of electrons with ions induced by half-cycle pulses and microwave fields has been reported [7-9]. These are essentially rescattering processes, rather similar to frustrated tunnel ionization. In sum, with respect to the production of highly excited states, Rydberg atoms in microwave fields are similar to ground-state atoms in strong laser fields. The notion that Rydberg atoms and microwaves constitute a good system for the study of strong field processes is not new; the first microwave ionization experiments were undertaken to connect tunneling and multiphoton ionization [10].

Until the recent 38-GHz microwave ionization measurements, there was only one measurement which showed the production of high-lying states, but in that experiment atoms initially in states of $n \approx 80$ were exposed to a five-cycle 11.5-GHz pulse, which resulted in as much as 35% of the population being left in states of n > 120 [11]. In all other experiments, with microwave pulses thousands of field cycles long, after the microwave pulse, the population was apparently either in the initial state or ionized [12]. No population was found in intermediate states. Why are the recent 38-GHz Li results so different from the previous results? Is it a question of experimental technique, something special about Li, or the relatively high frequency of 38 GHz? Here we report the ionization of Li and Na by a 16.9-GHz microwave field to address this question. As we show, the way the previous experiments were done would have precluded leaving atoms in high-lying states or detecting them. Furthermore, there is nothing special about 38 GHz or Li. Atoms are left in high-lying states when both Li and Na are ionized by a 16.91-GHz field. Finally, we show that the Na microwave ionization field of $E = 1/3n^5$ is only valid for |m| = 0, where *m* is the azimuthal orbital angular momentum quantum number. It is only observed for |m| = 1 when there is a static field of ~ 1 V/cm present. In the sections which follow we outline the experimental technique, present our results, and discuss the conclusions which can be drawn from them.

II. EXPERIMENTAL APPROACH

This section starts with a brief description of the experimental method followed by a more detailed explanation of each component of the apparatus. A thermal beam of ground-state lithium or sodium atoms crosses the electric field antinode at the center of a 16.91-GHz Fabry-Perot microwave cavity. There atoms are excited to a Rydberg state by a sequence of transitions induced by 20-ns dye laser pulses. Following laser excitation, atoms are subjected to a microwave pulse, typically 200 ns long, as shown in the timing diagram of Fig. 1. About 300 ns later atoms are exposed to a 1- μ s rise time field ionization pulse (FIP). Depending on the polarity of the FIP, either electrons or ions are sent upwards to a dual microchannel plate (MCP) detector. The MCP signal is amplified, captured by a gated integrator or oscilloscope, and recorded in a computer for later analysis.

The Fabry-Perot microwave cavity consists of two brass mirrors 40.5 mm in diameter with 119-mm radii of curvature. The on-axis spacing between mirrors is 101.5 mm. The cavity is operated on the TEM₀₁₂ mode at a frequency of 16.91 GHz with Q = 3200. The output of a Hewlett Packard (HP) 83550B sweep oscillator with a 83550A rf plug-in is sliced into pulses by a DM186B General Microwave pin attenuator. The resulting pulse has a variable width and rise and fall times of approximately 15 ns. Going through an HP 8494B attenuator, it is amplified in a Hughes 8020H traveling wave



FIG. 1. (Color online) Experimental timing diagram. Rydberg states created in laser excitation are subjected to the microwave pulse. About 300 ns later a slowly rising FIP ionizes atoms, and either electrons or ions are detected by the MCP, depending on the polarity of the FIP.

tube amplifier. It is then sent through an HP P375A variable attenuator into the vacuum chamber using SMA cables and into the cavity through an iris in one of the cavity mirrors. The microwave system generates a 16.91-GHz pulse with 0-300 V/cm amplitude and a variable width. The microwave field amplitude is calibrated using the method described by Cheng *et al.* [13], and we are able to determine the amplitude of the pulse with an uncertainty of 15%.

