Resonant production of high-lying states in the microwave ionization of Na

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We report microwave ionization experiments with Rydberg states of Na using several microwave frequencies near 80 GHz. We observe substantial ionization of states as low as $n = 26$ with microwave pulses of 170-V/cm amplitude. Unlike experiments at 38 and 17 GHz, microwave ionization is not always accompanied by the production of the extremely high-lying states just below the limit. It only occurs when the initial Rydberg state is in multiphoton resonance with the high-lying states. The resonance condition is apparent at 80 GHz because the ponderomotive energy shift of the limit, and of the high-lying states, is small compared to the microwave frequency, even at fields strong enough to produce ionization. At the lower microwave frequencies, the ponderomotive shift exceeds the microwave frequency, ensuring that the resonance condition is met and obscuring its importance. The same is true of many laser experiments.

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I. INTRODUCTION

In the ionization of ground state atoms by intense laser pulses, weakly bound Rydberg atoms are observed [1–3], leading to two questions; how are they produced and why do they survive the strong field of the laser pulse? Two mechanisms have been suggested for the production of Rydberg atoms. In recent observations of "frustrated tunnel ionization" the production of high-lying bound states has been attributed to rescattering of the electron from the ion core by the strong laser field [2,3]. In contrast, in earlier experiments the production of excited atoms was attributed to resonant multiphoton excitation, with the requisite tuning provided by the ac Stark shift due to the laser pulse [4–7]. Usually the ponderomotive shift of the ionization limit and excited bound states provided the tuning [4–6]. The ponderomotive energy $E^2/4\omega^2$ is the energy of oscillation of a free electron in the field $E\sin\omega t$. The large ponderomotive energy shifts and averaging over the focal volume obscured in most cases the resonant nature of the excitation to the high-lying states. However, if the averaging over the focal volume can be eliminated, the resonant character of the multiphoton excitation is clearly displayed. By detecting the ionization from a small spatial region of the laser focus, Jones observed the interference between multiphoton amplitudes on the rising and falling edges of a laser pulse [6]. Analogous microwave multiphoton transitions, in which all atoms saw the same field, have shown the same interference effect [8,9]. Finally, while it is very difficult to observe explicitly multiphoton resonance effects in laser experiments, it is straightforward to observe them in numerical simulations. In particular, calculations of Muller show that many phenomena are attributable to the resonant production of the high-lying states [10].

Although it may seem counterintuitive that a weakly bound Rydberg atom can survive a strong rapidly oscillating field, in a Rydberg atom the electron spends most of its time far from the ionic core. There the only effect of the field is to superimpose the rapid ponderomotive oscillation on the slow orbital motion of the electron, with no time average energy exchange. Only when the Rydberg electron returns to the ionic core, where it is moving rapidly, can it, together with the ionic core, absorb energy from the rapidly oscillating field. Consequently, if in multiphoton ionization an electron is launched from the core with slightly less energy than needed to escape from the core, and its Kepler orbital period is longer than the laser pulse, the resulting Rydberg atom survives the laser pulse, as first pointed out by Jones *et al.* [1] and verified in several contexts [11–13].

Here we describe the production of highly excited states (HLS) in the ionization of Na Rydberg atoms by 79-GHz microwave fields. The upper limit to the binding energy of the HLS we detect is 50 GHz, i. e., states of $n > 250$. The combination of the small ponderomotive shift and high microwave frequency allows us to observe explicitly the resonant nature of the production of the HLS. The resonant nature of the excitation of the HLS precludes their being formed by rescattering driven by the microwave field. Rather, they return to the ion core because they are reflected by the long range Coulomb potential, as suggested by Muller [10]. In the sections which follow we describe the experimental approach, present our observations, and discuss their implications.

II. EXPERIMENTAL APPROACH

In the experiment a thermal beam of Na atoms passes through the center of a Fabry-Perot microwave cavity where the atoms are excited by the route $3s \rightarrow 3p \rightarrow 3d \rightarrow nf$ by three 20-ns long laser pulses at a 1-kHz repetition rate. In most of the experiments described here, the atoms are exposed to a microwave pulse from 200 ns to 1 μs long subsequent to laser excitation. In this case microwave ionization of the initially excited Rydberg state, typically bound by fifty times the microwave photon energy, corresponds to ionization of ground state atoms by an intense laser pulse. The microwave field can also be present during the laser excitation, allowing the observation of the atom-field, or Floquet, states in a constant amplitude microwave field. Subsequent to the microwave field we can detect either the ions or electrons resulting from microwave ionization and field ionization by applying a field pulse 300 ns after the microwave pulse. The field pulse drives either ions or electrons to the microchannel plate detector. If we detect ions, we detect both free ions and the atoms in the

