fact that the peak of the experimental spectrum (Fig. 1) falls midway between the single- and double-transition frequencies, and from previous results on the quadrupole-induced lines,[7] we expect the single and double transitions to contribute roughly equal amounts to the spectrum of Fig. 1. Assuming exactly equal contributions would give $|\Phi| = 0.42ea_0^4$ and $|\Phi'| = 0.33ea_0^3$.

Finally, we mention other possible induction mechanisms, in addition to the hexadecapolar mechanism. The quadrupole-induced double transitions $S_0(1)+S_1(3)$, $S_1(1)+S_0(3)$, and $S_0(2)+S_1(2)$, which fall within the linewidth of Fig. 1, can be ruled out. To do this the group of double-S transitions near 5300 cm^{-1} (outside Fig. 1) were observed[8] and their intensity compared with the expected intensity of the double-S transitions in question; the latter is expected to be about 5% of the intensity of the lines near 5300 cm^{-1}. Since the observed peak of Fig. 1 is about 50% of the intensity of the 5300 cm^{-1} lines, we see that the double-S transition mechanism fails, by an order of magnitude, to explain the observed peak. Rough estimates of the dispersion and overlap mechanisms indicate that they too are small. The "level-mixing" mechanism of Herman[9] is estimated to be of importance here only if Φ is of order $0.1ea_0^4$ or less.

Work is now in progress to obtain accurate absorption coefficient for the pure rotational lines of H_2, and the lines of H_2–rare-gas mixtures. An analysis of all these experiments together should yield more accurate values for both $|\Phi|$ and $|\Phi'|$.

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Multiphoton Ionization of Highly Excited Hydrogen Atoms*

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A new method for the experimental study of multiphoton processes uses production of highly excited atoms in keV electron-transfer collisions. Such atoms can be ionized over a wide range of frequencies of an external electromagnetic field, permitting studies of multiphoton-ionization processes for regimes not yet achieved in laser experiments. Results for microwave ionization of hydrogen atoms with principal quantum numbers in the band $63 \leq n \leq 69$ are discussed in terms of the available theory.

The multiphoton ionization of atoms by intense oscillating electromagnetic fields is not well understood at present. Theoretical interpretation of laser-ionization experiments has treated the electric field $F\cos(\omega t)$ as a perturbing influence on the electronic motion within the atom, under conditions where the field oscillates rapidly with a period T short compared to the time of flight τ of the electron to a distance r_0 where the external field F would dominate over the attractive force F_0 of the atomic nucleus. However, when pulsed lasers of greater power become available for experiments on ground-state atoms, new regimes will be reached where either or both of the conditions $F \ll F_0$ and $T \ll \tau$ becomes inapplicable. It is the purpose of this paper to demonstrate that experiments within these new regimes can be done now by lowering the frequency into the microwave region to give larger T while reducing the field strengths required by using highly excited atomic electronic states with smaller F_0. For a simple one-electron system, the hydrogen atom, we present ionization data for $T \ll \tau$, $T \sim \tau$, and $T \gg \tau$. Our experiments constitute the first observations of the microwave ionization of highly excited atoms.

For a fixed frequency ω several possible breakdowns of perturbation theory (PT) can occur as F is increased. One theoretically considered situation occurs when the decrease of r_0 with increasing F makes $T \gtrsim \tau$, while $F \ll F_0$ still holds.[1] The ionization is then viewed analogously to the static-field case as a quantum mechanical tunneling of the electron through a time-varying barrier produced by the superposition of the external and Coulomb forces on the electron. Since the ionization rate should be a maximum when the field is near its peak value F, T can be taken as the ratio r_1/v of the minimum barrier width for an electronic state of binding energy $I = \frac{1}{2}\kappa^2$ to the electronic velocity v. Using atomic units[2] and considering only a hydrogen atom, for which the electron momentum $\kappa = n^{-1}$, we have for $F \ll \frac{1}{16}n^{-4}$ (the approximate classical ionization-threshold field), approximately, $v = \kappa$, $r_1 = \kappa^2 2F$, and hence $\tau = \kappa/2F$. In the PT limit the transition rate is given by a sum of energetically allowed terms involving increasing numbers of photons in a power series in a quantity proportional to field intensity. The breakdown of PT occurs when one such quantity useful for excited states, $1/4\gamma^2$, increases to order unity, where $\gamma \equiv 2\omega\tau = \omega/nF$ is called the adiabatic tunneling parameter[1,3] (see Ref. 3, Eq. 22). An untested theory for the adiabatic region $\gamma \ll 1$ has been developed, and the connections to the PT limit $\gamma \gg 1$ and the static limit $\gamma = 0$ indicated.[3,4] The region $\gamma \approx 1$ remains intractable in practice. This is also true for the situation $F \approx F_0$ where the energy levels of the atom undergo huge Stark shifts and Stark broadening, and the atom loses its low-field identity through strong mixing of the various bound and continuum states.

