Enhancement of Microwave Ionization by Quasicontinuum Production

P. Pillet,[(a)] C. H. Mahon, and T. F. Gallagher

Department of Physics, University of Virginia, Charlottesville, Virginia 22901

(Received 20 October 1987)

Small static fields ($\simeq 1$ V/cm) are observed to reduce the microwave fields required to ionize highly excited $25 \leq n \leq 50$ Li atoms by about an order of magnitude, from $\simeq 100$ to 10 V/cm. This occurs because the static field splits series of multiply degenerate sideband states to form a quasicontinuum of closely spaced states, in which a sequence of resonant absorptions occurs, allowing ionization at the lower microwave field.

PACS numbers: 32.60.+i, 31.50.+w, 32.80.Rm

The presence of a "quasicontinuum" of densely packed discrete states is thought to play a key role in processes in which photons are absorbed in a sequence of real transitions. An example of such a process is multiphoton dissociation in a molecule, which is generally thought to proceed by the selective resonant absorption of a few photons followed by the absorption of a series of photons by transitions through a quasicontinuum of closely spaced excited states.[1] Unfortunately, it is difficult to demonstrate definitively the importance of having a continuous, as opposed to a discrete, energy spectrum in multiphoton dissociation. Here we demonstrate the importance explicitly in a rather similar process. In particular, we describe the striking enhancement of the microwave ionization of Li Rydberg states which occurs when the multiply degenerate, but discrete, microwave sideband states are converted to a quasicontinuum by the application of a small static electric field. Specifically, we have observed that without a static field microwave ionization of Li atoms of principal quantum number $30 < n < 40$ requires fields comparable to those required to ionize H. The dominant process is direct field ionization, requiring a field of $1/9n^4$, 250 V/cm at $n = 40$ (unless indicated otherwise, we shall use atomic units).[2,3] However, when a small static field of $\simeq 1$ V/cm is applied, to produce the quasicontinuum, ionization begins to occur at microwave fields as low as $1/3n^5$, by a series of real transitions through the quasicontinuum, culminating in ionization. Thus, conversion of the discrete levels to a quasicontinuum reduces the microwave field required for ionization by about a factor of $n/3$, roughly an order of magnitude.

To understand how this occurs let us begin with a description of the microwave ionization which occurs for microwave fields $\geq 1/3n^5$. This process was first observed in Na and He and described in terms of Landau-Zener transitions.[4,5] At a static field of $1/3n^5$ the highest-energy $|m| = 0$ Stark state of principal quantum number n intersects the lowest-energy Stark state of principal quantum number $n+1$ as shown in Fig. 1(a). Here m is the azimuthal orbital angular momentum quantum number, and we restrict the present discussion to $m = 0$. For each n there is a manifold of n Stark states, but only the extreme members are shown in Fig. 1(a). The n and $n+1$ states shown do not actually intersect but have an avoided crossing of magnitude ω_0 as shown by the inset. Equivalently, these Stark states have a field-independent coupling matrix element of $\omega_0/2$. If the atoms are exposed to an oscillating microwave field of amplitude $E_{\mathrm{mw}} \geq 1/3n^5$, so that the avoided crossing is reached, the atom has a finite, $> 1\%$, probability of making the Landau-Zener transition $n \rightarrow n+1$ at the avoided crossing if the time to sample the crossing is $\simeq 1/\omega_0$. This in turn leads to the very rough requirement that the microwave frequency $\nu \simeq \omega_0$. If $\omega_0 \ll \nu$, then all the avoided crossings are traversed diabatically, and the Landau-Zener transitions which lead to microwave ionization at a field of $1/3n^5$ do not occur. Since the microwave field of $1/3n^5$ is in excess of the field required to make the $n+1 \rightarrow n+2$ and higher transitions, once the atom reaches the $n+1$ state it easily makes a series of transitions, culminating in ionization. Thus $E = 1/3n^5$ is the field required for the rate-limiting step, the $n \rightarrow n+1$ transition.