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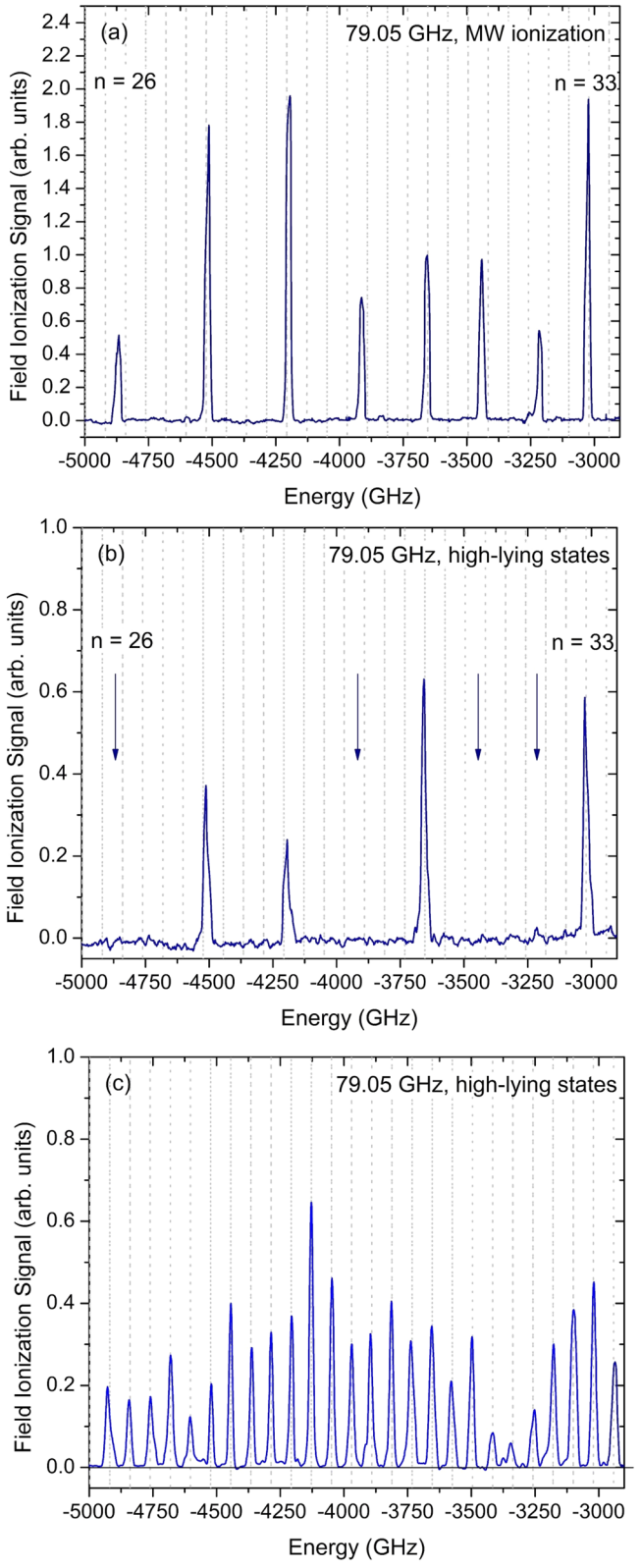

FIG. 1. Frequency scans of the third laser over the energy range from 5000 to 3000 GHz below the ionization limit, encompassing states from $n = 26$ to 33. The horizontal scale is the energy relative to the ionization limit. (a) Microwave ionization signal observed when the atoms are exposed to a 200-ns long, 170-V/cm, 79.05-GHz microwave pulse subsequent to laser excitation. For all states at least 40% of the atoms initially excited are ionized by the microwave pulse. (b) The same timing sequence and the microwave pulse as in (a), but atoms in the HLS are detected instead of microwave ionization. No HLS are observed for $n = 26$, 29, 31, and 32. The arrows indicate where the HLS signals should appear. The HLS are only observed when there is a multiphoton resonance between the initial Rydberg states and the HLS. The vertical dotted lines indicate the calculated locations of the Floquet states based on the HLS. (c) Spectrum observed when the laser excitation occurs in the presence of the 170-V/cm 79.05-GHz field. In this case a regular series of peaks 79.05 GHz apart is observed, extending from the limit to more than 5000 GHz below the limit. The peaks are Floquet states containing the HLS and occur at the locations of multiphoton resonances with the limit. The n states in (b) for which the HLS are observed are in multiphoton resonance with the HLS.

