Microwave Multiphoton Transitions between Rydberg States of Potassium

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We report the observation of sequences of from 1- to as many as 28-photon transitions between potassium $(n+2)s$ states and the lowest-energy Stark states of the n manifolds for $n=15-18$. The sequences are observed by the scanning of a static electric field from the field at which these pairs of levels nearly cross, to zero field where they are well separated. The maximum number of photons absorbed is observed to be proportional to the microwave field.

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The development of the laser has led quite naturally to the study and exploitation of highly nonlinear phenomena such as multiphoton excitation and ionization. For example, the 22-photon ionization of He by 1.06-μm light, scaling as $I^{21(2)}$, has been observed for $I \sim 5\times10^{14}$ W/cm^2.[1] Not surprisingly, describing such highly nonlinear processes by a high-order perturbation calculation is awkward and tedious. If, however, we express the above intensity as an electric field we see that it is $\sim 4\times10^8$ V/cm, a value comparable to the classical field required for ionization of a ground-state He atom, 3×10^8 V/cm.[2] The similarity of the field strengths together with the inherently low frequency of the radiation used to drive multiphoton processes suggest that approaching the problem as the interaction of an atom with a strong time-varying field might be profitable.

It has been appreciated for some time that one of the more fruitful ways of studying atoms in strong fields is to use Rydberg atoms. Since the Coulomb field experienced by the Rydberg electron at its classical turning point varies as n^{-4}, n being the principal quantum number, modest, easily characterized external electric fields produce significant effects. For example, the classical field required to ionize an $n=20$ atom is 2 kV/cm.[2] Furthermore, transitions to many nearby states are possible at microwave frequencies. These realizations led Bayfield and Koch to undertake their original measurements of the microwave ionization of hydrogen.[3]

Several recent ionization experiments on nonhydrogenic Rydberg states[4-6] with 8–15-GHz microwave fields have yielded results which differ dramatically from those obtained with static or quasistatic (~ 1 MHz) fields. Specifically, microwave field amplitudes of $\sim 1/3n^5$ (atomic units) have been required to ionize Na and He atoms of principal quantum number n, in marked contrast to the static-field requirement of $1/16n^4$.[4-6] The $1/3n^5$ field variation has been attributed to the rate-limiting step in the ionization process: a Landau-Zener transition from n to $n+1$ states, which occurs at or near the avoided crossing of the extreme upward-shifted n Stark state with the extreme downward-shifted $n+1$ Stark state, at a static field of $1/3n^5$. The transition occurs when an atom initially in level n is brought to the avoided crossing by the microwave field. If the field amplitude is enough to reach the avoided crossing and the size of the avoided crossing is roughly comparable to the microwave frequency, the transition occurs. Otherwise, it does not.

Given the usefulness of Rydberg atoms to an understanding of atoms in strong fields, it seems appropriate to study multiphoton microwave absorptions in Rydberg atoms as a way to understand analogous processes at much higher frequencies and intensities in ground-state atoms. A number of multiphoton microwave absorptions have been observed in atomic hydrogen[7,8] and another related effect, optical-microwave double resonance with many microwave photons, has been observed in hydrogen and sodium.[9,10] Such double resonances can also be viewed as optical transitions from a lower-lying state to microwave field produced sideband or Floquet states of a Rydberg state. The sideband states are easily described in terms of a Bessel function expansion.[11,12] As expected on the basis of the properties of Bessel functions,[13] the sidebands of a Stark state which has a linear Stark shift are observed to extend as far in frequency as the Stark shift produced by a static electric field of the same magnitude.[9,10] There is also, in priniciple, a decreasing tail of sidebands beyond this point which falls in amplitude by a factor of 10 in two sidebands.[13] Thus the Na $n \rightarrow n+1$ Landau-Zener transition mentioned previously can also be viewed as the overlap of the n and $n+1$ Floquet states when there is adequate coupling.[10]

To demonstrate more clearly this correspondence between the overlapping of sidebands and the microwave Stark shifting of states towards an anticrossing, it is desirable to make a systematic study of the intensity, field, and frequency dependence of microwave multiphoton transitions between two nearby Rydberg states. One such study was undertaken in helium, where a magnetic field was used to tune various Rydberg levels into resonance with one or more microwave photons,[14] although the multiphoton transitions were not easily identified.

Here we report the observation of entire sequences of multiphoton microwave resonances in potassium. Specifically, we have observed sequences of $(n+2)s \rightarrow (n,3)$ 1- to 28-photon transitions where (n,n_1) denotes the Stark state of principal quantum number n which adiabatically continues back to the zero field $l=n_1$ state.