The FIP is produced by a voltage pulse applied to the bottom plate. A negative (positive) voltage pulse leads to what we term a negative (positive) FIP and the detection of electrons (ions). The 300-ns delay between the microwave pulse and the FIP provides enough time for the electrons produced by photoionization or microwave ionization to leave the interaction region before the start of the FIP. Thus, with a negative FIP only atoms which are left in bound states after the microwave pulse are detected. Since the flight time of the electrons from the interaction region to the MCP is negligible compared to the rise time of the FIP, the time-resolved MCP signal yields the final bound-state distribution subsequent to the microwave pulse. With the 300-ns delay and a positive FIP we detect all ions produced: those due to photoionization, microwave ionization, and field ionization. With our maximum FIP, \sim 75 V/cm, we can ionize states as low as n = 60, and with a relatively small FIP, ~ 10 V/cm, we detect ions created by the microwave pulse, photoionization, and field ionization of only higher-lying states. Due to the fact that ions do not leave the interaction region in 300 ns and their long flight times to the MCP, it is impossible to distinguish a population left in the extremely-high-lying states from ions created by microwave ionization of the initial Rydberg state. (Laser photoionization is negligible.) It is, however, possible to discriminate between these ion signals and field ionization of atoms in states of $n \leq 90$. In the previous experiments, focused on microwave ionization of states of $n \leq 40$, ion detection was used, so it would have been impossible to discriminate between the production of high-lying bound states and microwave ionization.

To control or null the stray static field in the interaction region, the microwave cavity is surrounded by four copper plates, which, along with the cavity mirrors, can be grounded or separately biased to better control the static field. Low-pass filters are placed on all the bias leads but the one for the FIP, decreasing the effect of rf noise picked up by the plates or cavity mirrors. The minimum value of the static field we can obtain during this experiment is about 5 mV/cm; we estimate the static field by measuring the energy of the depressed ionization limit, a method discussed in detail elsewhere [5].

The experiment is triggered at the 1-kHz repetition rate of the frequency-doubled Nd:YLF laser 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. To excite Li atoms, the first 527-nm pulse pumps the 2s-2p and 2p-3sLittman-Metcalf-type [14] dye lasers, while the second one pumps a double grating Littman laser for the last 3s-np transition [15]. The linewidth of this laser is about 8 GHz, and its frequency can be continuously tuned to populate any Rydberg state of n > 20. A 52.42-GHz free spectral range etalon and an optogalvanic signal from the 16274.0212-cm⁻¹ $2p^{5}(2p_{3/2})3s-2p^{5}(2p_{3/2})2p$ Ne line provide relative and absolute frequency calibrations, respectively. Na atoms are excited via $3s \rightarrow 3p \rightarrow 3d \rightarrow nf$ transitions. The first 527nm pulse pumps a 3s-3p Littman-Metcalf dye laser. The second pump beam is used for a near-infrared Littman-Metcalf dye laser that drives the other two transitions. The linewidth of this laser is about 12 GHz. Amplified spontaneous emission of the second laser drives the 3p-3d transition, while the output of the oscillator populates an nf Rydberg state. The spontaneous emission can effectively drive the 3p-3dtransition, but it is not strong enough to excite an observable number of atoms to Rydberg states or to photoionize them. For both atoms, the dye 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. Unless stated otherwise, the laser field and microwave field are polarized vertically throughout the experiment.

III. OBSERVATIONS

A. Lithium

A major objective of the current work was to determine if population trapping in the extremely-high-lying states observed in 38.34-GHz microwave ionization also occurs in 16.91-GHz microwave ionization [5]. To detect atoms in these states, we use a negative FIP, narrow the integration gate to 50 ns, and position the gate in such a way that only signal from states that lie within 50 GHz of the ionization limit is recorded. We can scan the frequency of the 3s-np dye laser to obtain the spectrum of high-lying Rydberg states that remain bound after the microwave pulse. The spectrum obtained with a 300 V/cm microwave field is presented in Fig. 2. There is no signal above the limit, denoted by zero on the horizontal axis, because all photoionized electrons have time to leave before the FIP commences and are not detected. We denote the middle of the downward slope of this signal as the ionization limit depressed by the static field E_s present during the excitation. Explicitly, the limit is depressed by $\Delta W = 2\sqrt{E_s}$ [5]. Here and throughout the paper atomic units are used unless other units are given explicitly. In Fig. 2 the depressed ionization limit is at about -15 GHz, which corresponds to a 6.6 mV/cm static field. Figure 2 demonstrates that with a 300 V/cm microwave field we observe the high-lying states for initial states of $n \ge 21$, which is the lowest *n* state we can ionize