HLS. The latter are ionized early in the field pulse and can not be distinguished from free ions resulting from microwave ionization. If we detect electrons, the 300 ns time delay allows all electrons due to photoionization or microwave ionization to leave the interaction region, and we detect only bound atoms with the field pulse. Unless noted otherwise, the amplitude of the field pulse is ~0.4 V/cm, which ionizes states of $n > 250$, so the only bound atoms we detect are those in the HLS. The signal from the microchannel plates is captured in a gated integrator and saved in a computer for later analysis.

A critical aspect of the experiment is the reduction of the stray static fields to 3 mV/cm. Static fields in excess of 35 mV/cm ionize the HLS [14,15]. To this end we apply bias voltages to the two cavity mirrors and four plates surrounding the cavity. The Fabry-Perot cavity is composed of two brass mirrors of radius of curvature 52 and diameter 69.7 mm. The on-axis mirror separation can be varied from 93 to 89.7 mm, which allows us to have resonant frequencies from 77.02 to 79.8 GHz. We can change the resonant frequency by changing the mirror separation and the use of different TEM_{00n} cavity modes. Power is coupled into the cavity from a WR-12 waveguide through a 1-mm diameter iris in one of the cavity mirrors. The Q of the cavity is 8600, resulting in a filling time of 17 ns. The microwave source is a Hewlett Packard (HP) 8350B/83550A sweep oscillator operating at about 19.75 GHz. Its continuous wave output is sliced into pulses from 200 ns to 1 μs long, which are frequency doubled by a Phase One SP40 active doubler to 39.5 GHz and amplified with a Microsemi L3640-37 amplifier to a maximum power of 1 W. The 39.5 GHz microwaves pass through a HP R382A precision attenuator and then through the vacuum wall of the beam apparatus via a WR-28 waveguide. Inside the vacuum chamber the microwaves drive a Virginia Diodes D80R7 passive frequency doubler to generate the 79 GHz microwaves, which are transported to the cavity via a WR-12 waveguide. The relative power of the 79-GHz microwaves is measured on a continuous basis with a crystal detector on a 10-dB directional coupler in the WR-12 waveguide. The crystal detector was calibrated absolutely using a thermal power meter. We estimate the uncertainty in our field measurement to be 15%.

III. OBSERVATIONS

Striking evidence of the resonant nature of the production of the HLS is provided by Fig. 1, in which we show the signals

obtained when scanning the frequency of the last laser over the range from −5000 to −3000 GHz, which includes states of $26 \leqslant n \leqslant 33$. Figures 1(a) and 1(b) show the results of exposing the atoms to a 200-ns long 79.05-GHz microwave pulse of peak amplitude 170 V/cm, beginning 50 ns after the last laser pulse. Figure 1(a) shows the spectrum obtained when detecting the ions from microwave ionization and the HLS. Similar numbers of atoms are excited to each nf state in this range, but the fractional ionization varies due to the nonmonotonic variation in the ionization fields with n. In early microwave ionization experiments with Na, at microwave frequencies less than 20 GHz, a monotonic decrease in the ionization fields with n was observed, and the ionization fields were independent of the microwave pulse length from 200 ns to 1 μs [16]. As the microwave frequency is raised, low order multiphoton resonances begin to play a more important role, leading at 38 GHz to a nonmonotonic decrease in the ionization fields with n [17]. The nonmonotonic behavior is more pronounced at 79 GHz, and there is an obvious dependence of the microwave ionization probability on pulse length [18]. The important point, though, is that all states of $26 \leqslant n \leqslant 33$ exhibit substantial, roughly 50%, microwave ionization. In Fig. 1(b) we show the result of scanning the laser frequency when detecting the HLS by delayed electron detection. The HLS are only observed for $n = 27$, 28, 30, and 33; $n = 26$, 29, 31, and 32 are conspicuously absent. Why they are missing for $n = 26$, 29, 31, and 32 is suggested by Fig. 1(c), in which we show the spectrum obtained when detecting electrons with the delayed field pulse and laser excitation occurring in the presence of a constant 170-V/cm 79.05-GHz microwave field (the microwave pulse was simply moved to an earlier time). The resulting spectrum is a series of peaks spaced by the microwave frequency. It is simply a section of the spectrum shown in Fig. 2, which shows that the series of peaks extends from 5000 GHz below the limit to 500 GHz above the limit. A peak is located at the energy of the HLS, just below the limit, and all other peaks are displaced from it by multiples of the microwave frequency. Similar spectra have been observed with a 38-GHz microwave field [14]. In short, the peaks in Fig. 1(c) represent Floquet states containing the HLS [19–21]. Their temporal evolution is periodic, with the microwave frequency, and they are found at the energies which are in multiphoton resonance with the HLS. Comparing Figs. 1(b) and 1(c) shows that the initial states for which we observe the HLS are all multiphoton resonant with the HLS, while the missing states in Fig. 1(b) are not. To emphasize this point, in Fig. 1 we have added vertical dotted lines at the energies integral multiples of the microwave frequency below the ionization limit to show the locations of multiphoton resonances to the HLS.

