Microwave Ionization of Na Rydberg Levels

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The authors have observed the ionization of Na Rydberg states by strong microwave fields and have found that the field required for ionization is $1/(3.7n^5)$ (in atomic units) for $n = 20$–37. This field is close to the field $1/(3n^5)$ at which the Stark manifolds of principal quantum numbers n and $n+1$ intersect. The ionization can be shown to arise from transitions occurring near these intersections.

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Electric field ionization of atomic Rydberg states has been intensively studied both for the intrinsic interest of the process and for the potential applications in spectroscopy and collision experiments.[1-4] In spite of the fact that ionization by a pulsed field is a dynamic process, for both static and quasistatic (~ μs rise time) pulsed fields it has been found that the threshold electric field for ionization of a state of principal quantum number n is roughly equal to the classical field required for ionization, $1/16n^4$ (in atomic units), and in all cases scales as n^{-4}.

We have found that microwave-field ionization of sodium atoms is strikingly different from static-electric-field ionization.[1-4] Specifically, the microwave field required for ionization is given by $1/3.7n^5$. At $n=20$ this field is several times smaller than the static field required. Our results are also very different from the only previous systematic measurements[5,6] and calculations[7] of microwave ionization, carried out with hydrogen, in which the same field was required for ionization as in the static-field case, $1/16n^4$. We note that experiments with several He states have been performed recently which give results similar to those reported here for Na.[8] Here we describe our experiments and present a physical interpretation of the microwave ionization as a consequence of the rapid temporal variation of a classical microwave field, not as multiphoton ionization.

The experimental approach has been described elsewhere,[4] the only novelty being the utilization of a high-Q (~1400) cavity to apply the microwave field to the Na atoms. The cavity, shown in Fig. 1, is made of a section of X-band waveguide 20.32 cm long, closed at each end, and having usable, TE_{10n}, where n is odd, resonances ~3 GHz apart in the 7–16 GHz range. An insulated septum in the middle of the cavity (see Fig. 1) is used to produce inside the cavity a static (or quasistatic) electric field.

The Na atomic beam goes through the cavity as shown in Fig. 1 and is excited from the $3s_{1/2}$ ground state to the $3p_{3/2}$ state and then to a high s or d level by two successive pulsed tunable dye-laser beams. Then, 200 ns after the laser pulses, we apply a 500-ns square-shaped microwave pulse. Finally, a weak (30 V) positive pulse is applied to the septum, which accelerates the ions formed out of the cavity through a 1-mm-diam hole to a particle multiplier. The resulting signal is then measured with a boxcar averager.

We define the threshold amplitude for ionization, F_T, as the field required to produce an ion current equal to 50% of the maximum microwave ionization signal observed. Figure 2 shows the variation of the ion current versus the microwave field for the 24s and 23d states. As shown by Fig. 2, an increase from 10% to 90% of the max-

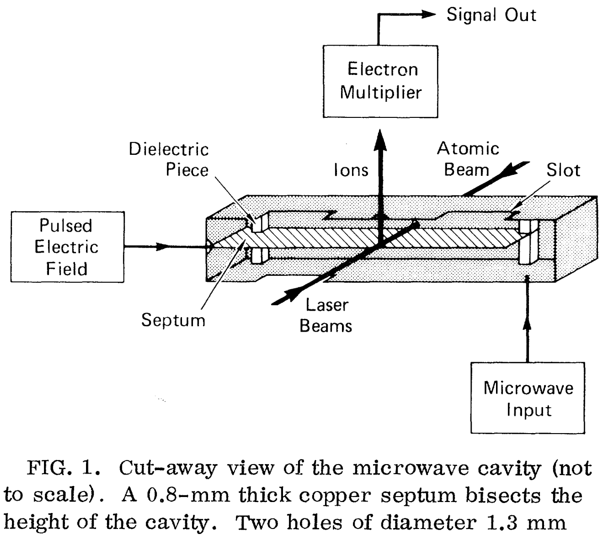

FIG. 1. Cut-away view of the microwave cavity (not to scale). A 0.8-mm thick copper septum bisects the height of the cavity. Two holes of diameter 1.3 mm are drilled in the side walls to admit the collinear laser and Na atom beams and a 1-mm hole in the top of the cavity is used for extraction of the ions. Input power of 14 W produces a field amplitude of 440 V/cm in the center of the cavity.