Alternatively we can describe the $n \rightarrow n+1$ transition as a resonant multiphoton transition. In the presence of a microwave field of amplitude E_{mw} and frequency ν, the n and $n+1$ Stark states do not each have a single energy, but each breaks into a spectrum of sideband states evenly spaced by ν as shown in Fig. 1(b).[6-9] The extent in frequency of the sidebands with significant amplitudes is the same as the shift produced by a static field of the same magnitude. Thus in a microwave field $> 1/3n^5$, the sidebands of the extreme n and $n+1$ Stark states overlap, as shown in Fig. 1(b). For a resonant $n \rightarrow n+1$ multiphoton transition to occur, the n and $n+1$ sidebands must overlap, and in addition the detuning Δ of sidebands of n from the sidebands of $n+1$ must be less than the coupling matrix element ω_0 between them. For this process to be driven by an arbitrary microwave frequency, for which $\Delta \simeq \nu/2$, we have again the loose requirement that $\nu \simeq \omega_0$ in addition to $E_{\mathrm{mw}} \geq 1/3n^5$. For small ω_0, $\omega_0 \ll \nu$, only very small detunings can be tolerated, producing very sharp resonances, precluding the

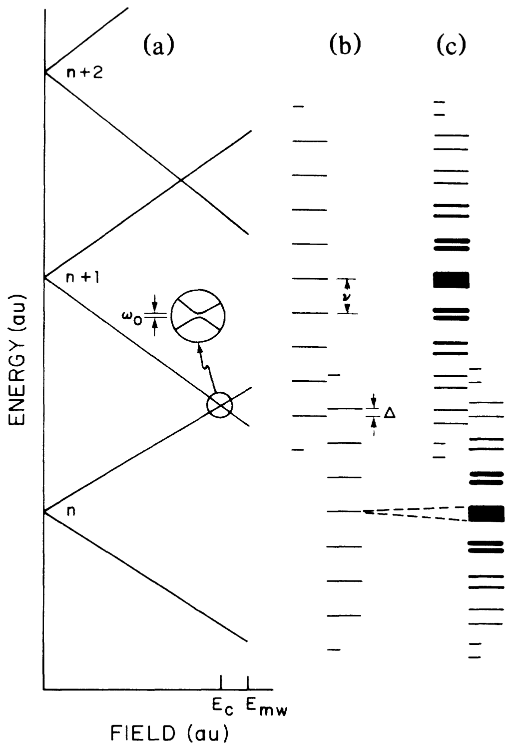

FIG. 1. (a) Static energy-level diagram showing the extreme $|m|=0$ Stark states of the n, $n+1$, and $n+2$ Stark manifolds. The extreme n and $n+1$ levels have an avoided crossing at a field of $E_c=1/3n^5$. As shown by the inset, the magnitude is ω_0. If a microwave field $E_{mw}\gtrsim E_c$ is applied, atoms can make the Landau-Zener transition $n\rightarrow n+1$ at the avoided crossing, followed by successive analogous transitions, culminating in ionization. (b) Spectrum of sideband states produced by a microwave field of amplitude E_{mw} and frequency ν acting on the extreme n and $n+1$ Stark states. The sidebands from all Stark states of the same n are degenerate modulo ν. In general there is a detuning Δ between the n and $n+1$ sideband states. (c) Spectrum of n and $n+1$ sideband states upon the application of a small static electric field which removes the degeneracy of the sideband states originating from different Stark states.

sequence of resonant transitions required to produce microwave ionization.

In Li, at $n=35$ we estimate the values of ω_0 for the $|m|=0$, 1, and 2 states to be 1.2 GHz, 0.26 GHz, and 6 MHz,[10] and the microwave frequency ν in our experiment is 8 or 15 GHz. Thus $\omega_0\ll\nu$, and $n=35$ Li atoms do not ionize at a field of $1/3n^5$ by the sequence of $n\rightarrow n+1$ transitions but rather by direct field ionization at the much higher field of $1/9n^4$.

If the n and $n+1$ states of Fig. 1(b) could be converted to a quasicontinuum, essentially ensuring that $\Delta\simeq\omega_0$, then the $n\rightarrow n+1$ transition and higher lying ones should occur. The conversion to a quasicontinuum of spacing ν/n can be effected in the following way. The extreme high-energy and low-energy n Stark states, shown in Fig. 1(a), have static Stark shifts of kE with $k=3n^2/2$ and $-3n^2/2$, respectively. The intermediate Stark states, which are not shown in Fig. 1(a), have Stark shifts that are evenly spaced between these values, with the states in the center of the Stark manifold having $|k|\ll 3n^2/2$. With no static field, the microwave field of E_{mw} converts each Stark state into a series of sideband states, spaced by ν, which span the energy range $\pm kE_{mw}$ around the zero-field energy. Thus the extreme high- and low-energy n Stark states lead to the degenerate pairs of sidebands shown in Fig. 1(b). A Stark state with a very small Stark shift, $|k|\ll 3n^2/2$, on the other hand, is only converted to a few sideband states, which are degenerate with the central sidebands of the extreme Stark states. Thus the sideband energies nearest the zero-field energy have the highest degeneracy, which falls roughly linearly from n at this point to 2 at the furthest displaced sidebands. In any event, all the sideband states are degenerate modulo ν.