To confirm that the production of the HLS is a resonant process we have tuned the frequency. Specifically, we have used the frequencies 77.27, 77.75, 78.67, 79.05, 79.23, 79.42, 79.65, and 79.77 GHz. This tuning range, 2.5 GHz, moves the Floquet states ∼4000 GHz below the limit by 125 GHz, which is adequate to probe the resonant behavior using an 80 GHz microwave field. When the microwave frequency is changed, and the amplitude and pulse length kept the same, the microwave ionization spectra are very similar to that shown in Fig. 1(a). In contrast, in the HLS spectra analogous

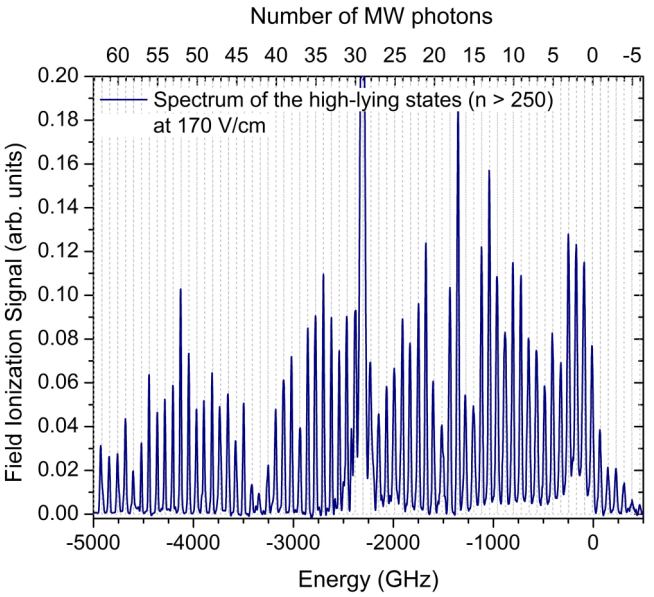

FIG. 2. Frequency scan of the third laser from 5000 below the limit to 500 GHz above the limit. The laser excitation occurs in the presence of a 170-V/cm 79.05-GHz microwave field, the same conditions as in Fig. 1(c). The spectrum shown in Fig. 1(c) is simply a section of this spectrum. As in Fig. 1, the vertical dotted lines indicate the calculated locations of the Floquet states based on the HLS.

to Fig. 1(b), different n states are missing for each cavity frequency. As a representative example, in Fig. 3 we show the HLS spectrum obtained at 79.65 GHz. The HLS are produced for only $29 \leqslant n < 32$; for $n = 26$–28 and 32, no HLS are