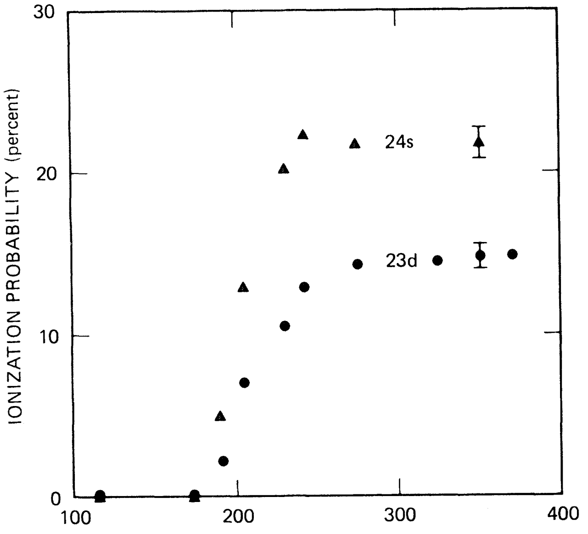

FIG. 2. Experimental ionization probability vs the microwave electric field for the $23d$ level (circles) and the $24s$ level (triangles).

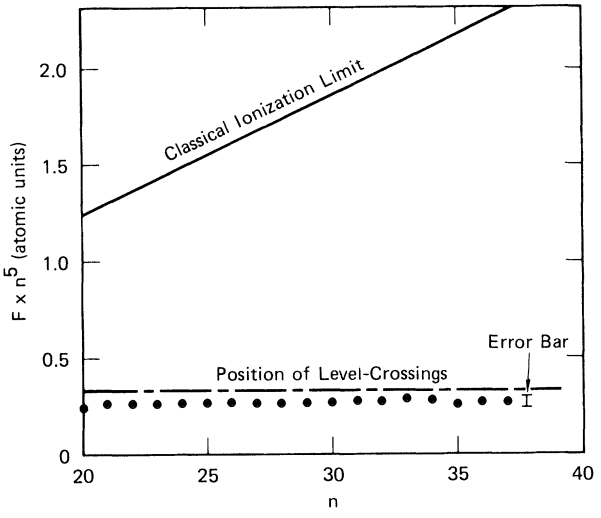

FIG. 3. Ionization threshold fields of nd levels multiplied by n^5, obtained with both lasers polarized parallel to the microwave field (circles).

imum ionization of the $24s$ state requires an increase of 20% in the microwave field, much more than the 2% increase required in the case of a quasistatic field.[4] Unlike the 100% ionization efficiency of the static field, apparently 20% of the atoms are ionized in the case of the s state and about 15% in the case of the d state. Why 100% ionization is not observed is not completely understood. Indirect evidence suggests that they go into high m states, which require much higher fields for ionization.[3,9] However, we note that such states are in many cases experimentally indistinguishable from lower n states.

The threshold fields for ionization have been measured for the ns states ($21 \leq n \leq 28$) and nd states ($20 \leq n \leq 37$) at a microwave frequency of 15.42 GHz. In Fig. 3 we plot the observed threshold fields of the nd states multiplied by n^5 (circles), as well as the classical field required for ionization multiplied by n^5, i.e., $n/16$ (solid line). As shown by Fig. 3 these ionization threshold fields are substantially different from the classical ionization field. The data of Fig. 3 may be expressed as

$$F_T = \frac{1.38\times10^9}{n^5}\ \text{V/cm} \simeq \frac{1}{3.7n^5}\ (\text{atomic units}),$$

which, to our knowledge, is the first observation of an n^{-5} scaling for an ionization threshold field. We estimate that the uncertainties in measuring losses and reflections of the microwave power lead to uncertainties of 10% in the absolute values of these threshold fields.

