

Tunneling Rates for Excited States of Sodium in a Static Electric Field*

Michael G. Littman, Myron L. Zimmerman, and Daniel Kleppner

Research Laboratory of Electronics and Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

(Received 28 June 1976)

We have measured ionization rates for selected Stark states of sodium in the range $n = 12-14$, $|m| = 2$. We find that the ionization rates for the lowest Stark components of each term are in excellent agreement with values calculated for hydrogen. The systematics of ionization of the higher sublevels, however, is in serious disagreement with hydrogenic theory because of effects of level mixing.

Ionization of hydrogen in a static electric field by electron tunneling has evoked continuing interest since the early days of quantum mechanics,¹⁻³ and has assumed new importance with the growing interest in highly excited atoms. Many useful experimental techniques based upon the ionization characteristics of these atoms have been developed.^{3,4} A detailed understanding of the ionization process is desirable to assure the validity of tunneling calculations, and is essential in view of the scope of experimental work whose interpretation rests on it. We present here first measurements of the ionization rates of individual Stark sublevels of sodium for $n = 12, 13, 14$; $|m| = 2$. The Stark structure of these states is well described by hydrogenic theory except in small regions near level anticrossings. We find that the ionization rates for the low-lying Stark sublevels of several terms are in excellent agreement with the rates that have been calculated for hydrogen. For the higher-lying sublevels, however, there is serious disagreement. The discrepancy is due to level mixing with ionizing levels which apparently has been neglected in tunneling calculations.

The early history of tunneling in excited states of hydrogen has been reviewed by Bethe and Salpeter.¹ A number of different theoretical approaches have been applied to this problem.² In particular, Bailey, Hiskes, and Riviere (BHR)⁵ have applied a WKB method developed by Rice and Good⁶ to calculate ionization rates for selected Stark levels up to $n = 25$. They found that the rates increase monotonically and rapidly with field. For the lowest component of the $n = 15$, $|m| = 0$ manifold, for instance, the ionization rate increases from 10^6 to 10^{11} sec^{-1} as the field increases from 10 to 12.5 kV/cm . The ionization-rate curves for adjacent sublevels are nearly parallel and are displaced toward higher field with increasing Stark energy. Several groups have used these calculations to interpret field-ionization experiments on fast beams of excited

hydrogen formed by charge exchange.^{3,7} Unfortunately, such experiments do not permit detailed study of the ionization rates since contributions to the ionization signal from numerous states overlap.

We have succeeded in measuring ionization rates for individual Stark states of sodium in a thermal atomic beam. A description of our method for accessing and identifying selected Stark sublevels has been presented previously.⁸ Ionization rates were determined by measuring the number of ions created as a function of time after excitation of the atoms in a static electric field by a 5-nsec laser pulse. The polarization axis of the laser, which excited transitions from the

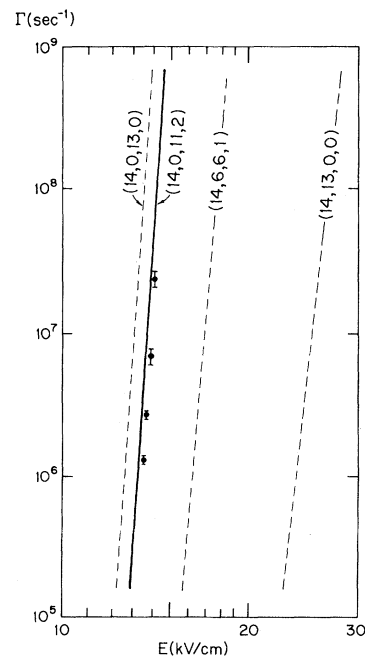


FIG. 1. Measured ionization rates versus field for the state ($n = 14, n_1 = 0, n_2 = 11, |m| = 2$). The theoretical curve (solid) was obtained by extrapolation from the ionization rates calculated by BHR (Ref. 5) for nearby Stark levels (dashed).