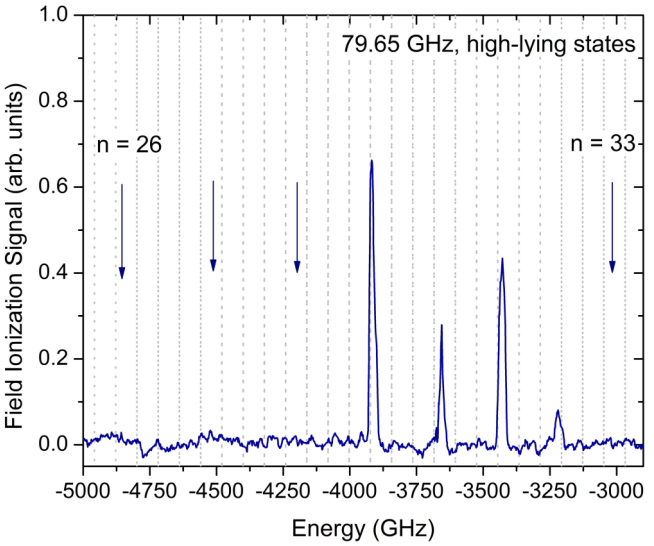

FIG. 3. Frequency scan over the same range as shown in Fig. 1. The microwave pulse is subsequent to laser excitation of the Rydberg states, and atoms in the HLS are detected. The microwave field is again 170 V/cm, but the frequency has been changed to 79.65 GHz. Now the HLS are produced for different n states than in Fig. 1(b). The arrows again indicate missing states. As in Fig. 1(b) the dotted lines indicate the calculated locations of the Floquet states based on the HLS. Only initial states in multiphoton resonance with the HLS lead to the observation of the HLS.

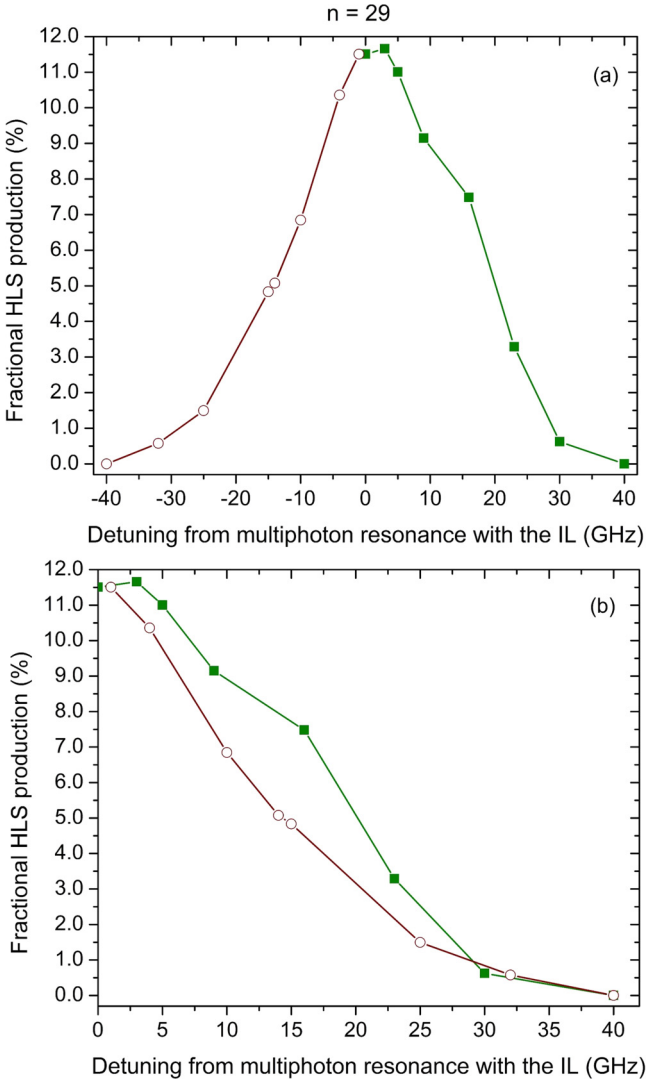

FIG. 4. (a) Fractional population in the HLS following the excitation of the $29f$ state vs the detuning between the $29f$ state and the multiphoton resonance with the HLS at the ionization limit (IL). (b) The same data as in (a) but plotted vs the magnitude of the detuning to show the difference between positive detunings (■) and negative detunings (○). The difference arises because the ponderomotive energy shift brings small positive detunings into resonance.