Thus $(n,3)$ indicates the lowest-energy $(m=0)$ member of the Stark manifold of principal quantum number n and it continues adiabatically to the zero-field nf state. To our knowledge this is the first observation of such sequences of multiphoton transitions.

The details of the atomic system are shown in Fig. 1, a potassium level diagram near $n=16$ in a static field. As shown by Fig. 1, the $n=16$, $l>2$ states are nearly degenerate at zero field, so that in a field of >10 V/cm they are better described as Stark states which have linear Stark shifts and therefore wave functions which are independent of the field. Only the two lowest (16,3) and (16,4) and the upper (16,15) Stark states are shown. The 18s state has virtually no Stark shift and is only weakly coupled to the $n=16$ Stark states, which are composed of $l>2$ states. The weak coupling is evidenced by the small size, 0.9 GHz, of the avoided crossing at 753 V/cm between the 18s and (16,3) levels.[15] The 16d state has a second-order Stark shift due primarily to its dipole coupling to the 16f components of the $n=16$ Stark states. The sequence of transitions we have observed, $18s+qh\nu \rightarrow (16,3)$, for $1 \leq q \leq 28$ is shown schematically in Fig. 1.

Our experimental approach is related to one used previously.[4] A beam of K atoms from an effusive source enters a microwave cavity through a 1.3-mm-diam hole in the cavity sidewall. The atoms are excited stepwise from the ground 4s state through the 4p state to the 19s state by two 5-ns pulsed-dye-laser beams entering the cavity through a 1.3-mm-diam hole in the opposite sidewall. A 0.5-μs pulse of microwave power at a frequency of 9.285, 10.353, or 11.535 GHz is used to drive the transitions. After 1 μs a high-voltage pulse is applied to a septum in the cavity, producing a field which ionizes the excited K atoms and expels these ions through a 1-mm-diam hole in the top of the cavity to a particle multiplier. The signal from the multiplier is captured with a gate set to accept the ionization signal from atoms in the $(16,n_1)$ states, but not the 18s state. The experiment is done by sweeping the static voltage applied to the septum and recording the increase in the $(16,n_1)$ population as the $18s \rightarrow (16,n_1)$ resonances are encountered.

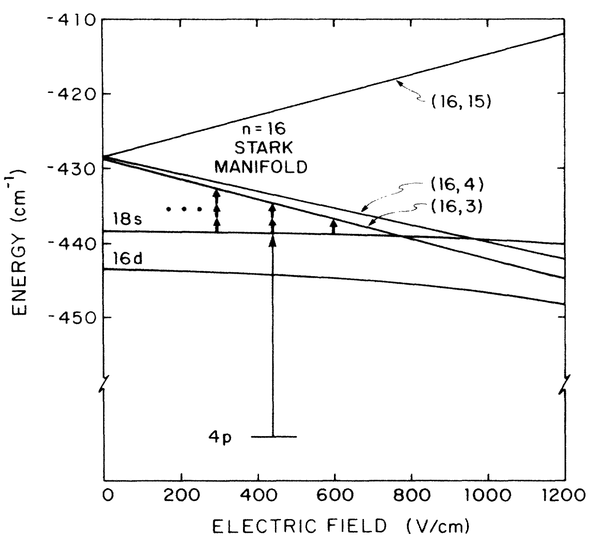

FIG. 1. Relevant energy levels near the $n=16$ Stark manifold. The manifold levels are labeled (n,n_1), where n_1 is the parabolic quantum number. Only the lowest two and highest-energy manifold states are shown. The laser excitation to the 18s state is shown by the long vertical arrow. The $18s \rightarrow (16,3)$ multiphoton rf transitions are represented by the bold arrows. Note that these transitions are evenly spaced in static field, and that transitions requiring more photons occur at progressively lower fields. For clarity, the rf photon energy shown in the figure is approximately 5 times its actual energy.

The microwave cavity is a piece of WR90 waveguide 20.32 cm long, closed at both ends, and containing a copper septum. The cavity has a Q of 2100 at 10.353 GHz and is driven by an Avantek 7872 yttrium iron garnet tuned oscillator, amplified by a Hughes 1277H or an Alfred 623A traveling wave tube amplifier with output powers of 20 and 1.3 W, respectively. With our experimental configuration, 1 W of power produced a field of 228 V/cm. We are able to determine the microwave fields with an uncertainty of $\pm 10\%$, and the static field homogeneity is 1%.