$3^2P_{3/2}$ intermediate state, was perpendicular to the electric field, yielding final states with $|m| = 0, 1, \text{ and } 2$.

Decay from the excited state was monitored by a single-event timing technique in which the distribution of arrival times of the first ion after laser excitation was measured with a time-to-digital converter. The decay of the excited state was observed to be exponential within statistical error, 2% or less. (Systematic timing errors are negligible at this level.) The method was used to measure ionization rates in the range of 10^6 to 10^8 sec^{-1} . Rates below 10^6 sec^{-1} could not accurately be determined because of time of flight of the beam ($\leq 2 \times 10^5 \text{ sec}^{-1}$) and because of corrections for radiative decay and collisions ($\leq 3 \times 10^5 \text{ sec}^{-1}$). The upper limit was set by timing resolution. (Rates in excess of 10^{11} sec^{-1} have been determined from broadening of the excitation curve—a decay rate of $5 \times 10^{10} \text{ sec}^{-1}$ corresponds to a frequency width equal to the laser linewidth, 0.5 cm^{-1} —but the results presented here were obtained entirely by direct timing.)

Data for the lowest-lying state of the $n = 14$,

$|m| = 2$ manifold, $(14, 0, 11, 2)$, are shown in Fig. 1. [The level indices give the parabolic quantum numbers $(n, n_1, n_2, |m|)$.] The rate calculated by BHR is shown for comparison. The theoretical curve is extrapolated from BHR's values for the extreme $|m| = 0$ levels⁵ and the central $|m| = 1$ level³ of the $n = 14$ manifold. The slopes of the theoretical and experimental curves agree, and to within the 3% uncertainty in calibration of the field the curves coincide.

Our observations of ionization rates for higher-lying components of the $|m| = 2$ Stark manifold are, however, in serious disagreement with BHR's calculations. The difficulty can be seen by examining the experimental excitation curves in Fig. 2(b) for the levels which are identified in Fig. 2(a). The signals were generated by ions collected following a 200-nsec delay after excitation. Disappearance of a signal with increasing field indicates that the lifetime of the level has fallen below 100 nsec, that is, its ionization rate exceeds 10^7 sec^{-1} . In regions where the field-ionization rate is low, the signal is due primarily to a small amount of collisional ionization.