FIG. 2. (Color online) Field-ionization signal from the highlying states of Li after exposure to a 300 V/cm microwave (MW) pulse as a function of the laser frequency, in terms of energy relative to the ionization limit. The inset shows the spectrum in the vicinity of the ionization limit obtained with the same narrow integration gate when no MW field is present.

with a 300 V/cm microwave field. To verify that we only detect high-lying states, we show in the inset of Fig. 2 a spectrum taken with the microwave field off. The signal is nonzero only very close to the ionization limit, from weakly bound states with n > 250. Although the width of the peak, 50 GHz, is more than five times as wide as our laser linewidth, further reduction of the amplitude of the FIP or the gate width does not lead to a narrower peak, only one diminished in amplitude. We believe that reduced collection efficiency for electrons produced by a small FIP requires a FIP of amplitude large enough to ionize states of n > 250. Using a positive FIP we can detect a microwave ionization signal for $n \ge 21$. It is worth mentioning here that with the negative FIP we detect zero signal when the laser frequency is scanned over the ionization limit when the microwave field is on. Thus, electrons are not spatially trapped in a plasma by ions in the interaction region before the arrival of the FIP, and what we detect is indeed a population left bound in the extremely-high-lying states when a Rydberg state is exposed to a strong microwave field [16,17]. We have, in addition, checked that the observed high-lying state signals are not dependent on the number of Rydberg atoms excited, ruling out collisional effects which might lead to the production of high-lying states.

To determine the fraction of atoms excited to the extremelyhigh-lying states, we tune the laser to a specific Rydberg state, and, using a negative FIP and electron detection, record the electron signal as a function of the microwave field amplitude. We use a narrow 50-ns gate to measure the population left in the high-lying states and a wider gate to detect surviving atoms in all detectable ($n \ge 60$) Rydberg states. To compare our results to the previous microwave ionization experiments, we, as well, detect ions with a positive FIP, the amplitude of which is not high enough to field ionize the initial state, but



FIG. 3. (Color online) Microwave ionization of the Li 73 *p* state. (a) Fraction of atoms surviving the MW field as a function of its strength detecting electrons from all bound states of $n \ge 60$ (circles) and fraction of atoms apparently ionized by the MW field as a function of its strength observed by detecting ions with the positive FIP (squares). The apparent microwave ionization signal collected with the positive FIP is due to both microwave ionization and field ionization of atoms in state of $n \ge 90$. Because the high-lying states are present in both signals, they are not complementary. (b) Fraction of atoms detected in high-lying states, $n \ge 250$, subsequent to the microwave pulse. Fraction of atoms in such high-lying states is also estimated from graph (a) as *ionization probability minus (100% minus survival probability)* for any given MW power (squares). When the MW field exceeds 30 V/cm, the only bound population remaining is in states with n > 250.

ionizes atoms with n > 90. With a positive FIP, population trapped in the extremely-high-lying states is indistinguishable from ions due to microwave ionization, so we term the positive FIP signal due to these two sources the apparent microwave ionization signal. We recorded such data for several Rydberg states and show a characteristic plot in Fig. 3 for n = 73. The black squares in Fig. 3(a) represent the apparent microwave ionization signal obtained by detecting ions with the positive FIP, while the (green) circles represent the surviving atom signals obtained by detecting electrons from all bound states of $n \ge 60$ with a negative FIP. These two detection techniques do not yield complementary signals because of the presence of the high-lying states in both. This point is made by Fig. 3(b) in which we show the electron signals from high-lying states measured with the narrow gate. All the negative FIP signal (green circles) at microwave fields over 30 V/cm in Fig. 3(a) is due to high-lying states. A second point shown by Fig. 3(b) is that population in the high-lying states appears at the same field as does microwave ionization. Figure 3 shows that atoms exposed to the microwave pulse are left in the initial state or nearby states, in the very-high-lying states, or ionized. As in previous experiments, energy levels are not populated in the region between the initial and high-lying states. Most important, we observe a substantial population transfer to states with n > 250. For n = 73 it is nearly 30% at the microwave field just above the threshold for ionization, and it decreases as the microwave field is increased.