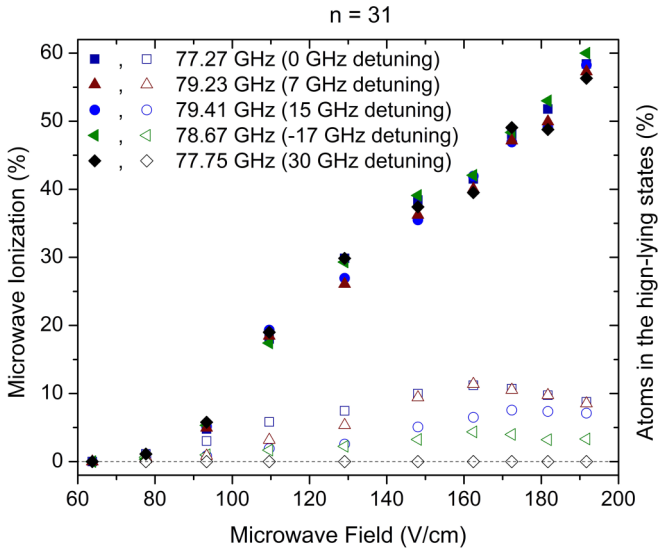

FIG. 5. Microwave ionization and the HLS production subsequent to the initial excitation of the $31f$ state vs detuning from multiphoton resonance with the HLS. The microwave ionization signals are shown as solid symbols and the HLS production as open symbols. 0 GHz detuning (77.27 GHz) (■,□), 7 GHz detuning (79.23 GHz) (▲,△), 15 GHz detuning (79.41 GHz) (●,○), −17 GHz detuning (78.67 GHz) (◀,◁), and 30 GHz detuning (77.75 GHz) (◆,◇). The microwave ionization signals are essentially identical, but the number of atoms left in the HLS is peaked at the multiphoton resonance with the HLS.

produced. As in Fig. 1(b), in Fig. 3 the energies of states not in multiphoton resonance with the HLS are indicated by arrows, and the vertical dotted lines show the location of the multiphoton resonances with the HLS.

A more systematic way of showing the resonant nature of the multiphoton transition to the HLS is presented in Fig. 4(a), where we show the HLS production starting from the $29f$ state as a function of the detuning between the $29f$ state and the multiphoton resonance with the HLS. The detuning is the $29f$ energy minus the energy of the nearest multiphoton resonance, or Floquet state, at zero microwave field. The detunings are accomplished by changing the cavity frequency as described earlier. In all cases the peak microwave field amplitude was 150 V/cm. As shown, the peak signal occurs near zero detuning, and the full width at half maximum of the resonance in Fig. 4(a) is 30 GHz. Also apparent, Fig. 4(a) is not symmetric. To emphasize the asymmetry, in Fig. 4(b) we show the HLS signal as a function of the magnitude of the detuning. At the half maximum there is a 7 GHz displacement between the two curves of Fig. 4(b), which is approximately equal to the ponderomotive shift $U_P = 9.5$ GHz at $E = 150$ V/cm. This displacement is expected since the ponderomotive shift brings the frequencies with positive detuning nearer to resonance and shifts frequencies with negative detunings further from resonance.

The 30 GHz width of the observed resonances can be understood in the following way. First, from the spectra of Figs. 1(b) and 2, we can extract the intrinsic width of the band of the HLS, or at least the upper limit of its width. The observed widths 3000 GHz below the limit, at $n = 30$, are ∼20 GHz. The laser linewidth is 10 GHz, so the intrinsic width of the band of the HLS is 15 GHz. This width is consistent with field ionization measurements which show that the band of the HLS extends no more than 50 GHz below the ionization limit. Since ionization and the production of the HLS from $n = 31$ are observed in microwave fields from 100 to 150 V/cm, it is reasonable to expect the resonance of Fig. 4(a) to be broadened by 5 GHz, the difference in the ponderomotive shifts in these two microwave fields. Accordingly, we expect the resonance of Fig. 4(a) to be 20 GHz wide, in qualitative agreement with the observed width of 30 GHz.

The dependence of the microwave ionization and the HLS signals on the microwave pulse amplitude is instructive, and in Fig. 5 we show these signals for the $31f$ state for several detunings of the multiphoton resonance with the HLS from