Past experimental studies of the multiphoton ionization of atoms and molecules have all concentrated on laser interactions. For the peak laser powers used the γ's have been in the range 2 up to 10^3.[5] As the laser studies are extended both to higher powers and to shorter pulse lengths, lower values of γ will become involved.

In the present paper we present results on the multiphoton ionization of a fast beam of highly excited hydrogen atoms with principal quantum number n near 66, using radio-frequency and microwave electric fields. To our knowledge this is the largest value of n studied in a laboratory experiment.

The highly excited hydrogen atoms H(high n) were usually produced by H^+-Xe electron-transfer collisions at an incident H^+ energy near 11 keV. Other inert gases have also been used, with the observed H(high n) ionization signals scaling with gas type according to the known cross sections for the production of states within the range $7 \leq n \leq 28$.[6,7] Our measurements are consistent with the Jackson-Schiff n^{-3} scaling rule for the n dependence of the H(high n) production cross section all the way up to $n = 80$.

Our identification of beam atoms in a narrow band of n values is based on the observation by Il'in[8] that the experimentally measured values of the static electric field strength F needed to produce a given ionization probability for each value of n satisfy the expression $F = \text{const} \times n^{-s}$, where the constant depends somewhat upon the geometrical factors for a given experiment and $s \sim 4$. This rule has been verified within the range $9 \leq n \leq 28$ in four different experiments, giving the expressions $F = 8.67 \times 10^8 n^{-4.07}$ V/cm,[6] $F = 1.66 \times 10^9 n^{-4.33}$ V/cm,[9] $F = 4.31 \times 10^8 n^{-3.85}$ V/cm,[10] and $F = 5.96 \times 10^8 n^{-4.00}$ V/cm.[11] This behavior is also supported by numerical calculations for $7 \leq n \leq 25$,[6,12] and is the classical answer to the question, "What state is at the top of the barrier for a given field?"[13] On the basis of extrapolating all the above-mentioned field-ionization data to $n \sim 65$, we feel that our values of n are known to within ± 5 units. In the present experiments a band of n's about five units wide was defined by switching a prequenching static electric field between two different field strengths and observing the resultant difference in field-ionization signals.[9]

The dependence on γ of the H(high n) multiphoton-ionization probability was measured with the apparatus shown schematically in Fig. 1. The fast beam leaving the Xe gas target cell (A) was charge analyzed by a very weak transverse electric field (B) and the pure neutral-atom component then passed through a transverse, static, n-band–defining electric field (C) followed by one of several highly-excited-atom field-ionization regions (D, E, F, G). The protons so produced were detected by the use of a carefully biased and secondary-electron–suppressed Faraday cup (H) on the beam axis. The n-band–defining field was gated at 44 Hz between field values of 28.5 and 41.0 V/cm corresponding to nominal values

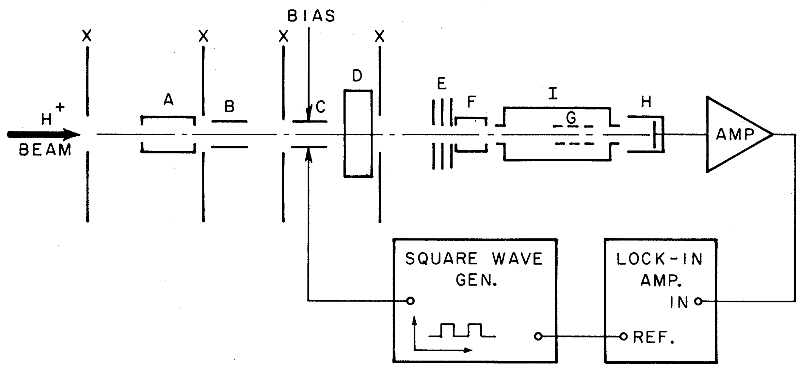

FIG. 1. Apparatus for the multiphoton-ionization experiments. A, electron-transfer gas cell; B, ion-removal plates; C, n-band–defining static field; D, broad-band rf structure; E,G, Einzel-lens axial static field structures; F, X-band microwave cavity; H, Faraday cup; I, liquid-helium cryopump; X, differential pumping apertures.

of n of 69 and 63, respectively. The resultant ac Faraday-cup current was amplified by a field-effect–transistor preamplifier and detected with a phase-sensitive lock-in amplifier.