If we apply a static field, E_s, the sidebands from the extreme high- and low-energy Stark states are shifted by $\pm\frac{3}{2}n^2E_s$, respectively, and intermediate Stark states by correspondingly smaller amounts. At the central sideband energy there are n states spread over $\pm\frac{3}{2}n^2E_s$ but at the furthest removed sideband energies there are only two, because of the variation in degeneracy mentioned above. The net result is the energy spectrum shown in Fig. 1(c), which is clearly much more continuous than the spectrum of Fig. 1(b). The static field required to generate the most nearly continuous distribution of these and higher lying states is that which produces a differential shift between the extreme Stark states equal to the microwave frequency. However, since both n and $n+1$ manifolds have approximately the same Stark shifts, only half this field is required to ensure a near degeneracy of n and $n+1$ sideband states, i.e.,

$$E_s=\nu/6n^2 \qquad (1)$$

With this field the central sideband states are quasicontinuous with spacing ν/n. As shown by Fig. 1(c), proportionately larger energy gaps develop as we progress to the most extreme sideband energies which only have two states. In arriving at Eq. (1) we have only considered the avoided crossings between the extreme n and $n+1$ Stark states. At slightly higher microwave fields the crossings between the intermediate Stark states are also encountered. In addition, the extent of the sidebands is proportional to E_{mw}. Thus how far the dense quasicontinuum extends and the number of accessible avoided crossings depend to some extent on the microwave field. If we take these points into consideration, it seems that the factor of 6 in Eq. (1) may not be exactly right, but it is clear that the field should scale as ν/n^2. In hydrogen the analogous n-$(n+1)$ crossings are not merely small, but vanish nonrelativistically, so that it seems that this effect should not be observable in hydrogen, and in fact

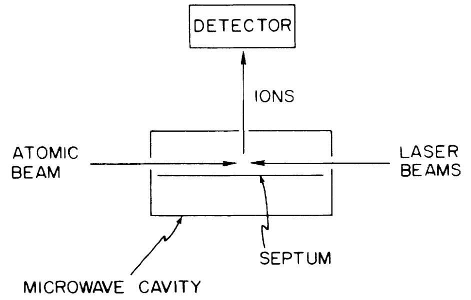

FIG. 2. Cross-sectional view of the microwave cavity in which the atomic beam and laser beams intersect.

it is not.[3]

As our apparatus is very similar to the one described previously,[2] our discussion here is brief. Li atoms from an effusive source pass through a hole in the sidewall of a 15.217- or 7.917-GHz microwave cavity as shown in Fig. 2, where they are excited by two counterpropagating pulsed laser beams entering through a hole in the opposite sidewall. The atoms are excited stepwise via the $2s$-$2p$ transition at 670 nm and the $2p \rightarrow nd$ transition at $\simeq 350$ nm. After a delay of 100 ns, a 500-ns pulse of microwave power is applied to the cavity. The 15- and 8-GHz cavities have Q's of 850 and 1300, respectively. Therefore the fields in the cavity reach 95% of their steady-state values in 18 and 52 ns, respectively. The maximum fields that we can produce with 20 W of microwave power are $\simeq 600$ V/cm. 100 ns after the end of the microwave pulse, a high-voltage pulse is applied to the septum in the cavity to field-ionize atoms remaining in the initially populated Li Rydberg state, or nearby states, and drive these Li^+ ions, as well as those formed by microwave ionization, out the hole in the top of the cavity to the detector. The hole is located at an antinode of the microwave E field. This and the short time between laser excitation and the field-ionization pulse ensures that all detected ions come from atoms which have experienced the same microwave electric field, to within $\pm 7\%$. The Li^+ ions from microwave ionization and those from field ionization have different flight times to the detector, and the corresponding signals can be separately recorded with two gated integrators set at different delay times after the high-voltage pulse. Finally a dc voltage is applied to the septum to produce the small static field.