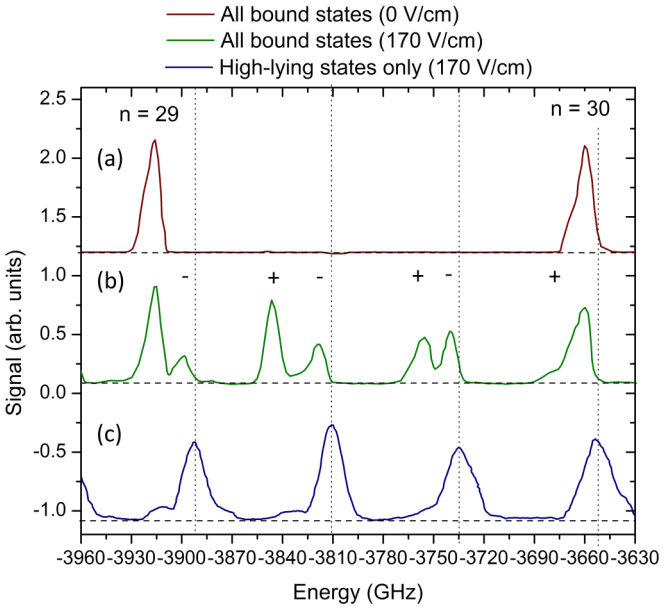

FIG. 6. Spectra observed when scanning the laser over the range including $n = 29$ and $n = 30$. (a) Laser excitation occurs in zero field, and the atoms are subsequently ionized by a 170-V/cm 79.05-GHz microwave pulse. (b) Atoms are excited in the presence of the 170-V/cm microwave pulse and ions are detected. In this case the only effect of the microwave field is the generation of sidebands displaced from the zero field states of (a) by multiples of 79.05 GHz. There are no measurable shifts. The lower $n = 30$ sidebands are indicated by $-$ and the upper $n = 29$ sidebands by $+$. (c) Spectrum observed when laser excitation occurs in the presence of the 170-V/cm field, and the HLS are detected. A spectrum similar to those shown in Figs. 1(c) and 2 are observed. This spectrum has no particular relation to the one shown in (b). As before, the dotted vertical lines indicate the calculated locations of the Floquet states based on the HLS.

the $31f$ state. The open symbols are the HLS signals, and the filled symbols are the true microwave ionization signals, that is the signals obtained using ion detection minus the HLS signals obtained using electron detection. As shown, the microwave ionization signals are independent of the detuning, but the HLS signals are not, as suggested by Figs. 1, 3, and 4. No detuning results in the largest HLS signal, and 30 GHz detuning results in no HLS signal. In addition, there is a noticeable difference between detunings of 15 and -17 GHz. The ponderomotive shift reduces (increases) the 15 (-17) GHz detuning from resonance, resulting in the observed difference. Finally, we note that the onsets of microwave ionization and the HLS production occur at the same field.

We have implicitly assumed that the initial Rydberg state is not shifted by the microwave field, and to confirm the validity of this assumption we have recorded the excitation spectra of the bound atoms in the presence of the microwave field. In Fig. 6 we show the bound state spectra from $n = 29$ to $n = 30$ in zero field and in a 170-V/cm microwave field. As shown, the principal effect of the microwave field is to generate sideband states displaced by integral multiples of the microwave frequency from the zero field $n = 29$ and $n = 30$ energies. There is no observable shift of the levels.

IV. DISCUSSION

The observations made with the 79 GHz microwave fields provide several insights. First, the width of the band of the HLS, 15 GHz, combined with the 10 GHz depression of the limit by the stray field places the center of the band of the HLS 28 GHz below the zero field limit, at $n = 350$, which has a Kepler orbit time of 6.6 ns, a time much shorter than our microwave pulse lengths. We do not, however, know when the HLS are produced during the microwave pulse, so it is not