A typical example of a sequence of transitions is shown in Fig. 2 for the $18s \rightarrow (16,3)$ transitions. These are scans of the (16,3) signal as a function of static field for a sequence of values of microwave field. For high static field, few photons are needed and thus low microwave power is required. As we progress to lower static fields, more photons are needed to make the transitions and thus more microwave power is required. The progression of $1,2,\ldots,28$ photons is quite apparent. We also note that there is negligible ac Stark shift until the very highest microwave powers, at which a slight shift to higher static field is observed. For transitions between a state with a linear Stark shift and one with almost no static Stark shift, only small ac Stark shifts are to be expected.[11,12]

Further support of our identification of the transition as $(n+2)s \rightarrow (n,3)$, irrespective of the static field value, is obtained by extrapolating to the zero-field intervals given in Table I. The intervals are consistent with transitions to hydrogenlike states of quantum defect $<5\times10^{-4}$, a requirement for the final states to be Stark states. It is interesting to note that the intervals are >15 GHz too large to match the $(n+2)s$-nf intervals (the nf quantum defect is 0.009[16]).

At a given microwave field, several multiphoton resonances appear with comparable strength. This is approximately what would be expected, based on the simplest Floquet description of the process in which several of the Floquet states most displaced from the original (16,3) level have comparable amplitudes.[11,13] The one- and two-photon transitions are observed with small, 3-V and 20-V/cm, microwave fields, but beyond that point the

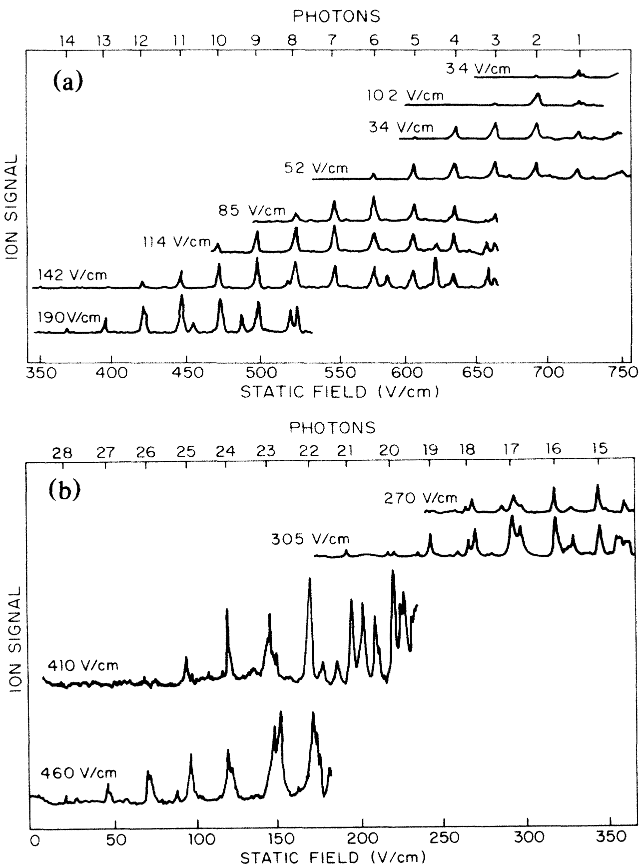

FIG. 2. (a) $18s \rightarrow (16,3)$ 1- to 14-photon transitions observed as the static field is scanned from 350–750 V/cm for the 10.353-GHz microwave fields indicated above each trace (3.4 and 190 V/cm). The regularity of the progression is quite apparent. Note the extra resonances in the 142-V/cm microwave field trace. These are due to $18s \rightarrow (16,4)$ transitions. (b) $18s \rightarrow (16,3)$ 15- to 28-photon transitions observed as the static field is scanned from 0 to 350 V/cm for 10.353-GHz microwave fields from 270 to 460 V/cm. Note the congestion of the 410-V/cm trace at static fields above ~200 V/cm, due to many overlapping $(18s \rightarrow 16,n_1)$ transitions.

maximum number of photons absorbed increases linearly with the microwave field. In the Floquet picture this two-photon offset corresponds to the decreasing tail of sidebands mentioned earlier.