Several additional observations provide further insights. The threshold fields obtained at 12.79 GHz are not significantly different, and the observed thresholds show little dependence on the laser polarization. The $(n+1)s$ and $(n+1)p$ levels have the same ionization threshold as nd, a result totally different from quasistatic-electric-field ionization in which $(n+1)s$, nd, and np states have the same threshold field but $(n+1)p$ has a much lower threshold field.[4] Finally, we have observed that adding a small dc field reduces the microwave field required for ionization. More precisely the sum of the dc field and microwave field amplitude required for the ionization of a given state is a constant, i.e., the maximum value of field occurring during each microwave cycle is the important parameter. This suggests the utility of treating the microwave as a field and not as photons.

These observations imply that the ionization is the result of the atoms interacting with a classical, time-dependent field. We first note that the ionization threshold field F_T is ~20% less than $1/3n^5$, the field F_x at which the Stark manifolds of principal quantum numbers n and $(n+1)$ intersect.[10] This suggests that the ionization process involves transitions between states with quantum number n and $(n+1)$ near the level crossing at the intersection of the Stark manifolds [we also show in Fig. 3, $F_x n^5 = \frac{1}{3}$ (dashed line)].

With this idea in mind, let us consider in detail how atoms are ionized by the microwave pulse. Figure 4 shows a simplified Stark diagram for the levels $n=20$ to 30. Assume that the atoms are excited to the 20*d* state before the application of a microwave field of steady-state amplitude 430 V/cm. As the microwaves fill the cavity they initially mix the states of the $n=20$ manifold. This occurs even at very low microwave fields for the crossing at zero field is necessarily traversed at a high slew rate. (Related effects due to quasistatic field reversals have been observed by MacAdam, Rolfes, and Crosby[11] and Higgs *et al.*[12]) We note that the 21*s* states and 21*p* are mixed into the $n=20$ manifold when the microwave field amplitude exceeds 380 and 200 V/cm, the fields at which these states intersect the $n=20$ manifold. Thus the *nd*, $(n+1)p$, and $(n+1)s$ states have the same threshold for ionization.

When the microwaves have reached their steady-state amplitude of 430 V/cm, those atoms in the highest energy $n=20$ Stark state are brought almost to the level crossing with the lowest energy $n=21$ Stark state, point *A* of Fig. 4. As the microwave field passes through its peak value some of these atoms make transitions to the lowest-energy $(n+1)$ Stark level as shown by the inset of Fig. 4. Since the intersections of higher-*n* Stark manifolds occur at lower fields, transitions to higher and higher *n* states are readily made on successive microwave cycles, so that the atoms are rapidly distributed over the Stark manifolds from $n=20$ up to $n\sim30$ where field ionization occurs.

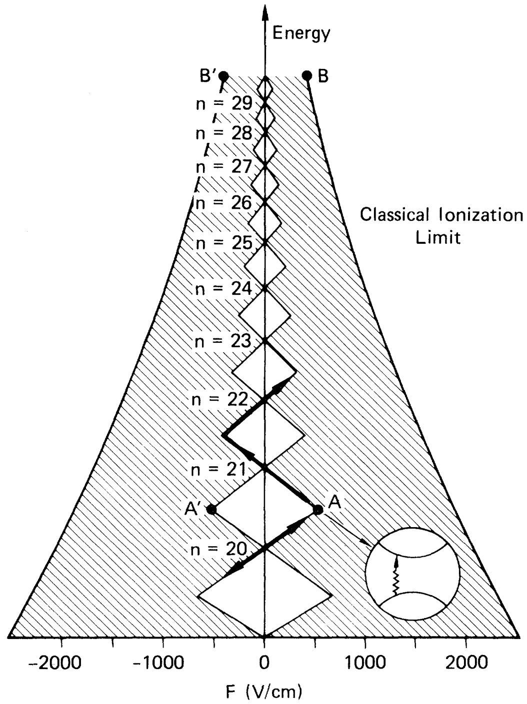

FIG. 4. Simplified Stark-level diagram for sodium. The bold arrows indicate the first steps leading ultimately to ionization at the classical ionization limit (point *B* or *B'*). Note that the bold arrows stop slightly short of *A* where the transition shown by the inset occurs.