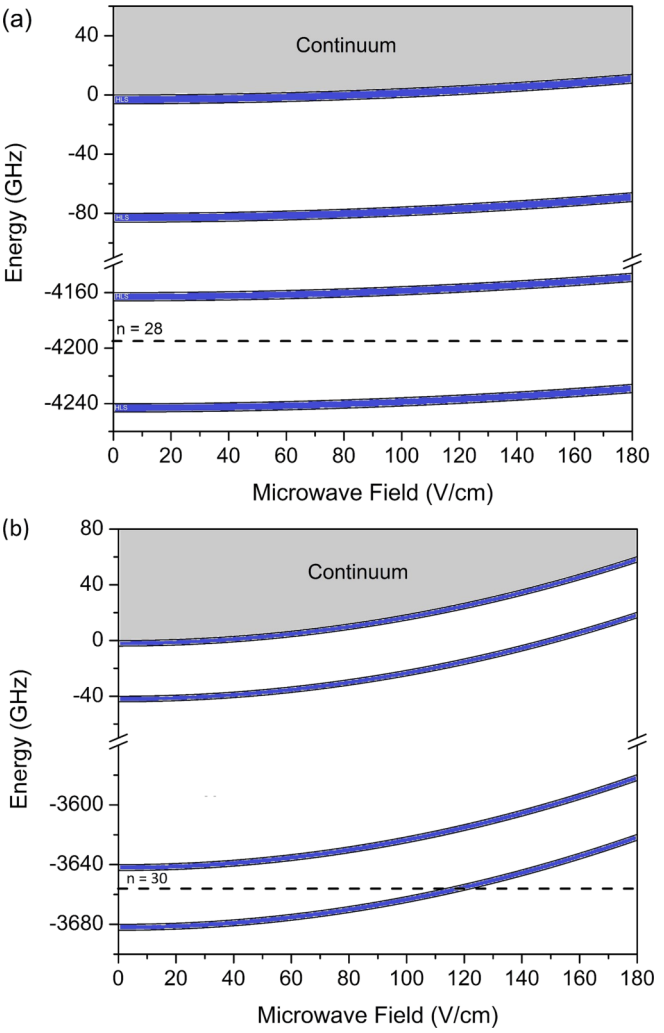

FIG. 7. (a) Energy level diagram vs amplitude of an 80-GHz microwave field. The band of the HLS is shown as a dark shaded band just below the continuum. It exhibits the ponderomotive shift to higher energy at higher microwave fields. There are Floquet states containing the HLS at integral multiples of 80 GHz below the HLS as shown. The $n = 28$ state lies between two of the Floquet states and does not come into resonance with the HLS before ionization occurs, and no HLS are produced. The HLS are only produced when a multiphoton resonance with the HLS exists. (b) The same energy level diagram as in (a) except that the frequency of the microwave field is 40 GHz. The ponderomotive shift is increased by a factor of 4 and the spacing between the Floquet levels is halved. Now, even a state with the maximum detuning, as shown for $n = 30$, comes into a multiphoton resonance with the HLS, and the HLS are produced for all initially populated Rydberg states.

obvious how long they must survive in the microwave field to be detected. In general, the orbit time is hundreds of times longer than the microwave period, so an electron returns to the core at a random phase of the microwave field and is equally likely to lose or gain energy in its encounter with the core. As a result, we expect an easily detectable fraction, ∼10% of the HLS produced, to survive several encounters with the core.

The most important aspect of this work is that these 79 GHz measurements show explicitly that the production of the HLS is a resonant process, occurring as shown in Fig. 7. The ponderomotive shift brings the band of the HLS into resonance with the initial state, and the HLS are produced if the multiphoton coupling is strong enough. At 79 GHz the ponderomotive shift is negligible, as shown in Fig. 7(a), so in zero field the initial atomic state must be very nearly N microwave photons below the limit. In microwave ionization by 17 and 38 GHz microwave fields as much as 20% of the initial population is also left in the HLS [17,22]. We suggest that the HLS are also produced by the same resonant multiphoton excitation in those cases, although the lower frequency and attendant larger ponderomotive shift obscure the resonant nature of the process. As shown in Fig. 7(b), drawn for 38 GHz, the ponderomotive shift can easily exceed the microwave frequency, ensuring that the resonance condition is met no matter what the energy of the initial state, as in the early laser experiments [4,5,7].

The third significant observation is shown in Fig. 5. The onsets of microwave ionization and the production of the HLS occur at the same microwave field, suggesting that they are the same process with slightly different final states, that is, the electron leaves the ion core with just enough or not quite enough energy to escape. For the measurements reported here, this connection is not surprising; the experiments are clearly in the multiphoton limit. Direct or tunnel ionization requires a field of $E = 1/9n^4$, and in a 150 V/cm microwave field it can only occur for $n > 42$. These 79 GHz experiments were done with $n = 30$ atoms, so direct or tunnel ionization is not possible. However, previous experiments on microwave ionization of Na at 15 and 17 GHz show that $m > 0$ states exhibit ionization threshold fields of $E = 1/9n^4$, i.e., by direct or tunnel ionization [16,22]. Here m is the azimuthal orbital angular momentum quantum number. In spite of the fact that the ionization mechanism is direct ionization, the production of the HLS also begins at this field, as shown by Fig. 7 of Ref. [22]. This observation is paralleled by the observation of resonant structure in the electron energy spectra observed in laser ionization in the tunneling limit with an infrared laser [23]. These observations are also ascribed to a multiphoton resonance [24].

V. CONCLUSIONS

The key insight which can be extracted from these measurements of ionization by a 79 GHz microwave field is that the production of the HLS is a resonant process, as proposed some time ago [10]. Due to the small ponderomotive shift at 79 GHz the resonance requirement is quite apparent, while at lower microwave frequencies and in many laser experiments it is not. The fact that the HLS are produced in a resonant process, necessarily requiring more than one microwave cycle, rules out rescattering driven by the microwave field as the source of the HLS. Rather, the HLS are due to reflection of the electrons by the long range Coulomb potential, as suggested by Muller [10]. Finally, it is almost certainly the case that the HLS reported here are intimately related to the zero energy electrons observed to accompany ionization by an intense infrared laser field [25,26].

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