The incremental fields required to absorb one additional photon are given in Table I. They scale as n^{-2}, as does the inverse of the static Stark shift. From these scalings, we can determine the microwave field needed to drive the resonance nearest zero static field (we do not have enough power to see the transitions near zero static field in all cases). These fields, scaling as n^{-5}, are given in Table I, along with the previously measured static-field crossings of the $(n+2)s$ and $(n,3)$ states.[15] As shown by Table I these microwave fields are only 60% of the static fields of the $(n+2)s$-$(n,3)$ avoided crossings. In this case, the $K(n+2)s$-$(n,3)$ interaction is through the nd state, which introduces a nonlinear Stark effect into the problem. Simple Floquet calculations and laser excitation spectra in the presence of microwave fields both indicate that the presence of the nd state slightly below the $nl>2$ states leads to the existence of Floquet or sideband states at energies below the lowest Stark state in a static field of the same magnitude. Experimentally, the $(n+2)s \rightarrow (n,3)$ transition is observed to occur when these Floquet or sideband states reach the s state.

For any given microwave field the transitions cannot be observed below some value of static field, because of the lack of microwave field. Above some value of static field, resonances which are not part of the $18s \rightarrow (16,3)$ series appear. At relatively high static fields and low microwave fields it is easy to identify the beginning of this congestion as the $18s \rightarrow (16,4)$ series of transitions, but at high microwave fields the spectrum becomes effectively continuous above some static field, and an unambiguous assignment is impossible. Nevertheless, the effectively continuous spectrum is due to the overlapping series of transitions to many $(16,n_1)$ Stark states.

The onset of the effectively continuous spectrum cor-

TABLE I. Incremental 10.353-GHz microwave field per photon absorbed, extrapolated zero-field intervals, microwave fields required for the resonances nearest zero static field, and $(n+2)s$-$(n,3)$ static anticrossing fields.

Transition	(Incremental microwave field)/(photons absorbed) (V/cm)	Extrapolated zero-field interval (GHz)	Microwave field of resonance nearest zero static field (V/cm)	Anti-crossing field (V/cm)
$17s$-$(15,3)$	23(2)	⋯	740(80)	1058[a]
$18s$-$(16,3)$	18(2)	295(2)	480(50)	753[b]
$19s$-$(17,3)$	17(2)	246(2)	378(40)	546[b]
$20s$-$(18,3)$	14(2)	226(2)	242(30)	404[b]

[a]Extrapolated from Ref. 16. [b]From Ref. 16.

responds to the threshold for nonresonantly driving the transition. If we set the static field to zero and increase the microwave field, we find a threshold for the $(n+2)s \rightarrow (n,3)$ transition, which in some cases exhibits structure reminiscent of that observed by Mariani *et al.* in He.[5] This observed structure is related simply to the location of the resonances near zero static field and the ac Stark shifts. For example, at 10.353 GHz the $19s \rightarrow (17,3)$ 25-photon resonance shifts through zero static field at a microwave field of ~450 V/cm. Thus in zero static field the $19s \rightarrow (17,3)$ transition occurs for microwave fields between 400 and 500 V/cm and only again for microwave fields > 750 V/cm, where the spectral congestion occurs at a field slightly higher than the $19s \rightarrow (17,3)$ level crossing. At 9.285 GHz, however, there is no such well placed resonance and a microwave field of ~750 V/cm is required.

Although the off-resonance threshold of ~750 V/cm coincides with the $19s$-(17,3) level crossing field and thus with the Landau-Zener prediction, it is apparently due to the overlapping of many transitions. This coincidence stems from the fact that, for microwave fields equal to or greater than the $(n+2)s$-$(n,3)$ crossing field, the Floquet states of many of the (n,n_1) Stark states have appreciable amplitudes near the $(n+2)s$ state, allowing the many overlapping transitions in the presence of adequate coupling. However, there is a more fundamental connection between the multiphoton resonance and Landau-Zener pictures. As was shown by Rubbmark *et al.*[17] for a somewhat more restricted case, when many cycles of the field are used, the Landau-Zener description goes smoothly over to the resonance description. Since the transition amplitude can build up over many coherent microwave cycles, it is hardly surprising that lower microwave fields are required to drive the resonant transitions. We note that in the Na $n \rightarrow n+1$ transitions rapid mixing of the n Stark levels probably removes any possibility of coherence over many microwave cycles, and the single-cycle Landau-Zener description applies. Not surprisingly, no clear resonances were observed in that case and a field equal to ~80% of the crossing field was required to drive the transitions. This is in marked contrast to the case at hand, in which the K s states are not mixed with any other states before making the $(n+2)s \rightarrow (n,3)$ transitions, and clear resonances are observed. In any case, it is clear that an understanding of the levels in a static field allows us to identify the relevant level crossings and thus predict the radiation fields required for multiphoton processes to within a factor of 2. This observation suggests that the field point of view might be employed more generally, to laser multiphoton processes for example, to yield useful insights with minimal calculational effort.

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