Our data consist of measurements of microwave-ionization probability as a function of both the static and the microwave fields, and we have collected the data in two ways as a consistency check. The first method is to fix the static field and sweep the microwave field. The second is to fix the microwave field and sweep the static field, typically from -5 to 10 V/cm. As shown explicitly by Pillet *et al.*,[2] we can observe an increase in the probability of microwave ionization as either a reduction of the field-ionization signal from the remaining atoms or an increase in the microwave-ionization signal. When we scan the static field, the microwave-ionization signal moves in time while the field-ionization signal does not, and so we record the field-ionization signal which can be monitored with a gated integrator set at a fixed time. In Fig. 3 we show the field-ionization signal obtained by sweeping the static field when the Li $36d$ state is excited, for several 15.217-GHz field amplitudes. All data shown have been normalized to the $E_{\mathrm{mw}}=0$ signal at the same static field to account for the fact that the static field affects not only the microwave ionization, but also the laser excitation. In more than 95% of the field regime shown in Fig. 3, the $n=36$ states are better described as Stark states. At the highest fields used in Fig. 3, 30% of the $n=36$ Stark states lie outside the 1-cm^{-1} laser bandwidth leading to a corresponding reduction in the number of atoms actually excited. Thus the data of Fig. 3 represent the probability of the atoms not being ionized by the microwaves. The data of Fig. 3 were obtained with both lasers polarized parallel to the static and microwave fields. However, with the lasers polarized perpendicular to the static and microwave fields, negligible difference was observed for this state or others that we studied. We therefore conclude that this is true for $|m|=0$, 1, and 2 states.

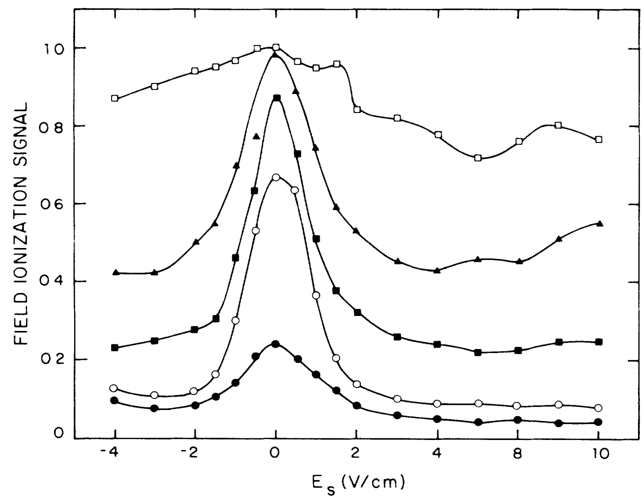

FIG. 3. Field-ionization signals from remaining atoms when the Li $36d$ state is initially excited in a variable static field with the 15.217-GHz microwave field amplitude varied as a parameter. The data are all normalized to the observed signals with $E_{\mathrm{mw}}=0$ at the same static field. $E_{\mathrm{mw}}=31$ (open squares), 44 (triangles), 70 (filled squares), 140 (open circles), and 307 V/cm (filled circles). Note that with $E_{\mathrm{mw}}=44$, 70, and 140 V/cm, there is a rapid decrease in the number of remaining atoms or, equivalently, a rapid increase in the microwave-ionization probability, at $\simeq 1$ V/cm, corresponding to the field required for quasicontinuum production.

As is evident from Fig. 3, with microwave fields of 44, 70, and 140 V/cm, there is a significant decrease in the

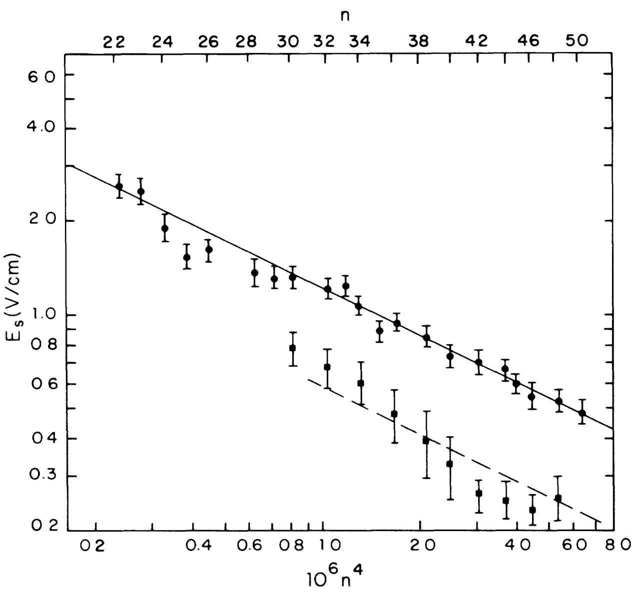

FIG. 4. Experimental points for $E_{1/2}$, the field at the center of the decrease in the signal of Fig. 3, for 15.217 (circles) and 7.921 GHz (squares). The lines are fits by n^{-2} dependences which give $E=0.15\nu/3n^2$ for 15.217 GHz (solid line) and $E=0.14\nu/3n^2$ for 7.921 GHz (dashed line).