A striking result of this experiment is the dependence of the production of the high-lying states on the static field, as shown in Fig. 4 for two different Rydberg states, 24p and 58p. We use a narrow 50-ns gate to detect only high-lying states and record the signal while continuously scanning the bias voltage on one or two plates. An example of such measurement is presented in the inset of Fig. 4 for n = 58 with a microwave field of 6 V/cm, where the production of high-lying states is



FIG. 4. (Color online) Relative number of atoms in the highlying states with n > 250 after the MW pulse as a function of static field in the interaction region. The laser frequency is tuned to two states: n = 58 and 24. The inset shows a relative number of atoms in the high-lying states as a function of bias voltage on one of the plates after excitation of the Li 58*p* state and exposure to a 6 V/cm microwave pulse. The main figure shows the relative numbers of surviving high-lying atoms as a function of the static field for the Li 24p (—) and 58p states. The microwave fields are 215 and 6 V/cm, respectively. In both cases these microwave fields are just above the ionization threshold fields. The two curves are almost identical, and for both states no bound atoms are detected when the static field is about 35 mV/cm, a field that ionizes states as low as n = 360.

at a maximum. For each of the bias voltages, we determine the field in the interaction region by measuring the depression of the ionization limit. Since we are never able to eliminate completely the depression of the limit, we believe there to be a time-dependent stray field of approximately 6.6 mV/cm. Converting the bias voltages into fields, we construct the static field dependence for both initial states, as shown in Fig. 4. In both cases the microwave field is set just above the threshold field, where the production of high-lying states is maximized. Figure 4 shows that the n = 24 and 58 curves exhibit the same behavior: the maximum high n signal occurs at the lowest static field, and we detect no signal when the field is increased to approximately 35 mV/cm. The 35 mV/cm static field is large enough to ionize all the population in the high-lying states of n > 360. Returning to Fig. 4, we notice that the middles of the curves nearly coincide, so, all in all, it is impossible to say which curve belongs to which state just by looking at the graph. That suggests that there is no dependence on the initial state, as long as the microwave field is strong enough for ionization. The decrease of population in the high-lying states with increasing static field is most likely due to either of two effects: the nonzero static field is destabilizing the electron's orbit in the microwave field, or the static field is directly ionizing all population in these states. To show that the latter is the dominant effect, we minimize the static field and introduce a 100-ns square pulse of small amplitude that starts after the microwave pulse and ends before the FIP. Using several values of amplitude of the square pulse, we detected no difference in the survival probability of the high-lying states compared to a corresponding measurement in a dc static field. In short, the dependence is essentially that shown in Fig. 4. A similar result has been reported by Zhao *et al.* [18], who showed that exposure to unipolar pulse trains biased to produce zero time-averaged field leads to minimal ionization while a nonzero time-averaged field leads to rapid ionization. The 35 mV/cm static field is strong enough to ionize population within \sim 32 GHz, or two microwave photons, of the ionization limit according to $\Delta W = 2\sqrt{E_s}$. Hence, we can infer that the observed high-lying states are states with n > 360.

B. Sodium

Another important objective of this work is to show that the excitation to the extremely-high-lying states by microwave ionization is a general phenomenon inherent to all Rydberg atoms and is not peculiar to lithium. To do so, we have repeated the measurements for sodium. In this experiment we were able to lower the static field to 5 mV/cm, which depresses the limit by 13 GHz. As in Li, we observe excitation to the extremely-high-lying states with n > 250 after the microwave pulse. Scanning the 3d-nf laser frequency while detecting the high-lying states yields a spectrum essentially identical to that shown in Fig. 2. High-lying states are again observed for $n \ge 21$ because the maximum microwave field is the same.