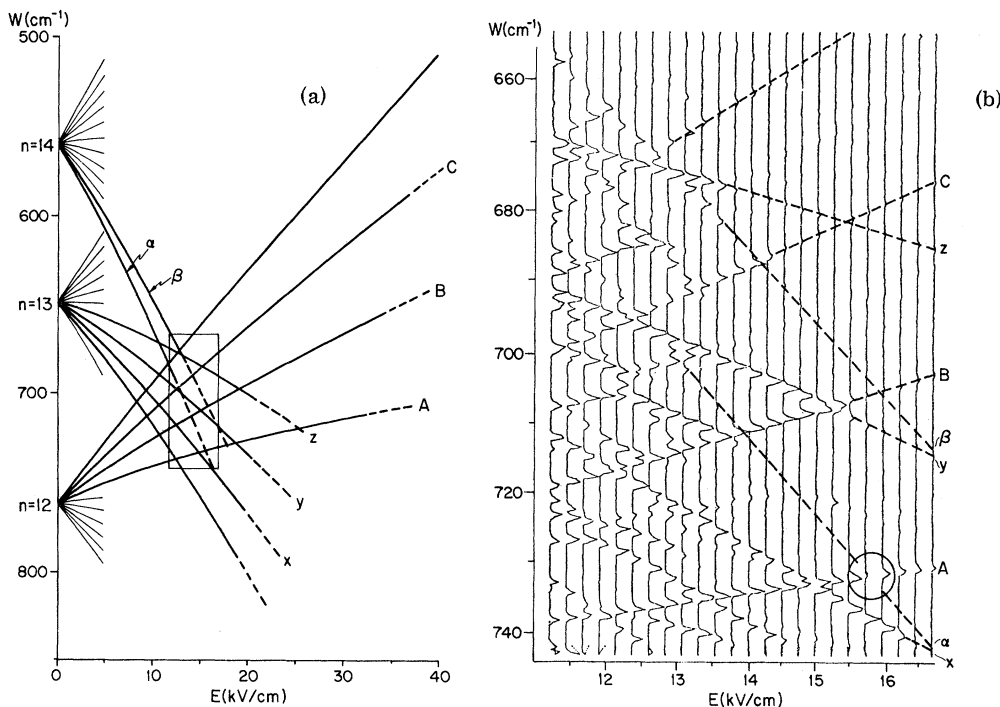


FIG. 2. (a) Stark structure of states $n = 12, 13, 14$; $|m| = 2$. The dashed lines indicate that the ionization rates exceed 10^7 sec^{-1} according to BHR. The quantum numbers for the states are A, $(12, 6, 3, 2)$; B, $(12, 7, 2, 2)$; C, $(12, 8, 1, 2)$; x, $(13, 2, 8, 2)$; y, $(13, 3, 7, 2)$; z, $(13, 4, 6, 2)$; α , $(14, 0, 11, 2)$; β , $(14, 1, 10, 2)$. (b) Excitation curves in sodium obtained by scanning the laser over the region indicated in (a). The signals, which appear as horizontal peaks, were generated by ions collected following a 200-nsec delay after excitation. Disappearance of a signal with increasing field indicates that the lifetime of the level has fallen below 100 nsec (i.e., ionization rate is in excess of 10^7 sec^{-1}). The dashed lines commence at the observed disappearance of each $|m| = 2$ level. (The other unlabelled peaks have $|m| = 0$ or 1.) The circled feature is explored in greater detail in Fig. 3.

The dashed lines in Fig. 2(b) commence at the observed disappearance of each $|m| = 2$ level. For comparison, the field at which the ionization rate exceeds 10^7 sec^{-1} , extrapolated from BHR's calculations, is indicated by dashed lines in Fig. 2(a). It is apparent that many levels begin to ionize at substantially lower fields than expected.

A clue to this early onset of ionization is provided by the circled feature in Fig. 2(b). The level $A [(12, 6, 3, 2)]$ momentarily disappears at

$$W - \frac{1}{2} i \Gamma = \frac{1}{2} (W_A + W_\alpha - \frac{1}{2} i (\Gamma_A + \Gamma_\alpha)) \pm \{ [W_A - W_\alpha - \frac{1}{2} i (\Gamma_A - \Gamma_\alpha)]^2 - 4 |V_{A\alpha}|^2 \}^{1/2}.$$

W_A and W_α are the unperturbed energies for the levels, Γ_A and Γ_α are their decay rates, and $V_{A\alpha}$ is the matrix element of $\vec{E} \cdot \vec{r}$ which couples the two states. We have computed $V_{A\alpha}$ by diagonalizing the energy matrix for $|m| = 2$ with n ranging from 10 to 15, using hydrogenic dipole matrix elements corrected for small effects of core polarizability and penetration.⁸ Γ_α , which varies rapidly with E , was determined from the results of BHR. At the A - α crossing, $\Gamma_\alpha \approx 10^{10} \text{ sec}^{-1}$. (Γ_A , due primarily to radiation, was experimentally determined to be $3 \times 10^5 \text{ sec}^{-1}$. Since $\Gamma_A \ll \Gamma_\alpha$ its value does not need to be known precise-