We have used three different multiphoton electric field structures. The first (D) was a broad-band parallel-plate rf structure which permitted the study of both transverse static fields and transverse rf fields with frequencies in the region 0.9–1.8 GHz. The second structure (F) was an X-band axial microwave field from one of several available TM_{01n} modes of a high-Q cylindrical cavity with its axis along the beam direction. The third structure was an electrostatic Einzel lens (G) which, because of the beam velocities used, applied a one-cycle wave to the highly excited atoms with a principal Fourier frequency component of about 30 MHz. For these experiments, theory predicts the behavior at this last frequency and our field strengths (i.e., small γ) to be similar to that for a completely static field.[3]

The results for several of the direct-ionization experiments are shown in Fig. 2, where the field-ionization signal S for the n band $63 \leqslant n \leqslant 69$ is plotted versus γ for the various multiphoton-transition structures, using the central value of n of 66 and peak values F of the applied electric field strength. The data when plotted as a function of peak electric field qualitatively confirm one prediction of the adiabatic theory,[3] namely, that for γ's near the adiabatic limit ($\gamma \ll 1$) and for the same peak field strength F, higher-frequency waves are more effective for ionization than lower-frequency waves. We emphasize, however,

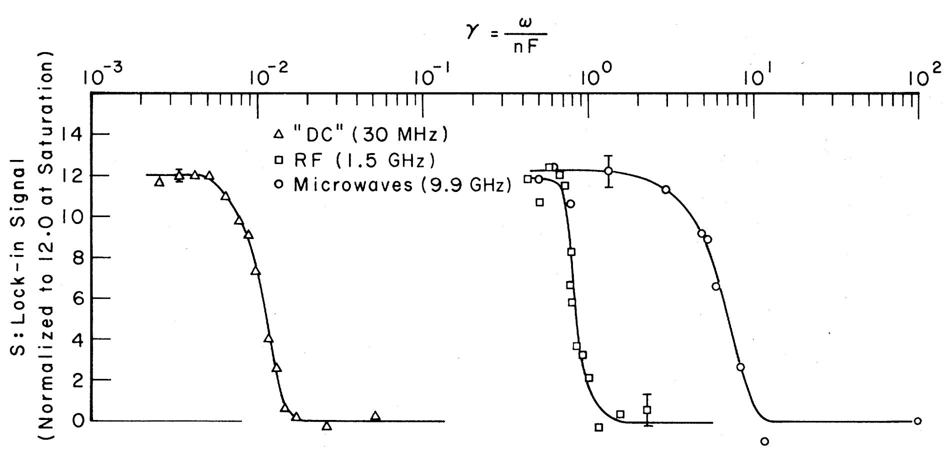

FIG. 2. The ionization of hydrogen atoms with principal quantum numbers in the band $63 \leqslant n \leqslant 69$, as a function of the tunneling parameter γ. Shown are curves for (1) an essentially static axial applied electric field, (2) a transverse 1.50-GHz rf field, and (3) an axial 9.9-GHz microwave field. Smooth curves have been drawn through the data points to aid the eye. Each curve exhibits experimental saturation at lower values of γ (higher values of field strength F).

that most of the 1.5-GHz data points of Fig. 2 are in the region $\gamma \approx 1$ where no theoretical predictions are available.

In the absence of saturated resonant intermediate states[1,14] or an ionization potential lowered by the intense field,[3,15] the number k (multiplicity) of photons required for ionization would be equal to $[I/\hbar\omega + 1]$, where $[x]$ refers to the integer value of x. For the 1.5-GHz ($\gamma \sim 1$) data and the 9.9-GHz ($2 \lesssim \gamma \lesssim 9$) data k is 502 and 76, respectively. The actual or effective multiplicity is defined by the slope of a log-log plot of S versus F^2. These values are about 2.0 and 1.5, respectively. We are not aware of any multiplicity-reduction theory directly applicable to our data, but in some limits the theory of Ref. 3 predicts that k is decreased by a number of order n, a large effect for $n = 66$ and $k = 76$.

As these experiments studied extremely delicate atoms, many different auxiliary tests of the apparatus were made. Among the principal results was a determination that the dominant effect (> 90%) of the applied field was indeed ionization rather than bound-bound transitions either to states of lower principal quantum number n or to states of somewhat higher n. In addition, another type of experiment was performed to check directly that the ions produced were caused by the applied electric fields and not by spurious effects such as collisions with the background gas. In the experiment the ionization of an atom in a static axial electric field region "labeled" with a positive electric potential resulted in an acceleration of the ion as it left the region. The corresponding increase in velocity labeled the kinetic energy of only those protons produced in this spatial region, which were then isolated for detection using kinetic-energy analysis. We varied the pressure of the background gas in such a region located inside a liquid-helium cryopump from our normal 10^{-8}-Torr operating range to a measured pressure of less than 10^{-11} Torr with no observable (< 4%) change in results.

Let us, in conclusion, indicate some further possibilities for the study of multiphoton processes using highly excited atomic beams. An important improvement for some atomic species will be a narrowed high-n band produced by laser pumping specific, more-populated, lower-energy states present in the beam. In addition, microwave powers orders of magnitude larger than presently used would permit studies at lower values of n and γ. It also appears possible to use lasers to extend our field-ionization measurements to optical frequencies. Different systems (alkali atoms, for example) could be used with the goal being the study of possible inner-shell core effects on the multiphoton processes. Finally, one should be able to do experiments oriented towards searching for the transparency of atoms to superstrength ($F \geq F_0$) electromagnetic fields.[16,17]

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