In the following, the 3d-nf laser was fixed at several different states, and survival and ionization probabilities were taken as described in the previous subsection. In Fig. 5 we show the apparent microwave ionization signals obtained with the positive FIP by solid circles, triangles, and squares for the 24f, 30f, and 40f states, respectively. The static field is in all



FIG. 5. (Color online) Fractional apparent ionization for the Na 24f (solid circle), 30f (solid triangle), and 40f (solid square) states obtained with a positive FIP. The signals contain both microwave ionization and field ionization of high-lying states with n > 250. Vertical lines correspond to the 50% ionization threshold of |m| = 0 and 1 states measured by Pillet *et al.* [19]. Fractional high-lying state signals obtained for the 24f (open circles), 30f (open triangles), and 40f (open squares) states obtained with a negative FIP and a narrow integration gate. As in Li the high-lying states begin to be observed at the field at which microwave ionization starts, and in all cases roughly 15-20% of the atoms are left in the high-lying states.

cases 5 mV/cm. The open symbols represent the signals due to high-lying states observed with the negative FIP. As shown, we observe a substantial fraction of the initial population in states with n > 250 after the microwave pulse, with maximum transfer up to 20%. As none of these low Rydberg states can be ionized by the FIP, a normalization of the population in the high-lying states is done by comparison of the MCP signals to a corresponding signal recorded from one of the higher Rydberg states, for instance, n = 73 that can be completely ionized by the FIP. As in Li, the population observed in the high-lying states as the microwave field is increased, and the highly excited states appear at the same microwave field as the onset of microwave ionization.

It is useful to compare the microwave ionization thresholds, shown by the solid symbols of Fig. 5, to those reported by Pillet *et al.* [19]. In their work, with the laser polarization parallel to the microwave field only *nd* states of |m| = 0 and 1 were excited, and one threshold field for ionization was observed, with 50% ionization at $E = 0.28n^{-5}$, a field slightly lower than the Inglis-Teller field of $1/3n^5$. The Inglis-Teller field is the field at which the |m| = 0 Stark states of *n* and n + 1 intersect. For |m| = 0 and 1 the *n* and n + 1 energy levels do not cross but exhibit substantial avoided crossings due to the large quantum defects, 1.35 and 0.85, of the *s* and *p* states. Ionization of |m| = 0 and 1 states occurs at the field E = 0.

 $1/3n^5$ since a microwave field of this amplitude samples the avoided crossing between the n and n + 1 levels, allowing the $n \rightarrow n + 1$ transition to occur. The coherent effect of multiple cycles is required, and it is a resonant process [20]. Similar transitions through higher-lying levels occur, culminating in ionization. The vertical dashed lines in Fig. 5 mark the 50% |m| = 0 and 1 threshold fields measured by Pillet *et al.* With laser polarization perpendicular to the microwave field, so as to excite |m| = 0, 1, and 2 levels, they observed two thresholds, the |m| = 0 and 1 threshold at $E \cong 1/3n^5$, and the |m| = 2threshold at $E = 1/9n^4$, the static ionization field for the red hydrogen Stark state. In other words, the Na |m| = 2 states ionize by field ionization of the initial *n* state. Transitions through higher-lying states do not occur since the largest |m| =2 quantum defect is the d quantum defect of 0.015. Returning to Fig. 5, we see that the 24f and 30f ionization curves have ionization thresholds at $E = 1/3n^5$, as expected, but the 40 f curve exhibits two thresholds, not one. The first is at $E \cong 1/3n^5$, as expected, and the second is near $E = 1/9n^4 =$ 223 V/cm, the hydrogenic ionization field. We attribute the first 40 f threshold to |m| = 0 and the second to |m| = 1, ionizing by field ionization, as previously observed for |m| =2. While the 24 f and 30 f signals of Fig. 5 exhibit the expected threshold fields, they differ from those of Pillet et al. in that at most 40% ionization is observed, not 100%. From Fig. 5 alone it is not apparent that the 24 f and 30 f ionization signals reach only 40% ionization, but it becomes apparent when we add a static field. Figure 6 shows 30 f ionization threshold curves taken for several static fields. As the static field is raised from 10 mV/cm to 1.5 V/cm the |m| = 1 ionization threshold moves from $E = 1/9n^4$ to $E = 1/3n^5$. The curve taken at