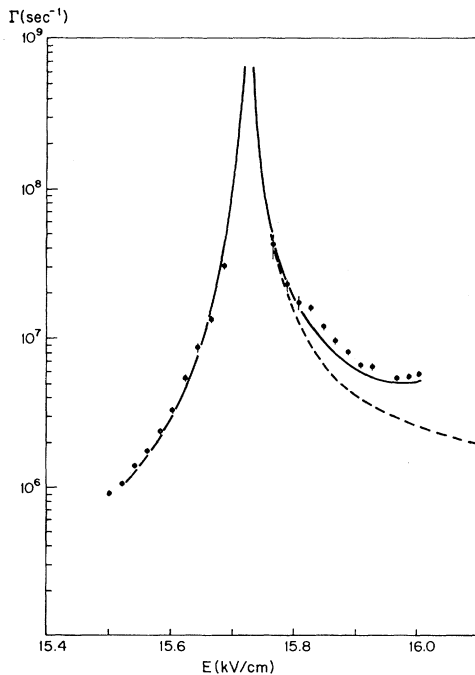


FIG. 3. Ionization rates for the level $A [(12, 6, 3, 2)]$ in the region of the crossing with the rapidly ionizing level $\alpha [(14, 0, 11, 2)]$. The solid line is the theoretical curve described in the text. The dashed line is calculated neglecting the A - θ crossing at 17.3 kV/cm.

a field of 15.75 kV/cm. We ascribe this behavior to level mixing with the rapidly ionizing state $\alpha [(14, 0, 11, 2)]$ at the A - α level crossing. Quenching phenomena of this type, which can lead to violation of the "no crossing" theorem, have been discussed by Lamb.⁹ Figure 3 shows data for the ionization rate of state A in the vicinity of this crossing. The theoretical curve in Fig. 3 is calculated using the following expression for the complex energy of two interacting levels¹⁰:

ly.) A second ionizing level, $\theta [(15, 0, 12, 2)]$ [not shown in Fig. 2(a)], crosses level A at 17.3 kV/cm and was included in the calculation, assuming that the A - α and A - θ crossings are independent.

The sharp increase in the ionization rate at a level crossing offers insight into the disappearance of the other levels shown in Fig. 2(b). If crossings occur with a series of broad levels, and if the coupling matrix elements are large, then the ionization rate never returns to the value for the isolated level. The level is effectively coupled into the continuum in a manner similar to that of an autoionizing state.

The dramatic effects of level mixing in field ionization of sodium naturally raise questions about the role of level mixing in hydrogen. Level crossings in hydrogen, which appear to arise chiefly from relativistic effects, are much sharper than in sodium. Nevertheless, even a minute admixture of a high-lying state with a large ionization rate can dominate the ionization process. The problem needs further study.

The question also arises as to whether our agreement for the ionization rate of the $(14, 0, 11, 2)$ level of sodium with BHR's results for hydrogen may be simply fortuitous. To investigate the possible role of level mixing, we projected the sodium wave function onto a hydrogenic basis set for fields in the range 13–15 kV/cm. The major perturbations to the ionization rate, due to $n = 15, 16$ manifolds, were found to be negligible. Thus, we believe that the $(14, 0, 11, 2)$ state in sodium is indeed suitable for testing hydrogenic theory.

We conclude that in the absence of level mixing our results are in good agreement with the rates calculated by BHR, but that when level mixing is important both quantitative and qualitative features of ionization can be significantly altered.

We would like to acknowledge helpful conver-

sations on level crossing in hydrogen with David R. Herrick and with Gregory Hatton.

*This work was supported by U. S. Air Force Office of Scientific Research Contract No. F44620-72-C-0057 and by Joint Services Electronics Program Contract No. DAAB07-75-C-1346.

¹H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms* (Academic, New York, 1957).

²Recent theoretical work on field ionization in hydrogen: M. H. Rice and R. H. Good, Jr., *J. Opt. Soc. Am.* **52**, 239 (1962); D. S. Bailey, J. R. Hiskes, and A. C. Riviere, *Nucl. Fusion* **5**, 41 (1965); B. M. Smirnov and M. I. Chibisov, *Zh. Eksp. Teor. Fiz.* **49**, 841 (1965) [*Sov. Phys. JETP* **22**, 585 (1966)]; J. O. Hirschfelder and L. A. Curtiss, *J. Chem. Phys.* **55**, 1395 (1971); N. A. Guschina and V. K. Nikulin, *Chem. Phys.* **10**, 23 (1975); L. B. Mendelsohn, *Phys. Rev.* **176**, 90 (1968); M. Alexander, *Phys. Rev.* **178**, 34 (1969); J. Fauchier and J. D. Dow, *Phys. Rev. A* **9**, 98 (1974); M. Hehenberger, H. V. McIntosh, and E. Brändas, *Phys. Rev.*

A **10**, 1494 (1974); D. R. Herrick, to be published.