number of atoms surviving the microwave pulse as the static field is increased from zero. This is due to the enhancement of the microwave ionization by the conversion of the discrete sideband states to a quasicontinuum. For reference we note that $E=1/3n^5=28$ V/cm, and $E=1/9n^4=340$ V/cm. Data taken by the first method, scanning the microwave power, are consistent with Fig. 3 and show in an explicit fashion the reduced microwave field required for ionization.

As a straightforward way of quantifying the field required for quasicontinuum production, we define the field $E_{1/2}$ as the field at which the signal has fallen halfway from its value at $E_s=0$ to its value at $E_s=6$ V/cm. As E_{mw} is raised from 44 to 70 V/cm, $E_{1/2}$ decreases from $\simeq 1.1$ to 0.95 V/cm, where it remains even when E_{mw} is increased to 140 V/cm. Thus we determine $E_{1/2}$ from these two microwave fields. Although $E_{1/2}$ is convenient, we must remember that the field required to saturate the ionization via the quasicontinuum is roughly a factor of 3 higher than $E_{1/2}$. Using this procedure we have obtained values of $E_{1/2}$ for microwave frequencies of 15.217 and 7.921 GHz, and these data are shown in Fig. 4. From Fig. 4 it is apparent that $E_{1/2}$ is proportional to ν and to $1/n^2$, as required by Eq. (1). Fitting all the experimental points by a single ν/n^2 dependence gives $E=0.30\nu/6n^2$, i.e., a value 3 times lower than that predicted by Eq. (1). This corresponds to the previously mentioned factor-of-3 difference between $E_{1/2}$ and the field at which 90%–100% ionization occurs by quasicontinuum production.

In summary, we feel that our observations are in quite good, probably slightly fortuitous, agreement with the proposed model of quasicontinuum production and, more important, show in an explicit fashion the importance of the quasicontinuum for such sequential processes.

It is a pleasure to acknowledge helpful discussions with D. J. Larson and L. A. Bloomfield. This work has been supported by the U.S. Air Force Office of Scientific Research under Grant No. AFOSR-87-0007.

---

[a] Permanent address: Laboratoire Aimé Cotton, Centre National de la Recherche Scientifique II, Batiment 505, F-91404 Orsay Cédex, France.

[1] D. M. Larsen and N. Bloembergen, Opt. Commun. **17**, 254 (1976).

[2] P. Pillet, H. B. van Linden van den Heuvell, W. W. Smith, R. Kachru, N. H. Tran, and T. F. Gallagher, Phys. Rev. A **30**, 280 (1984).

[3] K. A. H. van Leeuwen, G. V. Oppen, S. Renwick, J. B. Bowlin, P. M. Koch, R. V. Jensen, O. Rath, D. Richards, and J. G. Leopold, Phys. Rev. Lett. **55**, 2231 (1985).

[4] P. Pillet, W. W. Smith, R. Kachru, N. H. Tran, and T. F. Gallagher, Phys. Rev. Lett. **50**, 1042 (1983).

[5] D. R. Mariani, W. van de Water, P. M. Koch, and T. Bergeman, Phys. Rev. Lett. **50**, 1261 (1983).

[6] S. H. Autler and C. H. Townes, Phys. Rev. **100**, 703 (1955).

[7] J. E. Bayfield, L. D. Gardner, Y. Z. Gulkok, and S. D. Sharma, Phys. Rev. A **24**, 138 (1981).

[8] H. B. van Linden van den Heuvell, R. Kachru, N. H. Tran, and T. F. Gallagher, Phys. Rev. Lett. **53**, 1901 (1984).

[9] R. C. Stoneman, D. S. Thomson, and T. F. Gallagher, Phys. Rev. A (to be published).

[10] I. V. Komarov, T. P. Grozdanov, and R. K. Janev, J. Phys. B **13**, L573 (1980).