FIG. 6. (Color online) Na 30 f signal obtained with the positive FIP vs microwave field for static field E_s values from 10 mV/cm to 1.5 V/cm. The vertical dotted line corresponds to the 50% ionization threshold as measured by Pillet *et al.* [19] at 0.28n⁵ for n = 30. With $E_s = 10 \text{ mV/cm}$ the |m| = 0 threshold is roughly 68 V/cm, and the |m| = 1 threshold is at $E = 1/9n^4 = 700$ V/cm, too high a field to be observed. As the static field is raised the |m| = 1 threshold moves to join the |m| = 0 threshold at $E = 1/3n^5$.



FIG. 7. (Color online) Fractional apparent ionization for the Na 55f state (solid squares) obtained with a positive FIP. The lasers are polarized perpendicular to the microwave field to excite |m| = 2 states. The obtained curve exhibits two thresholds: multiphoton ionization of |m| = 0 and hydrogenic tunneling ionization of |m| = 1 and 2. Fractional high-lying state signals obtained for the 55f (open circles) obtained with a negative FIP and a narrow integration gate. The population in high-lying states clearly exhibits two increases, corresponding to the multiphoton and tunneling ionization thresholds. Without the excitation of |m| = 2 levels the signal would decrease with increasing microwave field, as shown by the broken line.

1.5 V/cm is the same as observed by Pillet *et al.* [19]. The observations shown in Figs. 5 and 6 provide clear evidence that static fields on the order of 1 V/cm were present in the initial microwave ionization experiments done in a waveguide cavity [19]. Under such circumstances, even with electron detection no high-lying states would have been observed.

From Fig. 5 it is not apparent that ionization of the 40f|m| = 1 atoms leads to the production of high-lying states, but the onset of the |m| = 1 ionization is gradual, which obscures the production of additional high-lying states. To obtain a more definitive result, we have polarized the lasers perpendicular to the microwave field to excite $|m| \ge 2$ states, which exhibit a sharper ionization threshold. In Fig. 7 we show the apparent microwave ionization signal obtained with the positive FIP and the high-lying state signal obtained with the negative FIP for the 55 f state. The sharp hydrogenic $|m| \ge 2$ ionization threshold in the positive FIP signal at E = 51 V/cmis apparent, as is the corresponding increase in the high-lying state signal detected with the negative FIP. Figure 7 also shows a dashed curve mimicking a behavior of the high-lying states as the microwave field is increased in the case when 50% ionization threshold is at the Inglis-Teller limit. It is clear that there are more atoms detected in the high-lying states when the second threshold is reached and more atoms are being ionized. Evidently, the high-lying states are produced whether microwave ionization occurs by the absorption of many photons or by field ionization.

The static fields which affect the survival of high-lying states and those which alter the ionization threshold fields differ by almost two orders of magnitude. This point is made



FIG. 8. (Color online) The static field dependence of the highlying state signal and the microwave ionization probability of the 30f state after exposure to a 120 V/cm microwave pulse. Only states with |m| = 0 and 1 are prepared. The high-lying state signal was observed by detecting electrons with a negative FIP (solid line). The microwave ionization probability was observed by detecting ions with a positive FIP (dashed line). The half widths of the two curves differ by a factor of 40.

graphically by Fig. 8, in which we show, as functions of the static field, negative and positive FIP signals for the 30 f state exposed to a 120 V/cm microwave field. The solid line shows the high-lying state signal observed, which has a half width of 20 mV/cm. In contrast, the rise in the microwave ionization signal due to the |m| = 1 states, shown by the broken line, has a half width of 800 mV/cm.