³Review articles on field ionization experiments in fast beams: A. C. Riviere, in *Methods of Nuclear Physics*, edited by B. Bederson and W. L. Fite (Academic, New York, 1968), p. 208; R. N. Il'in, in *Atomic Physics 3*, edited by S. J. Smith and G. K. Walters (Plenum, New York, 1973), p. 309.

⁴For example, T. W. Ducas, M. G. Littman, R. R. Freeman, and D. Kleppner, *Phys. Rev. Lett.* **35**, 366 (1975); R. F. Stebbings, C. J. Latimer, W. P. West, F. B. Dunning, and T. B. Cook, *Phys. Rev. A* **12**, 1453 (1975).

⁵Bailey, Hiskes, and Riviere, Ref. 2.

⁶Rice and Good, Ref. 2.

⁷J. E. Bayfield and P. M. Koch, *Phys. Rev. Lett.* **33**, 258 (1974); P. M. Koch, thesis, Yale University, 1974 (unpublished).

⁸M. G. Littman, M. L. Zimmerman, T. W. Ducas, R. R. Freeman, and D. Kleppner, *Phys. Rev. Lett.* **36**, 78 (1976).

⁹W. E. Lamb, *Phys. Rev.* **85**, 259 (1952).

¹⁰D. Kleppner, in *Atomic Physics and Astrophysics*, edited by M. Chrétien and E. Lipworth (Gordon and Breach, New York, 1971).

Observation and Simulation of Effects on Parylene Disks Irradiated at High Intensities with a 1.06- μm Laser

W. C. Mead, R. A. Haas, W. L. Kruer, D. W. Phillion, H. N. Kornblum, J. D. Lindl, D. R. MacQuigg, and V. C. Rupert

Lawrence Livermore Laboratory, University of California, Livermore, California 94550

(Received 3 May 1976)

Parylene (C_8H_8) disks have been irradiated with a 1.06- μm laser at fluxes of 10^{15} – 10^{17} W/cm^2 . The spatial and temporal scattered light distributions, x-ray spatial and spectral emission properties, and ion spatial and energy distributions were measured. The results, together with two-dimensional magnetohydrodynamic code simulations, imply absorption via collective processes, laser generation of suprathreshold electrons, and transport inhibition consistent with the presence of megagauss-level thermoelectric magnetic fields.

Recent experiments using Nd-glass lasers to heat DT filled glass microspheres suggest that light absorption, plasma heating, and electron conduction play determining roles in achieving compression and thermonuclear neutron production.¹ In order to investigate these processes in simpler systems, many workers have employed plane target geometry.²

In the present work,³ a series of irradiations of parylene- N^4 disks were performed at the JANUS laser facility.⁵ The 150- μm -diam by 10- μm -thick C_8H_8 disks were mounted on glass stalks, placed at varying positions with respect to the system's best focus, and irradiated with energies of 6–12

J, and durations of 60–120 psec. Nominal target-spot diameters of 90, 30, and 10 μm were used to cover the intensity range of 10^{15} – 10^{17} W/cm^2 . An extensive diagnostic package observed plasma behavior on each shot, and detailed comparisons have been made between observations and two-dimensional computer simulations.

The threshold for target damage by prepulse energy was established by using only the oscillator and preamplifiers to apply low-energy pulses to the target. Detailed target inspection showed no observable damage at energy densities below 4.5 J/cm^2 . The laser prepulse diagnostic detection limit (2–5 μJ) was sufficiently low to assure no