The ionization process is apparently the result of a multistep process, the rate-limiting step of which is the transition from the *n* to $(n+1)$ Stark manifold. Thus the observed microwave ionization thresholds should be the fields at which the transition probability from the highest-energy *n* Stark state to the lowest-energy $n+1$ Stark state becomes appreciable.

The transition probability is easily calculated with the Landau-Zener approach of Rubbmark *et al.*[13] We have considered the lowest-energy level of the $n+1$ Stark manifold and the highest level of the *n* Stark manifold. With no coupling they would cross at F_x, however, they are coupled by the nonzero quantum defects, and thus there is an avoided crossing at F_x as shown by the inset of Fig. 4. Extensive numerical calculations indicate that at 15 GHz even when the microwave field amplitude is only $0.9F_x$ the transition probability is ~1% after one microwave cycle. As the Rydberg atoms are in the microwave field for several thousand cycles, it is evident that the transition occurs even though the field does not reach the first level crossing. This is predicted by the Landau-Zener theory[14] but has not to our knowledge been unambiguously observed previously.

As expected, the calculated transition probability is roughly proportional to the size of the coupling, or equivalently to the size of the avoided crossing. Thus in hydrogen, where there is negligible coupling between the *n* and $n+1$ Stark manifolds, ionization should not occur by this mechanism at a field of $\sim1/3n^5$, and in fact microwave ionization occurs only at the higher classical field required for ionization $1/16n^4$.[5] Finally we note that our calculations indicate that in the Na states of $n\sim20$ for frequencies below 2 GHz the transition probability drops significantly. Thus for frequencies less than 2 GHz we expect the field required for ionization to be more nearly given by $1/16n^4$.

To summarize, we have observed for the first time a microwave ionization threshold law for an alkali atom, $F_T \simeq 1/3.7n^5$, giving a much lower threshold field than is found for quasistatic fields. The $1/n^5$ ionization threshold field dependence, the additivity of the applied microwave field and a small dc electric field, and the weak frequency dependence suggest that the observed ionization is due to a rapidly varying classical field.

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[1] S. Liberman and J. Pinard, Phys. Rev. A 20, 507 (1979).

[2] M. Littman, M. Kash, and D. Kleppner, Phys. Rev. Lett. 41, 103 (1978).

[3] T. Jeys, G. Foltz, K. Smith, E. J. Beiting, F. Kellert, F. B. Dunning, and R. F. Stebbings, Phys. Rev. Lett. 44, 390 (1980).

[4] T. F. Gallagher, L. M. Humphrey, W. E. Cooke, R. M. Hill, and S. A. Edelstein, Phys. Rev. A 16, 1098 (1977).

[5] J. E. Bayfield and P. M. Koch, Phys. Rev. Lett. 33, 258 (1974).

[6] J. E. Bayfield, L. D. Gardner, and P. M. Koch, Phys. Rev. Lett. 39, 76 (1977).

[7] J. G. Leopold and I. C. Percival, Phys. Rev. Lett. 41, 944 (1978).

[8] P. M. Koch, J. Phys. (Paris), Colloq. 43, C2-187 (1982).

[9] J. H. M. Neijzen and A. Dönszelmann, J. Phys. B 15, L87 (1982).

[10] M. L. Zimmerman, M. G. Littman, M. M. Kash, and D. Kleppner, Phys. Rev. A 20, 2251 (1979).

[11] K. B. MacAdam, R. Rolfes, and D. A. Crosby, Phys. Rev. A 24, 1286 (1981).

[12] C. Higgs, M. A. Finemann, F. B. Dunning, and R. F. Stebbings, J. Phys. B 15, L697 (1982).

[13] J. R. Rubbmark, M. M. Kash, M. G. Littman, and D. Kleppner, Phys. Rev. A 23, 3107 (1981), and references therein.

[14] L. D. Landau and E. M. Lifshitz, *Quantum Mechanics, Non-Relativistic Theory* (Pergamon, New York, 1965), pp. 171–174 and pp. 322–330.