IV. DISCUSSION

Since high-lying states had only been observed in the 38-GHz ionization of Li and in no other case, our original motivation was to see if there was something special about high frequencies or Li. The results reported here show that high-lying states with n > 360 are also observed in the 16.91-GHz ionization of both Li and Na, and they are probably produced quite generally in microwave ionization. The most probable reason for their not being observed previously is the requirement of such a low stray field, a field roughly four orders of magnitude weaker than the microwave fields used for ionization. It is not yet clear what sets the upper limit on the static field in which high-lying states can be observed, but it clearly increases with the microwave frequency. In the previous measurements at 38 GHz, the static field at which half the high-lying states disappeared was \approx 35 mV/cm, whereas in these measurements it is ≈ 17 mV/cm. These static fields depress the ionization limit by 33 and 22 GHz, respectively, so the requirement might be as simple as depressing the limit by the microwave frequency. Applying this criterion to ionization of a ground-state atom by an 800-nm laser yields a static field of 3×10^5 V/cm, one unlikely to occur accidentally. To

understand the origin of the static field limitation will require experiments over a wider frequency range.

An unexpected result of this work is the realization that stray fields almost certainly altered the Na microwave ionization threshold fields in previous experiments. Both |m| = 0 and 1 states were thought to ionize at $E = 1/3n^5$. Now it appears that the |m| = 1 states ionize by field ionization, at $E = 1/9n^4$, unless there is a field present, in which case ionization can occur at a microwave field as low as $E = 1/3n^5$. The dramatic effect of a small static field on microwave ionization was first observed by Pillet et al. [21], who found the same phenomenon in the microwave ionization of Li |m| = 0 states, which have 0.35 as the only quantum defect greater than 0.05. The Na |m| = 1 states are somewhat similar; 0.85 is the only quantum defect larger than 0.015. Why the small static field reduces the microwave ionization field from $1/9n^4$ to $1/3n^5$ can be understood as follows. The $n \rightarrow n+1$ transition is a resonant multiphoton transition which can be envisioned as being due to the near degeneracy of the dressed, or sideband, states formed from the Stark states of n and n + 1. If the quantum defect is small, the multiphoton Rabi frequency is as well, and the $n \rightarrow n+1$ transition does not occur unless the microwave frequency is almost perfectly resonant with the atomic multiphoton transition. In zero static field the dressed or sideband states of the same *n* are degenerate, but adding a small electric field lifts the degeneracy, and the sideband states from different Stark states form a quasicontinuum, allowing the resonance condition to be met. Their experiment showed that the static field E_s required is that which shifts the extreme Stark state by half the microwave frequency, i.e., $E_s = \omega/6n^2$. Applying this requirement to n = 30 with a frequency $\omega/2\pi =$ 16.91 GHz yields $E_s = 1.5$ V/cm, in good agreement with

Fig. 8. Apparently the Na p quantum defect alone is not large enough to allow microwave ionization at $E = 1/3n^5$, unless there is a field present to convert the dressed, or sideband, states from the Stark states into a quasicontinuum. The realization that there was a field of approximately 1 V/cm present in the earlier experiment implies that microwave ionization of the Na |m| = 2 states is unaffected by this field [19]. Apparently the field-induced quasicontinuum is too coarse grained for the extremely small |m| = 2 avoided crossings, and the multiphoton transitions do not occur.

V. CONCLUSIONS

This work shows clearly that the production of high-lying states in microwave ionization occurs quite generally; it is not peculiar to Li or high microwave frequencies, those in excess of 30 GHz. Furthermore, the example of Na shows that it occurs both when microwave ionization exhibits a $1/3n^5$ threshold field, indicating multiphoton ionization, and when it is exhibits a $1/9n^4$ threshold field, indicating field ionization. In experiments to date, high-lying states have only been detected if the static field is of order 10 mV/cm or less. The tolerable field increases with the microwave frequency, but its precise dependence on the microwave frequency is still an open question.

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